Ions, Atoms & Molecules in Intense Fields:of FELs:

from the visible . . to x-ray FELs

1. huge integrated flux → dilute samples

2. huge peak intensities → multi photon

3. extreme short times → electron dynamics

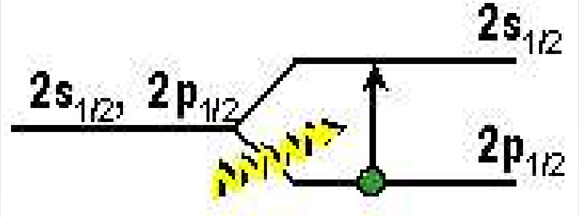
Joachim Ullrich

Max-Planck-Institut Heidelberg

at least three

New Fields of Research

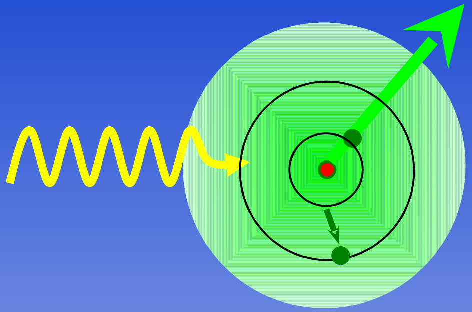
I. Precision Spectroscopy & Ionization : Ions



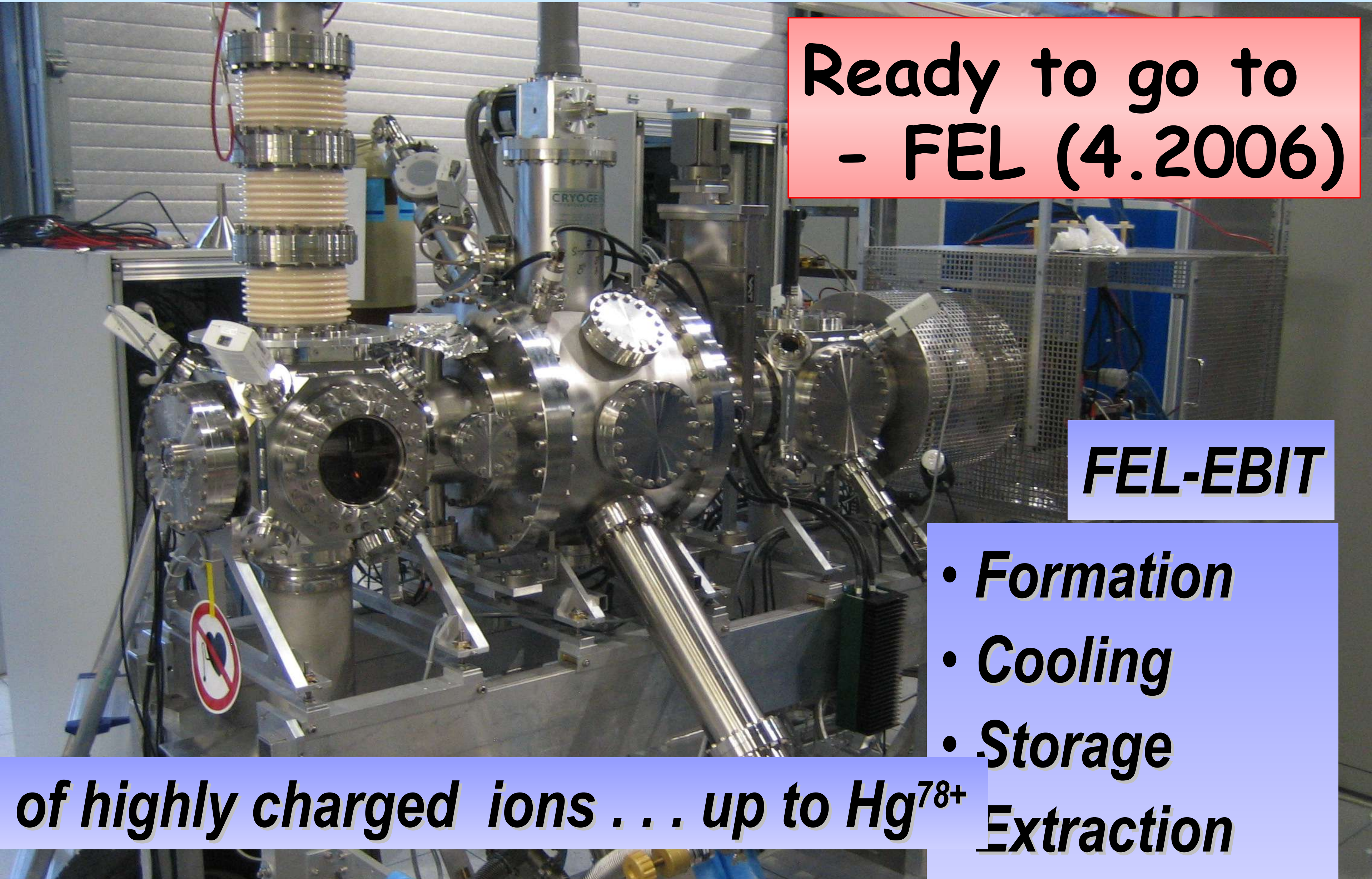

Ar^{15+}: 31.866(1) eV

⋮

U^{89+}: 280.83(10) eV



Highly charged ions



Ready to go to
- FEL (4.2006)

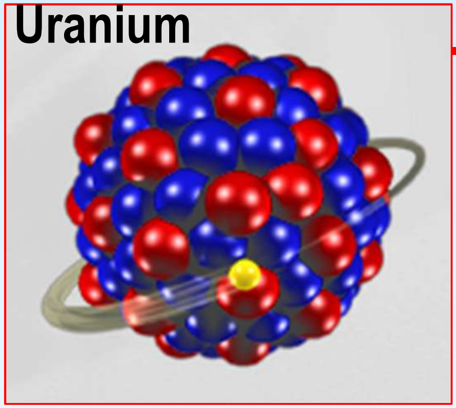
FEL-EBIT

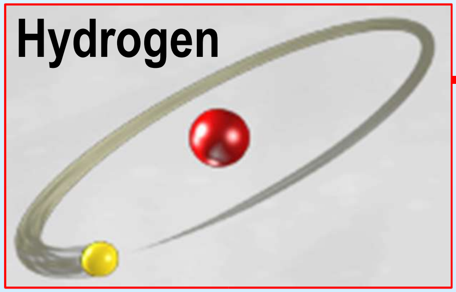
- Formation
- Cooling
- Storage
- Extraction

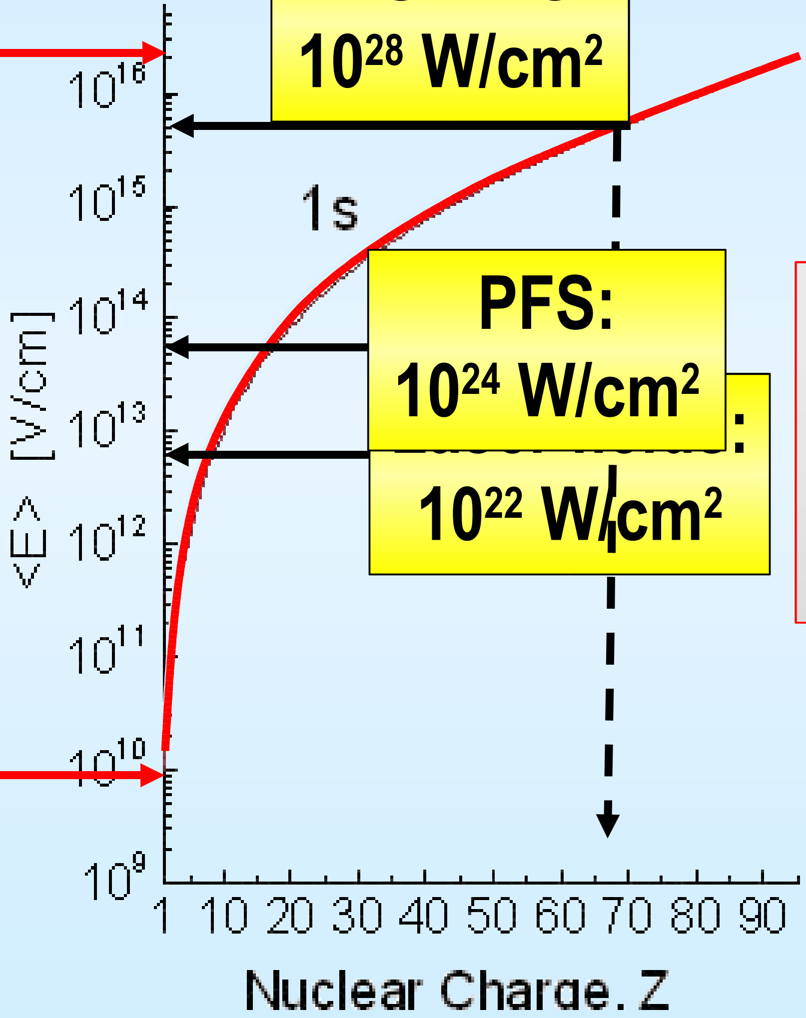
of highly charged ions . . . up to Hg^{78+}

Spectroscopy of Ions

Max–Planck–Institut für Kernphysik







PFS-HHG:
10^{28} W/cm^2

PFS:
10^{24} W/cm^2

10^{22} W/cm^2

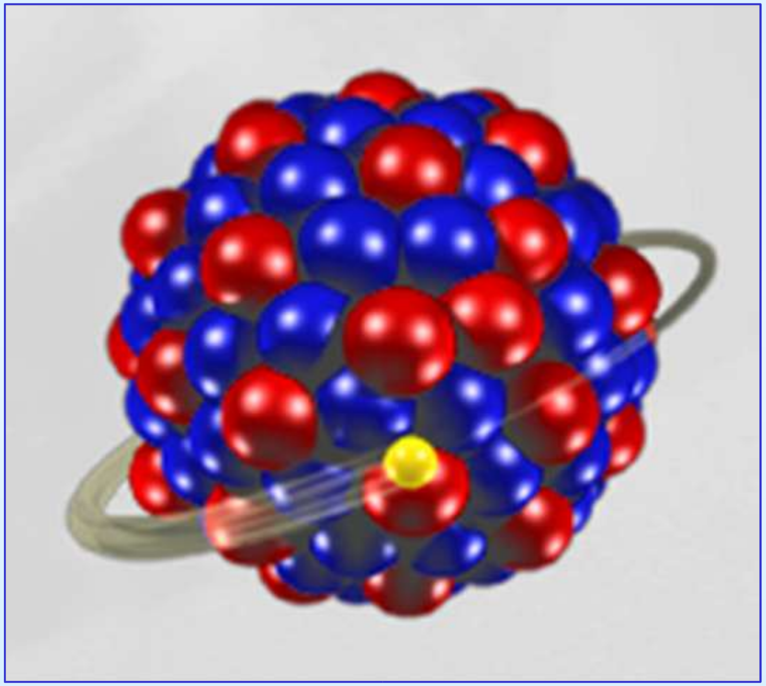
$\Delta E \approx 500$ eV
$Z \cdot \alpha \approx 1$

Quantum
Electro-
Dynamics

$\Delta E \approx 10^{-6}$ eV
$Z \cdot \alpha \approx 10^{-2}$

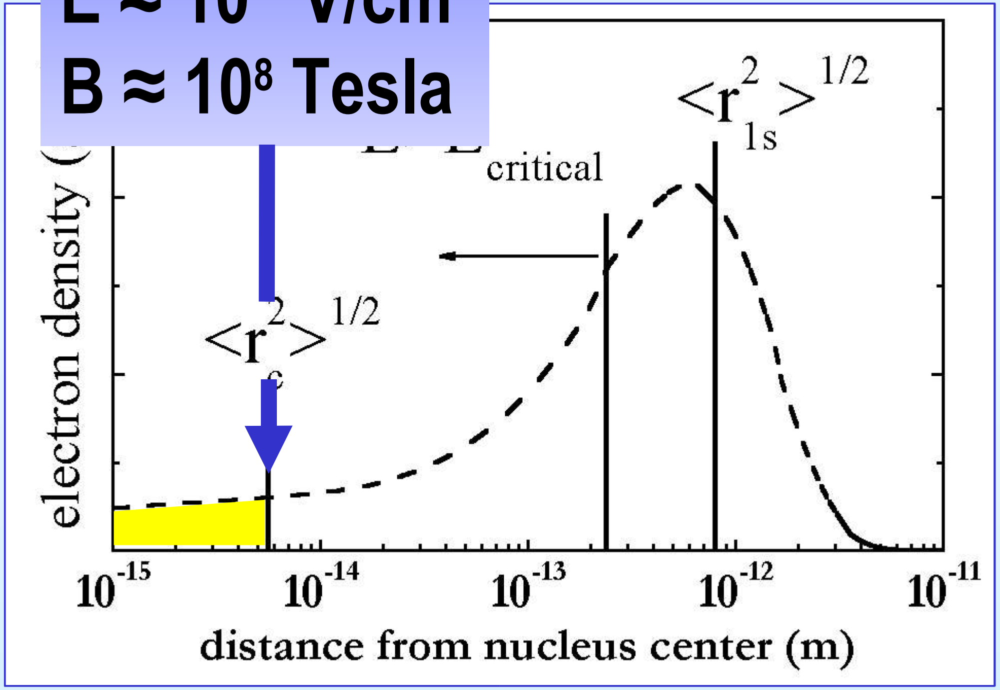
Spectroscopy of Ions

Explore the Nucleus



$E \approx 10^{19}$ V/cm

$B \approx 10^{8}$ Tesla



?

- Fundamental Symmetries

Extracted Beams

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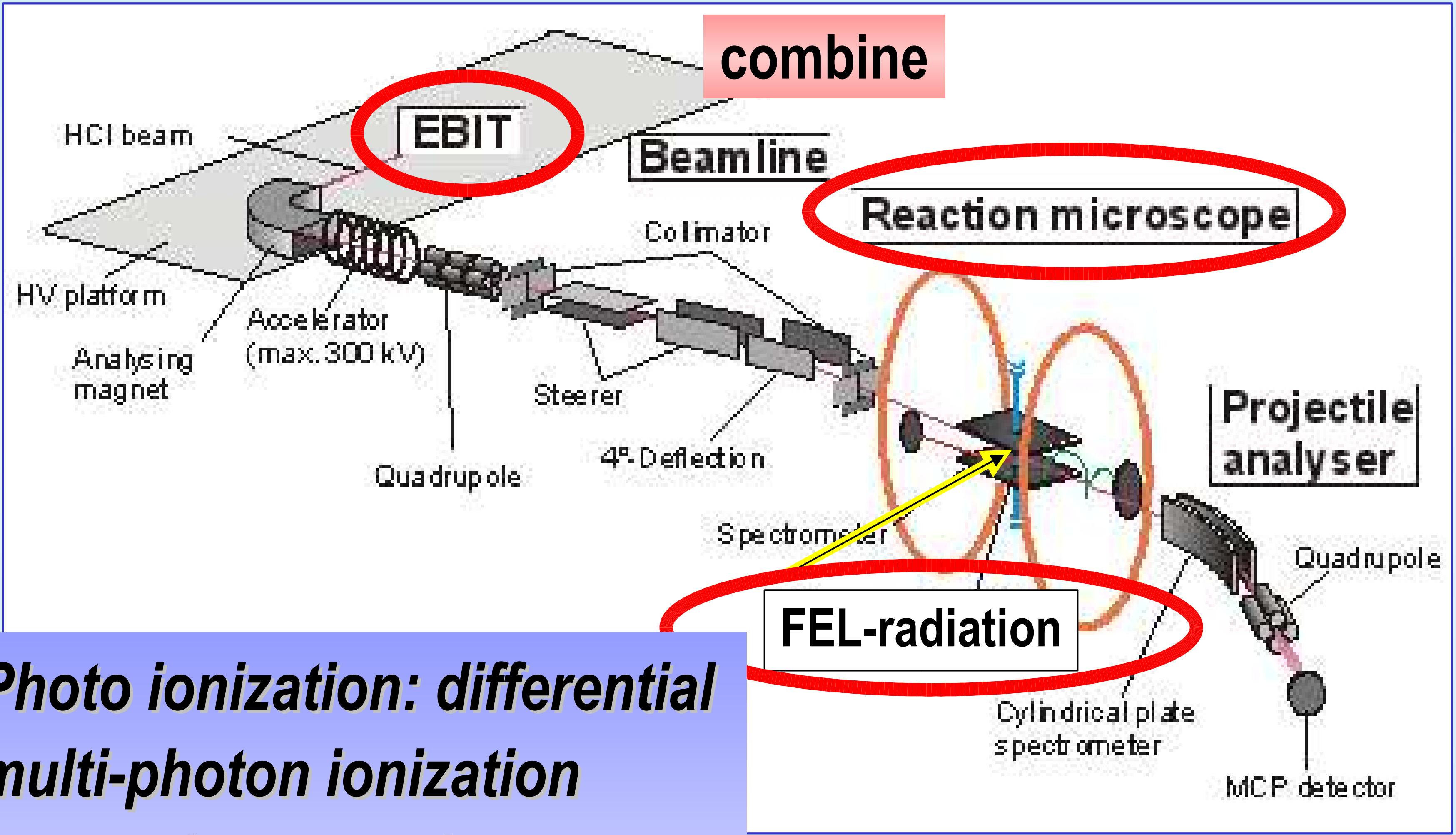
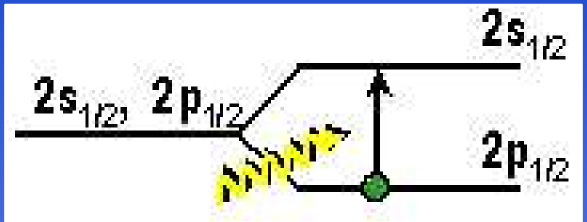


- ***Photo ionization: differential***
- ***multi-photon ionization***
- ***harmonic generation . . .***

I. Precision Spectroscopy & Ionization : Ions



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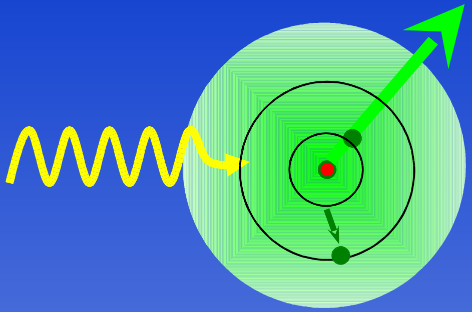
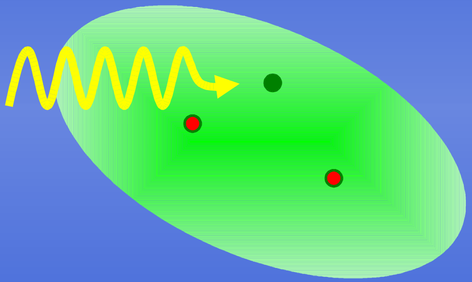
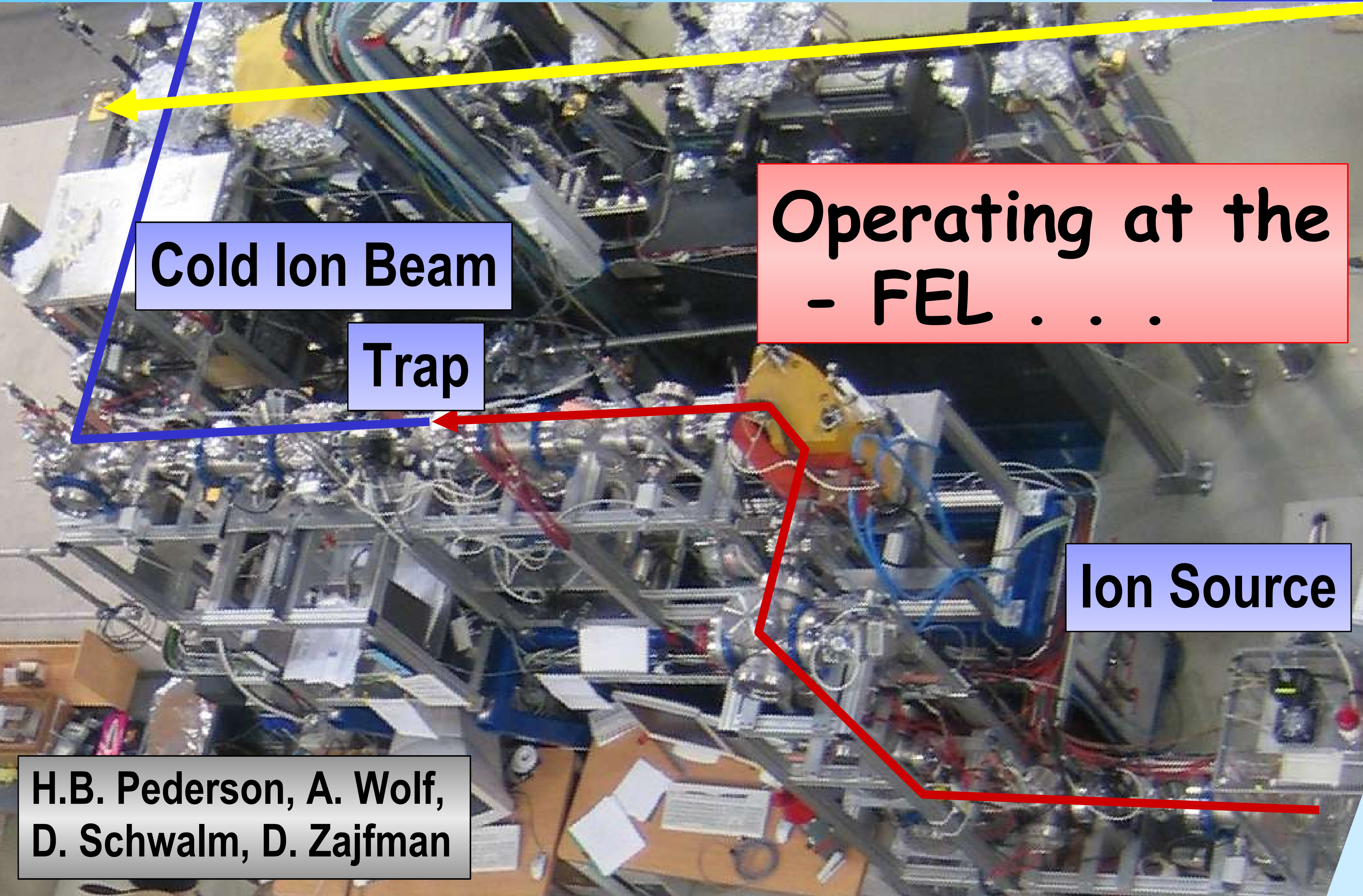


I. Interaction with Molecular Ions

…. very dilute beams



Cold molecular ions



Cold Ion Beam

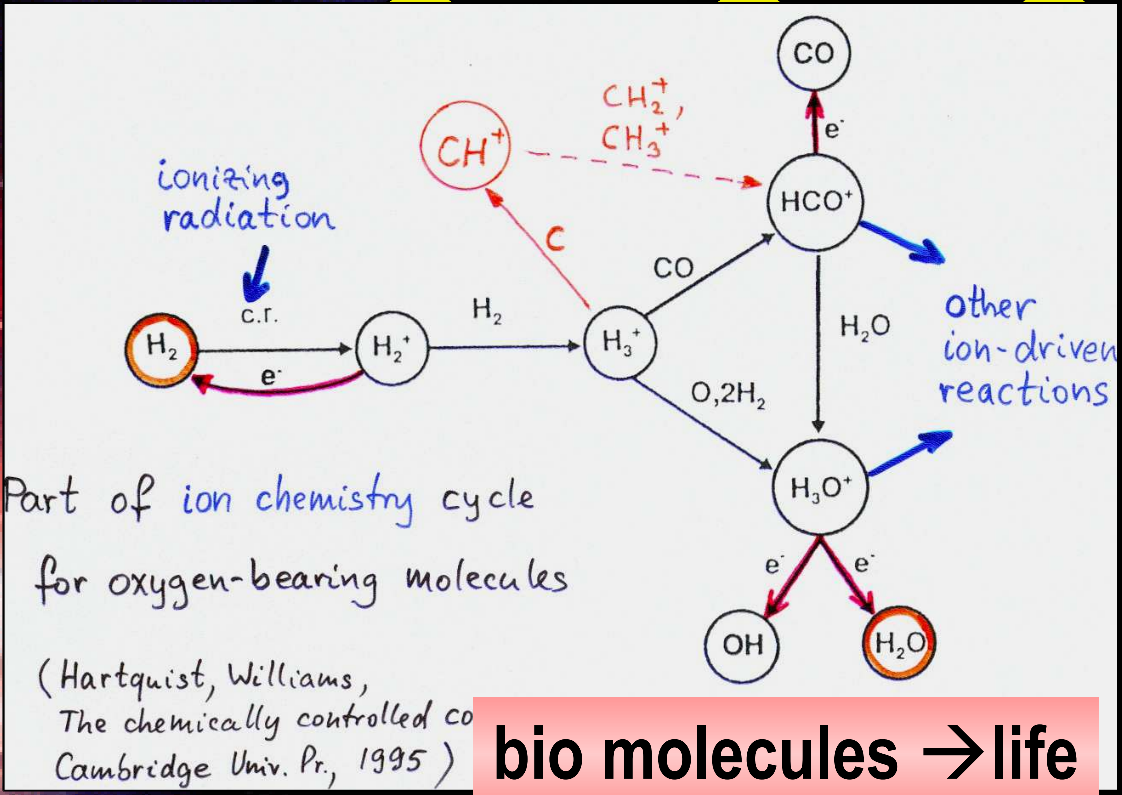
Trap

Operating at the - FEL . . .

Ion Source

H.B. Pederson, A. Wolf, D. Schwalm, D. Zajfman

Cold molecular ions



Part of ion chemistry cycle for oxygen-bearing molecules

(Hartquist, Williams, The chemically controlled co Cambridge Univ. Pr., 1995)

bio molecules → life

How do molecules form in interstellar space?

Cold molecular ions

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Photoreaction	C	α	β	Reference
	C	α	β	Reference

Relevant

- Interste
- Close to

$H_3O^+ + \nu \rightarrow OH$

$O_2^+ + \nu \rightarrow O + O$

$CH^+ + \nu \rightarrow H^+ +$

$CH_2^+ + \nu \rightarrow CH^+$

$CH_3^+ + \nu \rightarrow CH_2^+$

$CH_3^+ + \nu \rightarrow CH^+$

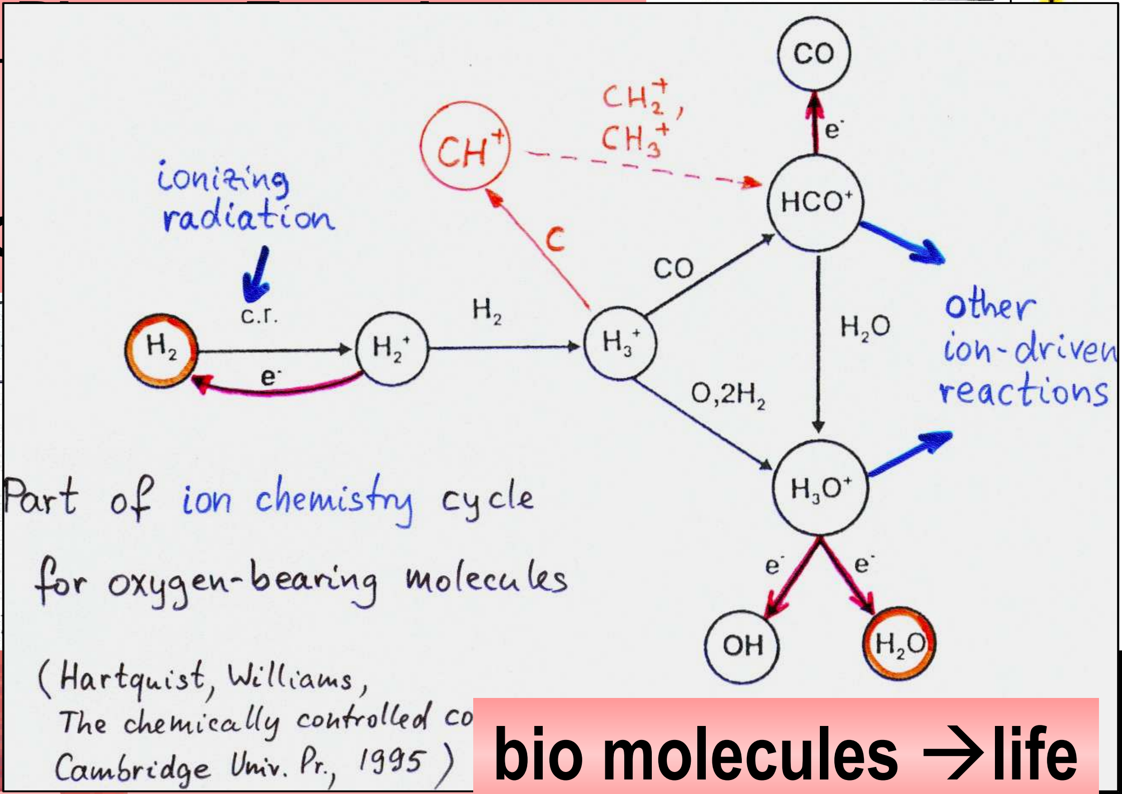
$CH_4^+ + \nu \rightarrow CH_2^+$

$CH_4^+ + \nu \rightarrow CH_3^+$

$CH_5^+ + \nu \rightarrow CH_4^+$

$CH_5^+ + \nu \rightarrow CH_3^+$

$SH^+ + \nu \rightarrow S^+ +$



Part of ion chemistry cycle for oxygen-bearing molecules

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bio molecules → life

At the FEL:

feasible for the first time

10^{17} photons/sec

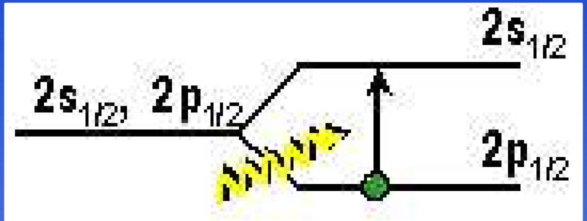
Sternberg & Dalgarno, Chemistry in dense photo dominated regions

et density

m^{-3})

How do molecules form in interstellar space ?

I. Precision Spectroscopy & Ionization : Ions

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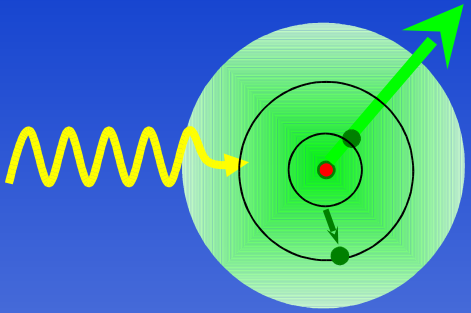

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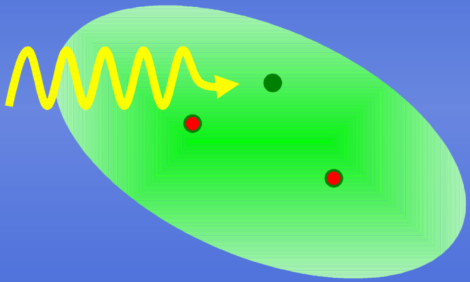
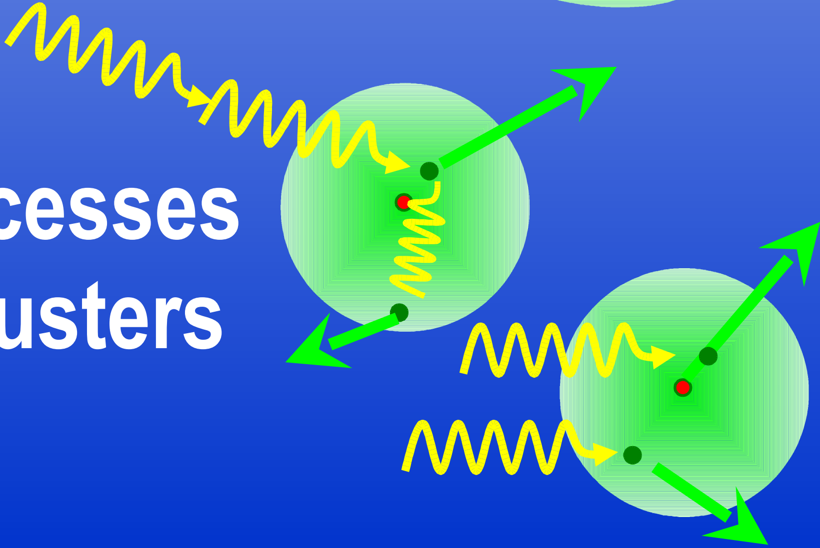


III. Few- to Multi-Photon Processes in Atoms, Molecules & Clusters

Ultra-Fast Phenomena



Reaction Microscopes

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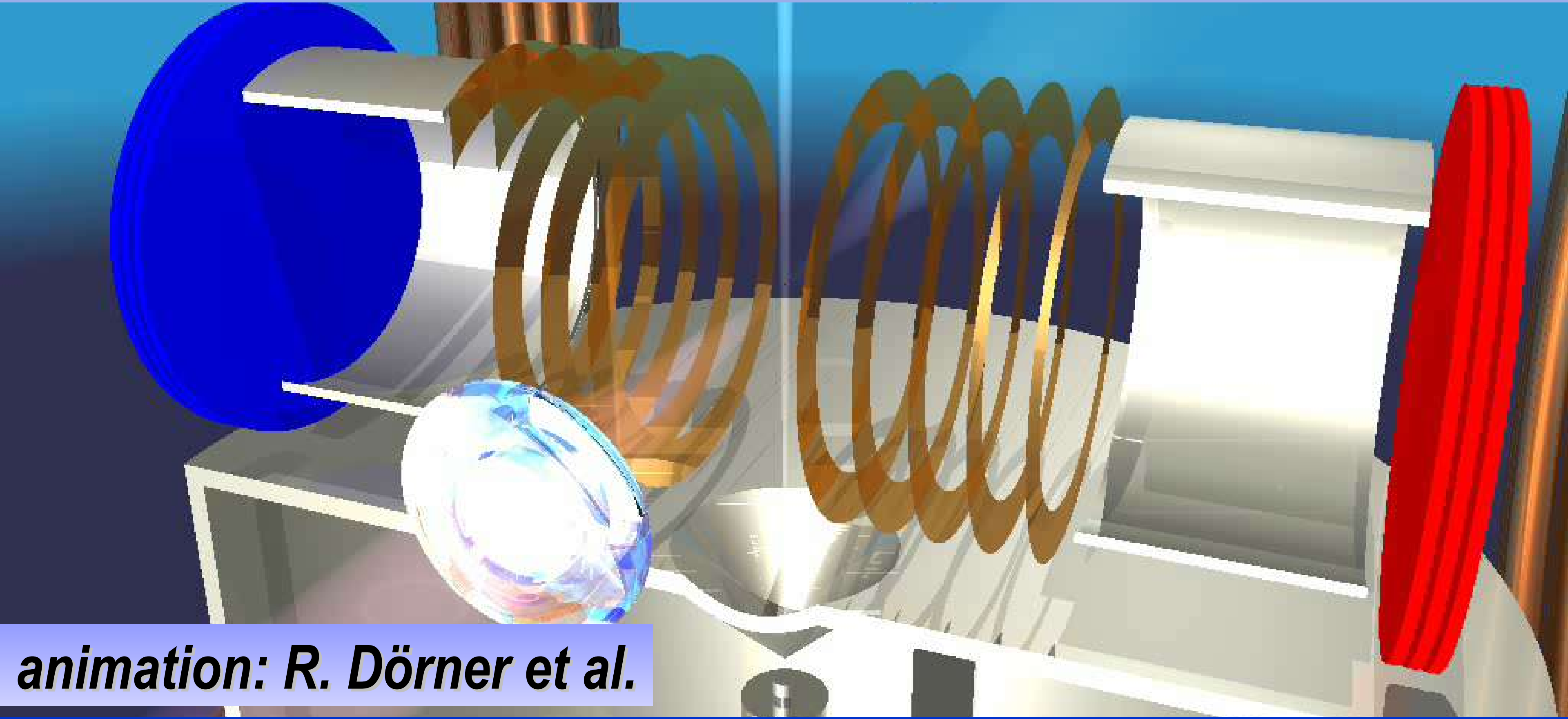
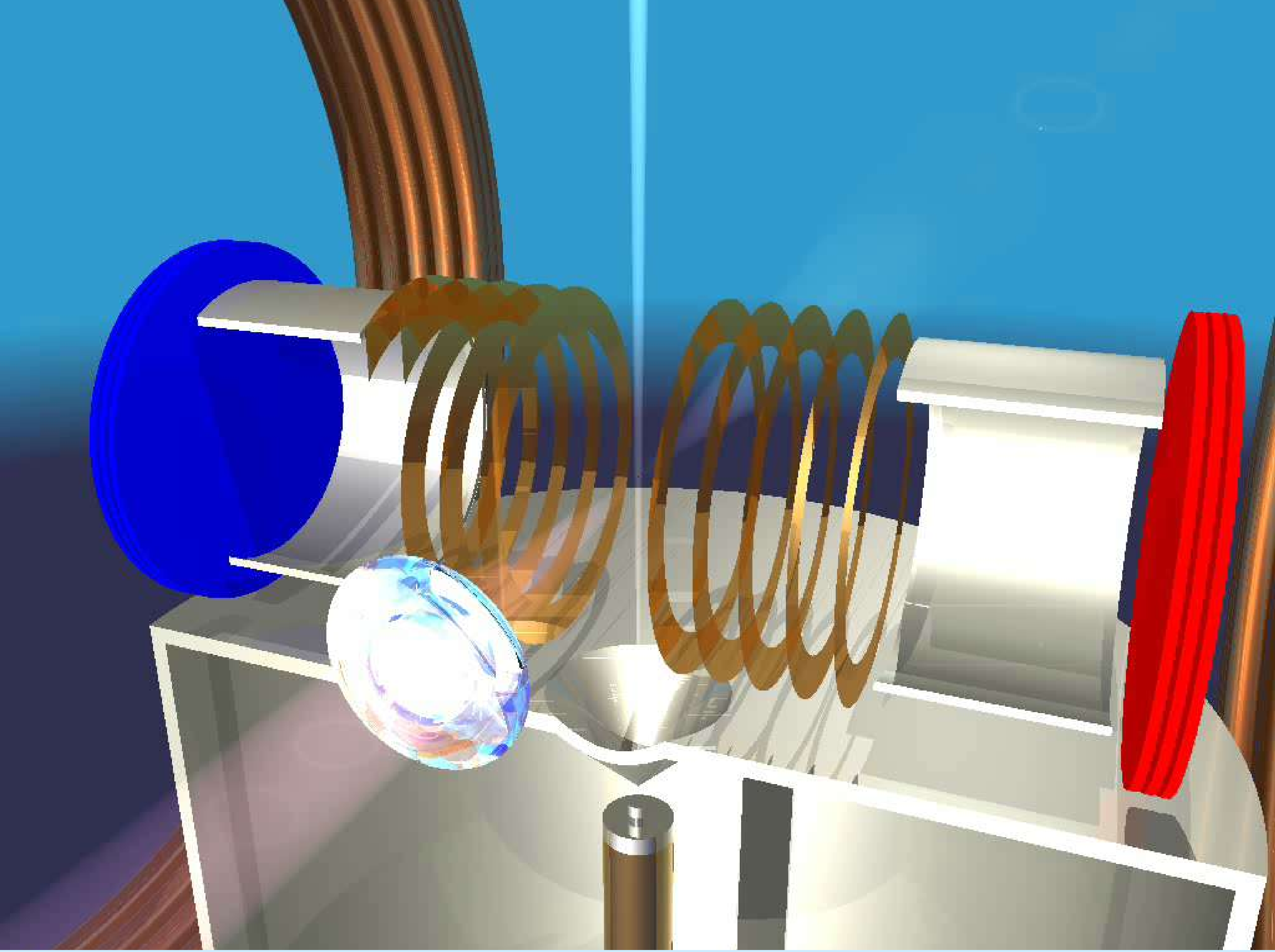
The “Cloud Chambers”



animation: R. Dörner et al.

of Atomic & Molecular Physics



FEL Reaction Microscope

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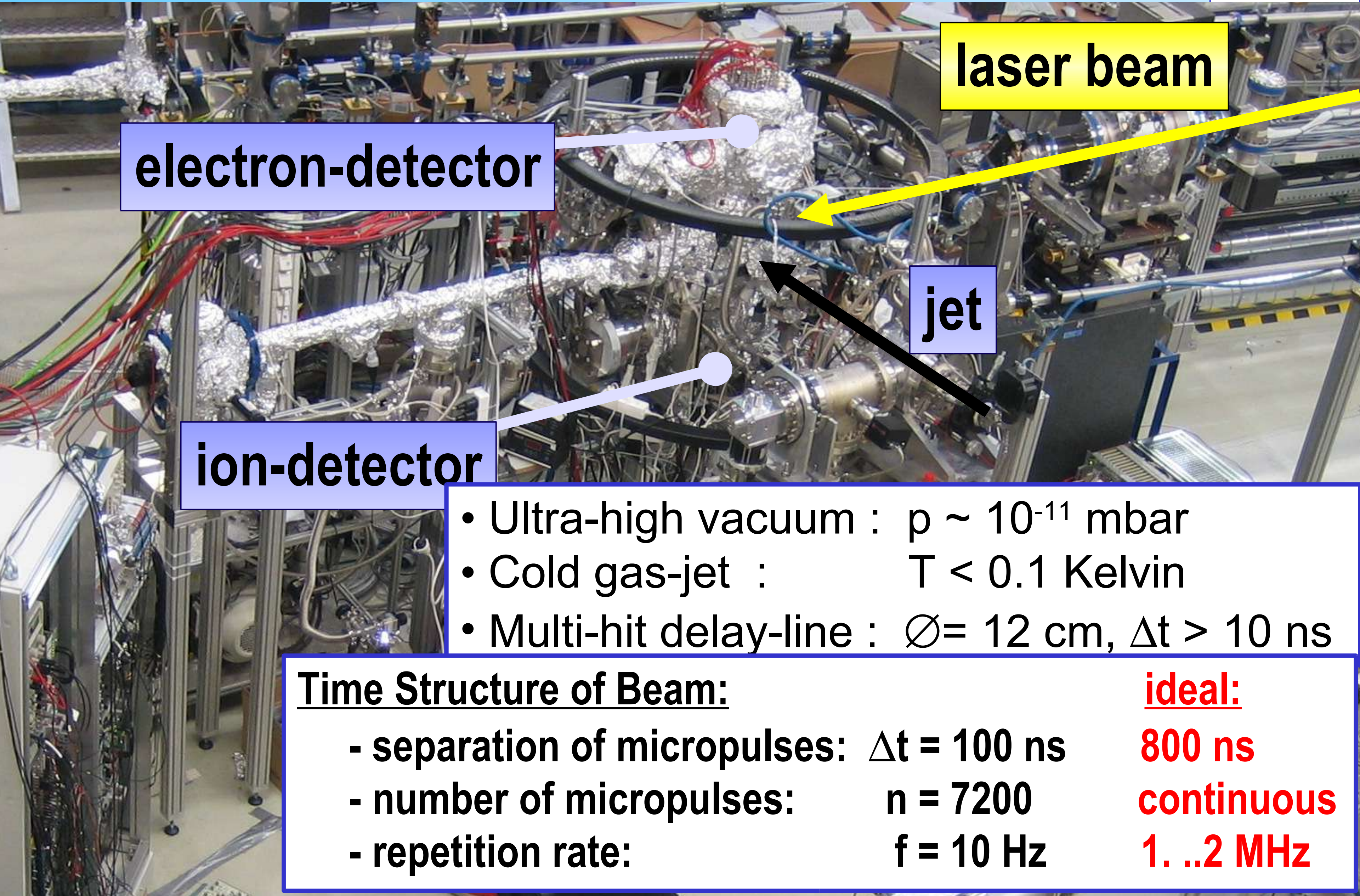

- Ultra-high vacuum : $p \sim 10^{-11}$ mbar
- Cold gas-jet : $T < 0.1$ Kelvin
- Multi-hit delay-line : $\varnothing = 12$ cm, $\Delta t > 10$ ns

Time Structure of Beam:		**ideal:**
- separation of micropulses:	**$\Delta t = 100$ ns**	**800 ns**
- number of micropulses:	**n = 7200**	**continuous**
- repetition rate:	**f = 10 Hz**	**1. ..2 MHz**

The Heidelberg Group

U

eber

F

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Outline of the Talk

Max–Planck–Institut für Kernphysik

- Reaction Microscopes
- Introduction
  - Atoms in Intense Fields
  - Attosecond Streak Effect
- From Single-Photon to Multi-Photon
  - Double Ionization
  - Multiple Ionization
- Ultra-Fast Phenomena
  - Attosecond Correlation
  - Pump-Probe: The Molecular Movie

Atoms in Intense Fields

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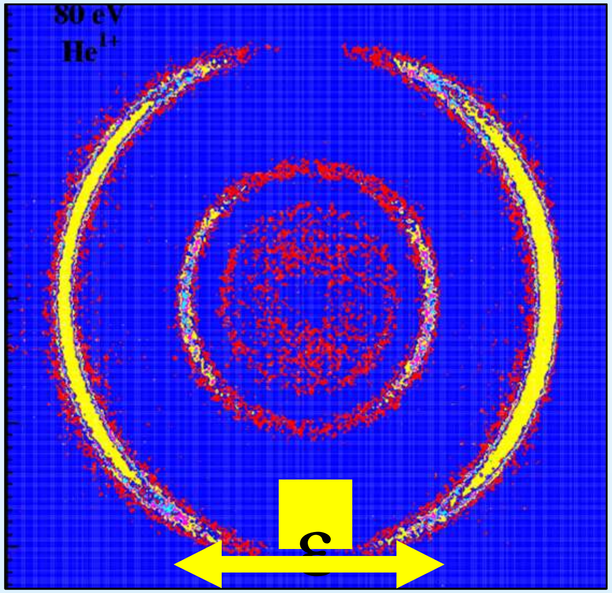
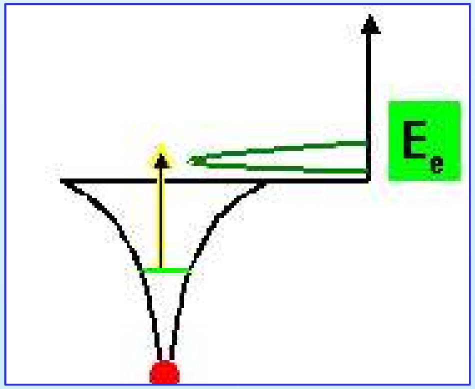
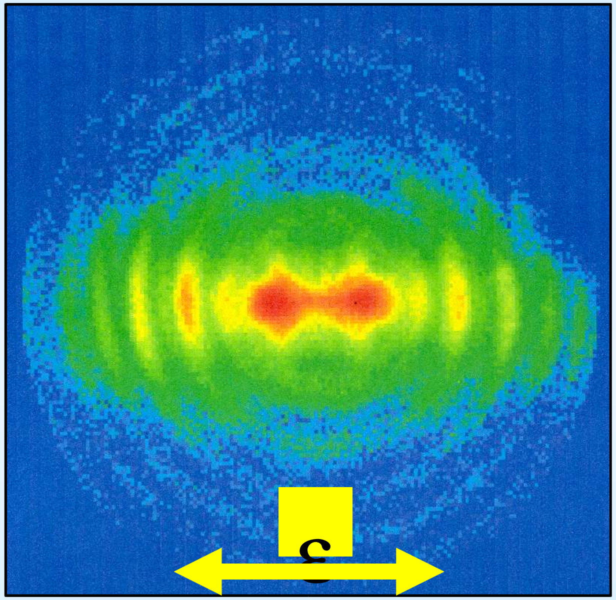
für Kernphysik

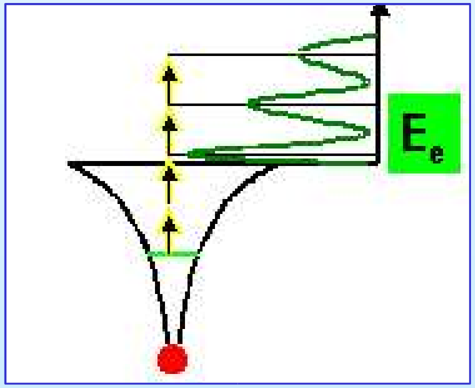
photo absorption
Einstein 1905





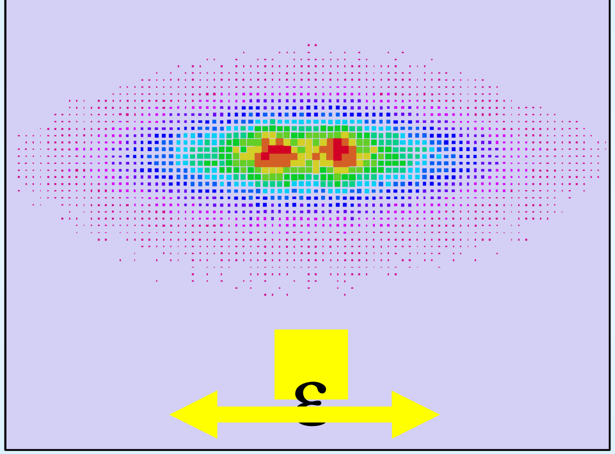
multi-photon ionization
Göppert-Meier 1931

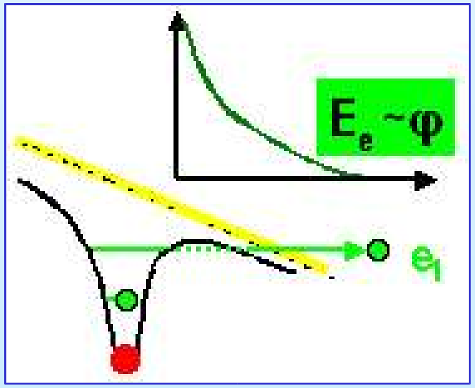




tunnelling ionization

Mechanism?





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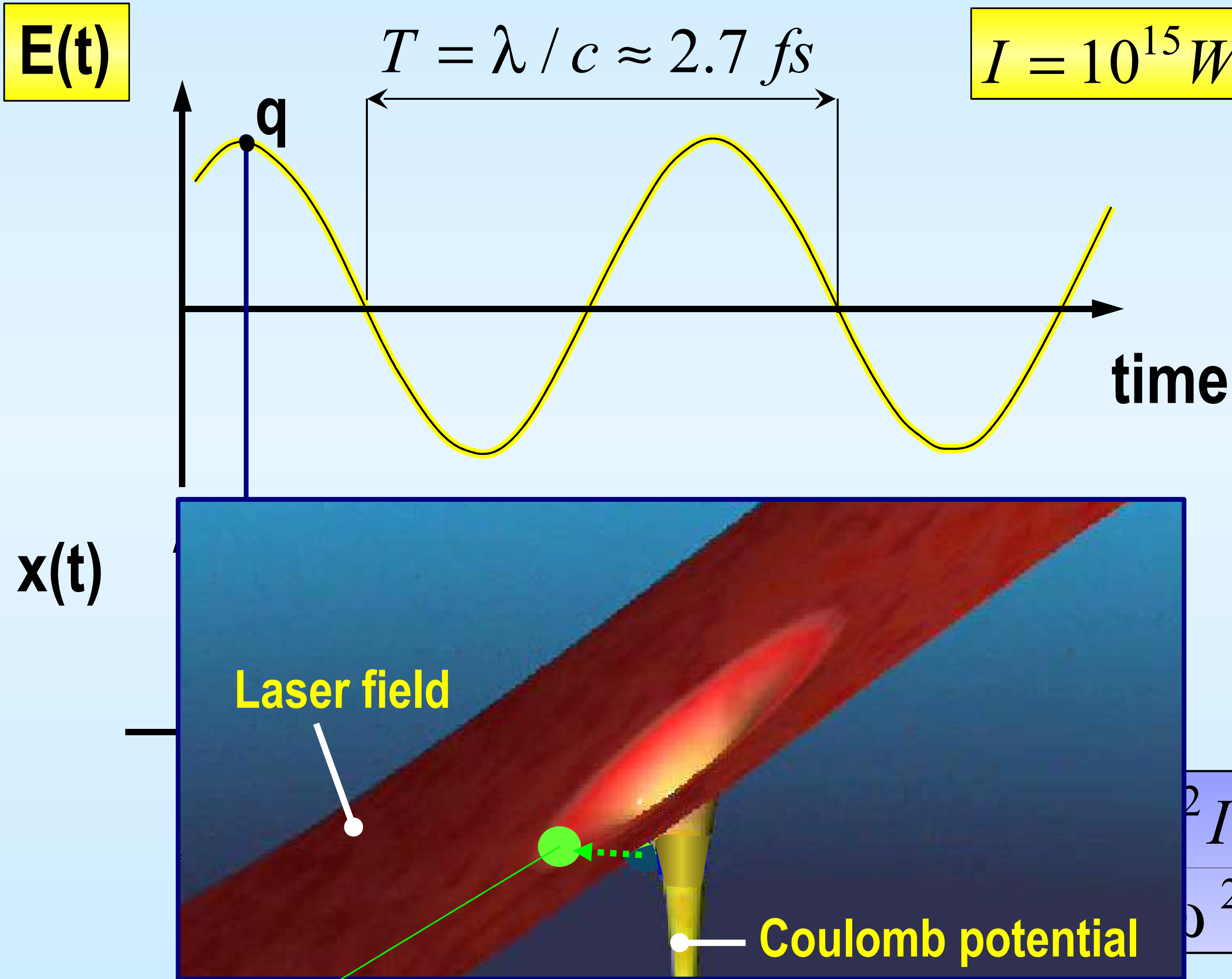
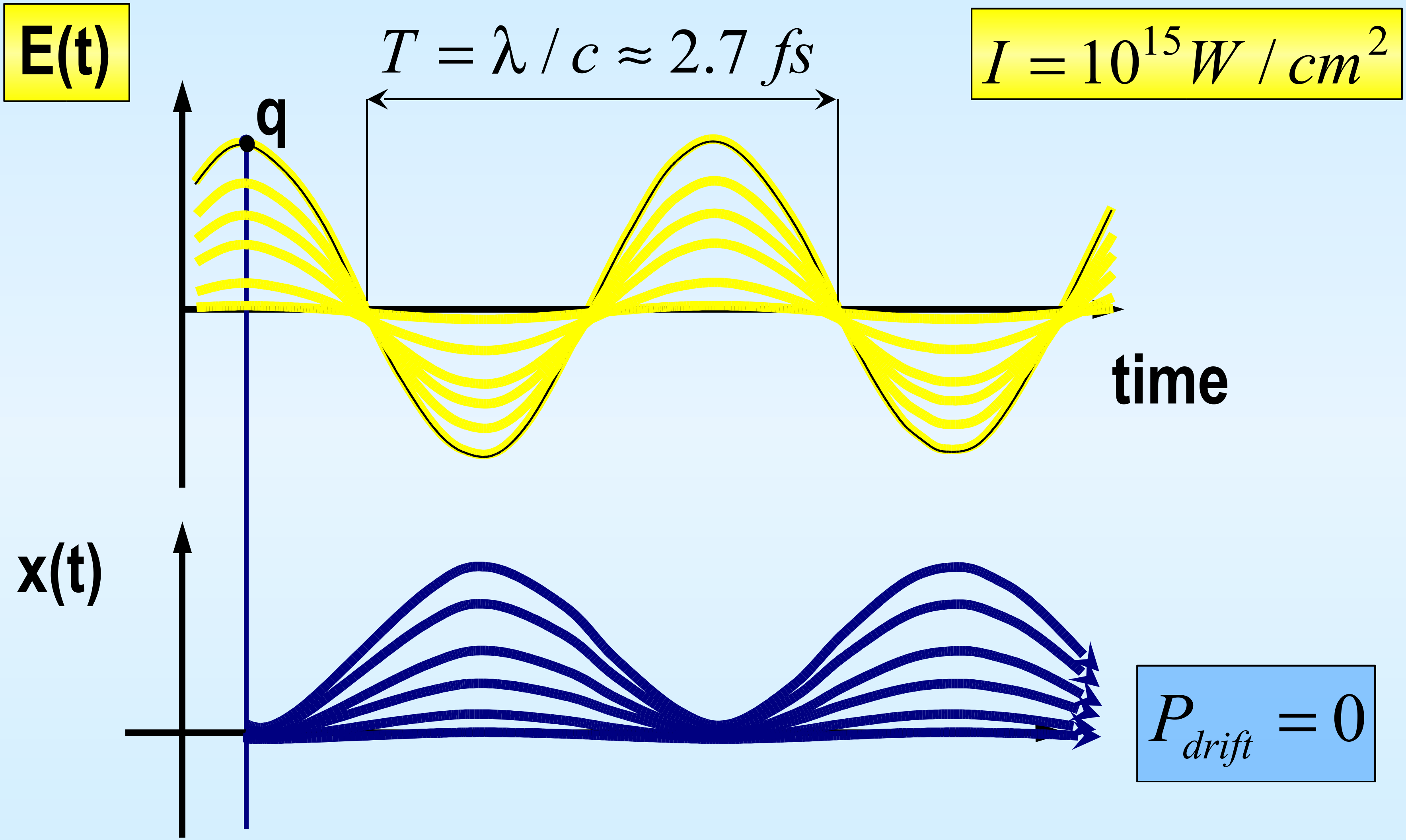
Tunneling Ionization



$$I = 10^{15} W / cm^2$$

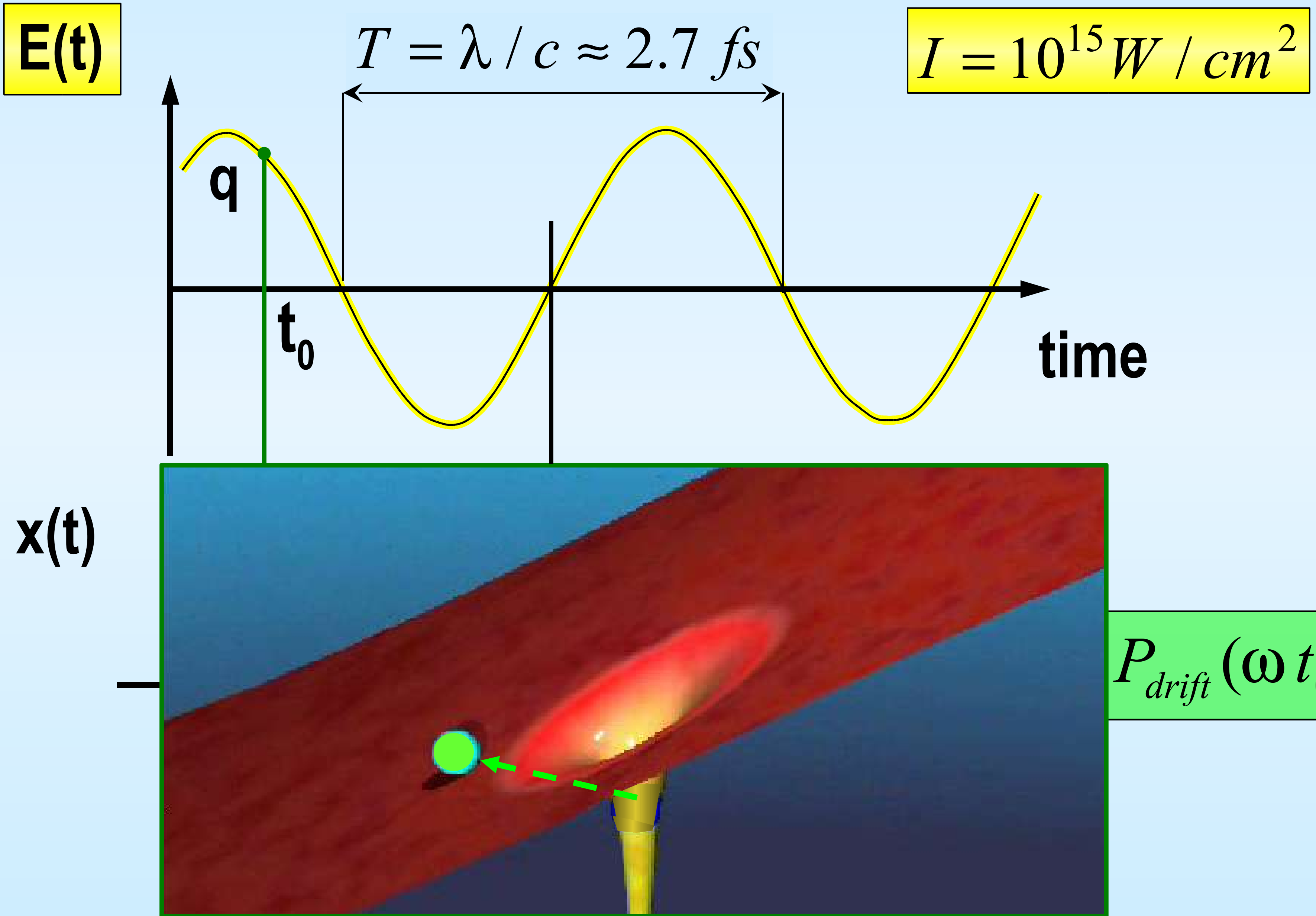
$$\frac{{}^{2} I}{\,{}^{2}} \approx 50\,eV$$

Tunneling Ionization

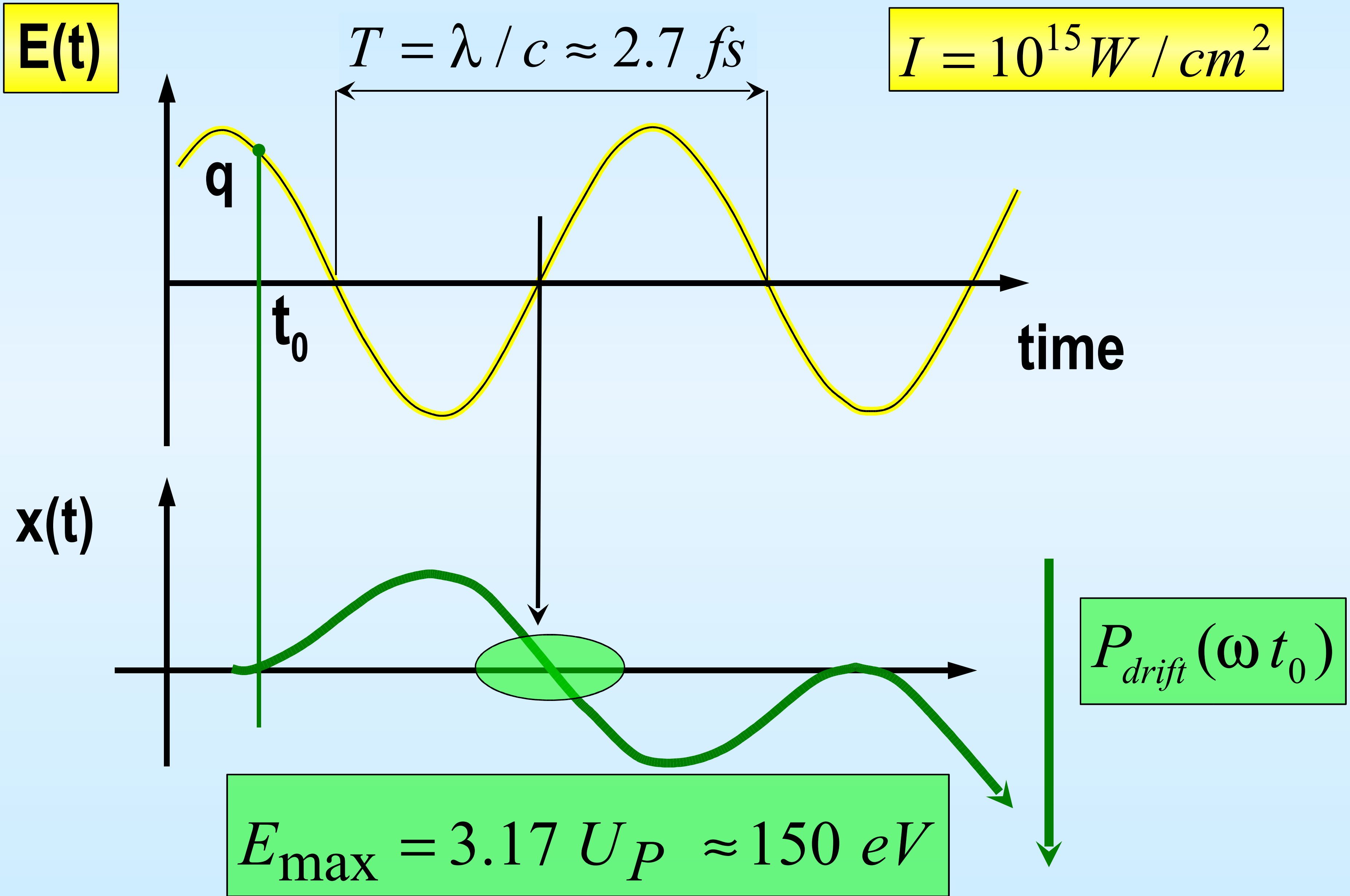


A Photoelectron/-ion with zero energy!

Tunneling Ionization



Tunneling Ionization

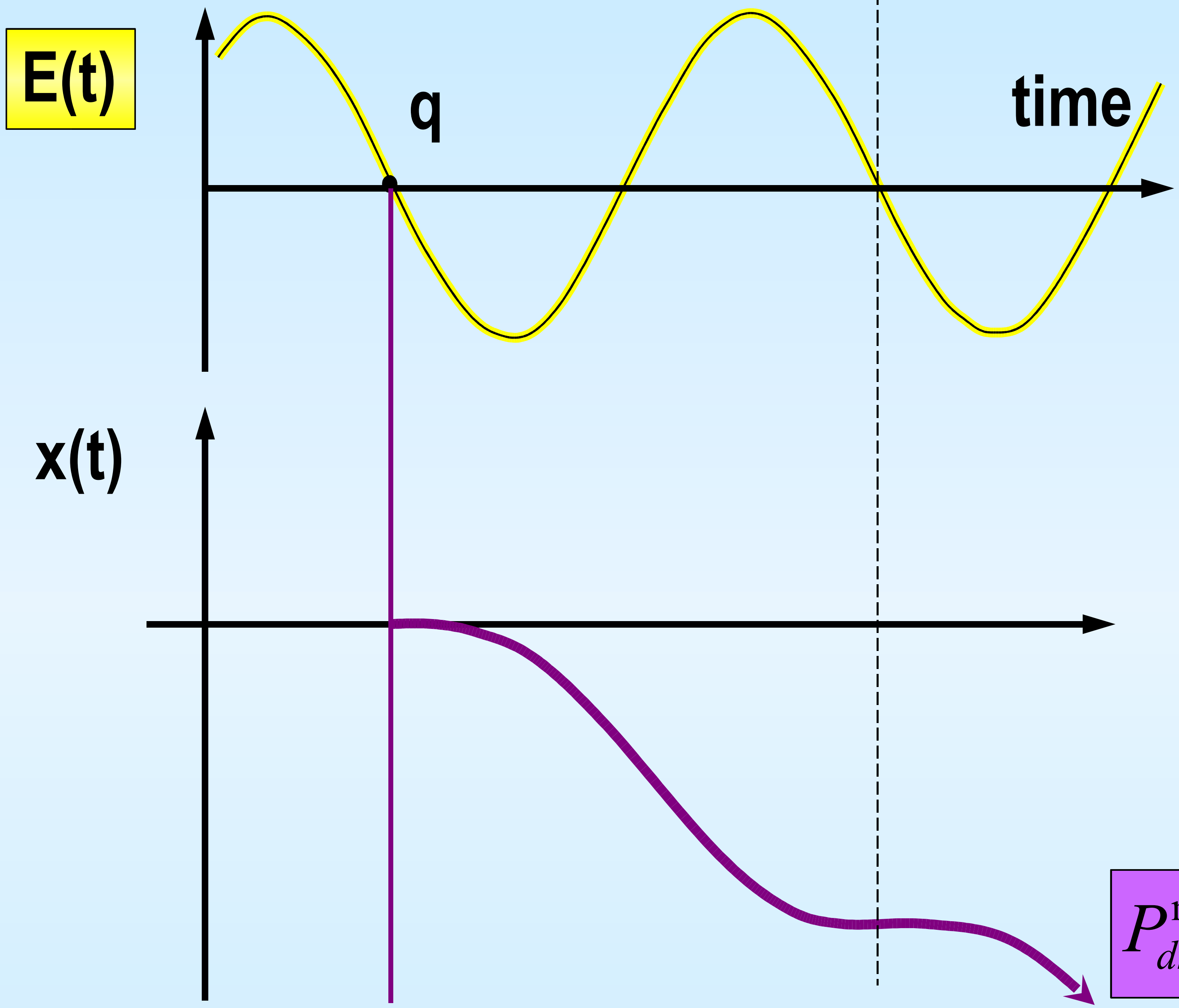


$T = \lambda / c \approx 2.7\ fs$

$I = 10^{15} W / cm^2$

$P_{drift}(\omega t_0)$

$E_{\max} = 3.17\ U_P\ \approx 150\ eV$



$$P_{drift}^{\max} = 2\sqrt{U_p}$$

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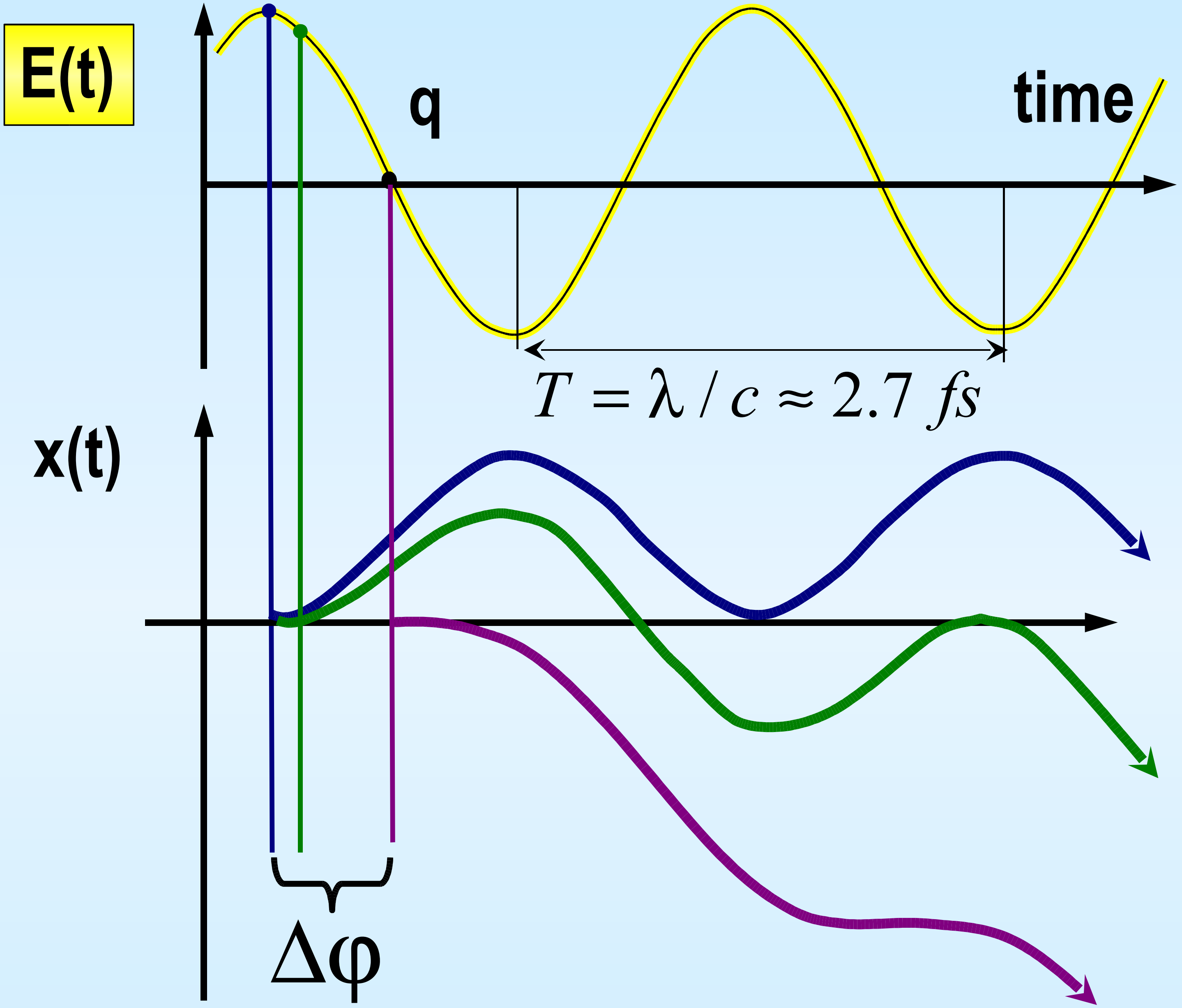
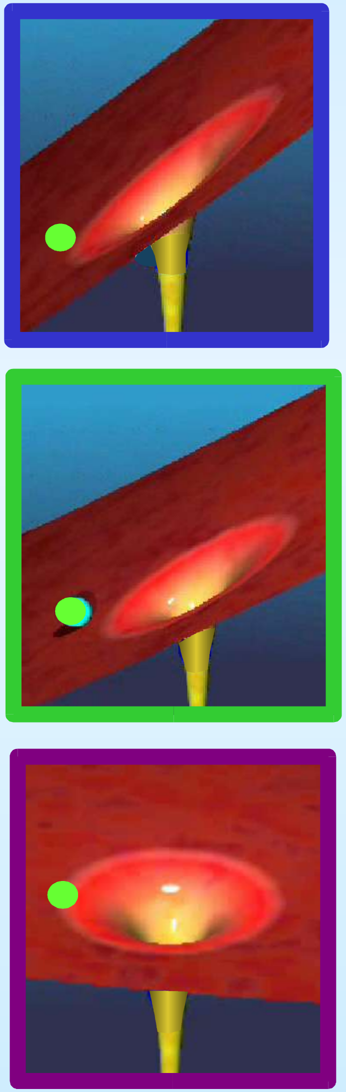
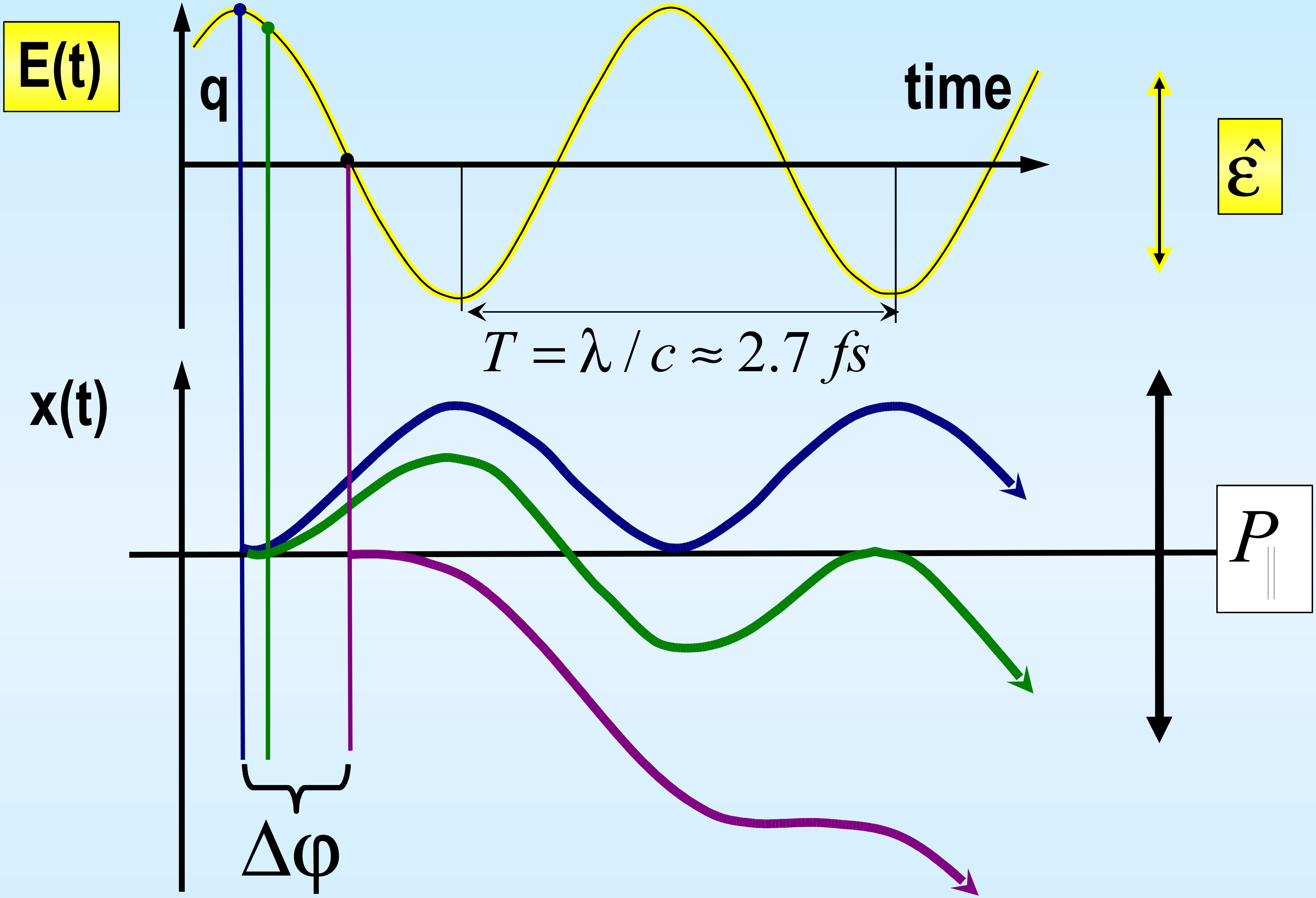
für Kernphysik





$$\Delta t = 80\, as \ \ for \ \Delta p_{rec} = 0.1\ a.u.$$



$$\Delta t = 80\, as \ \ for\ \Delta p_{rec} = 0.1\ a.u.$$

Attosecond Streak Camera

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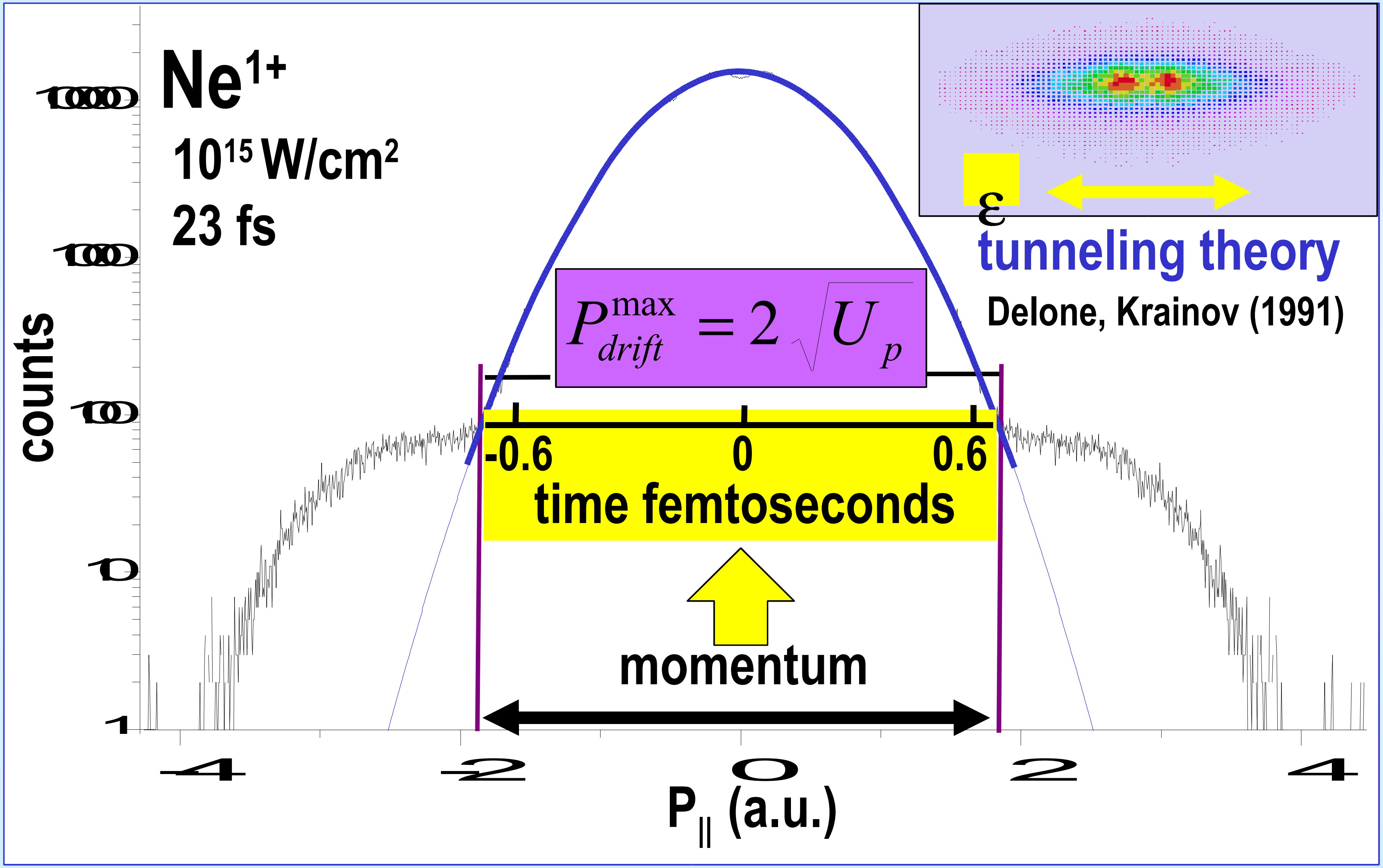


Ne^{1+}

10^{15} W/cm^2

23 fs

$$P_{drift}^{max} = 2\sqrt{U_p}$$

time femtoseconds

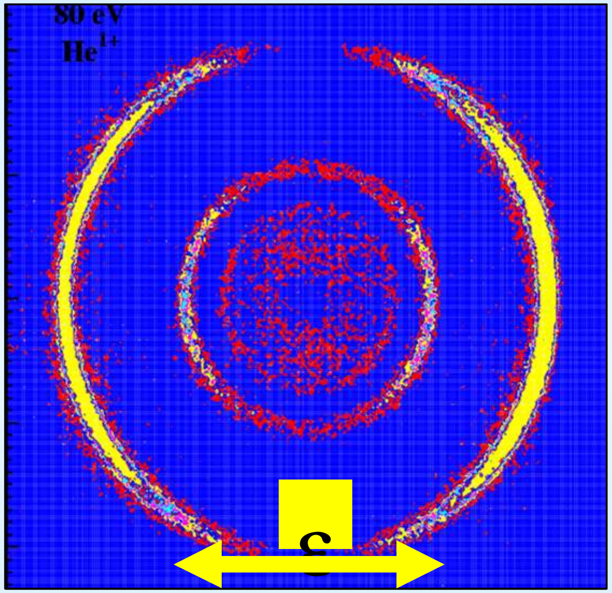
momentum

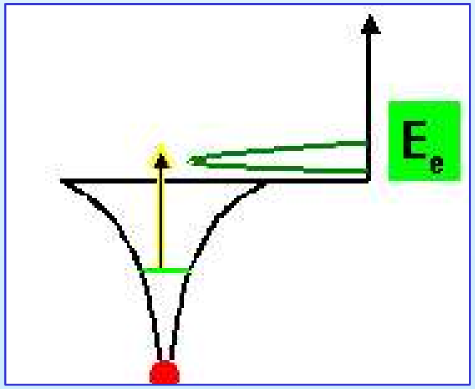
tunneling theory

Delone, Krainov (1991)

Atoms in Intense Fields

photo absorption
Einstein 1905





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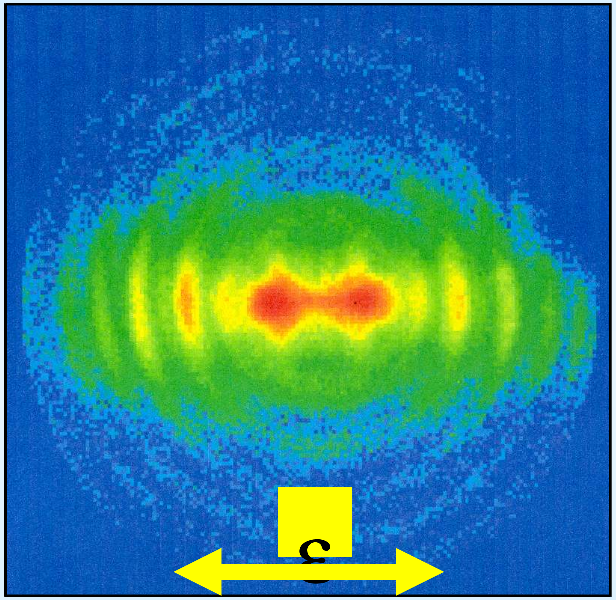
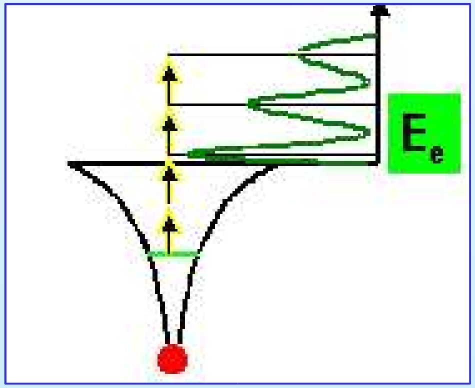
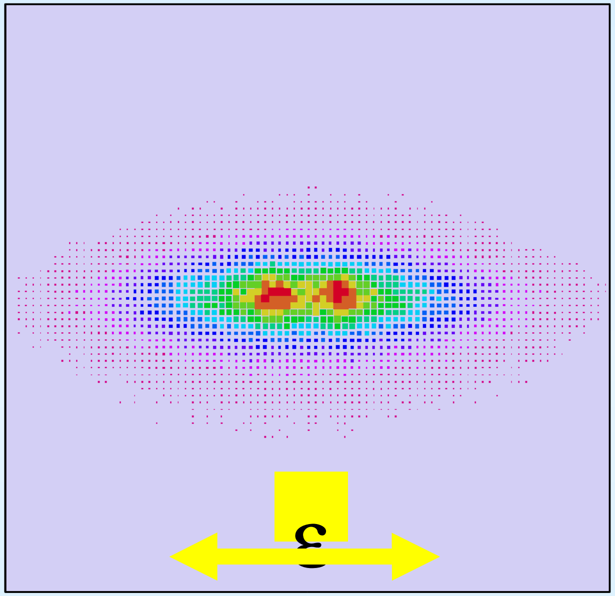
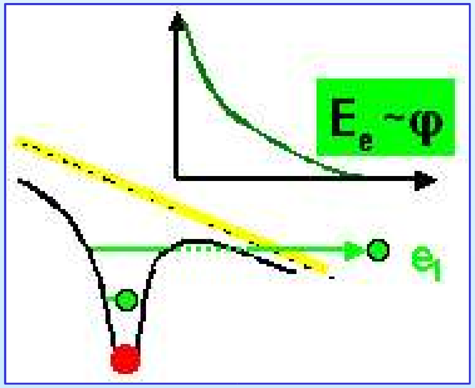




tunnelling ionization





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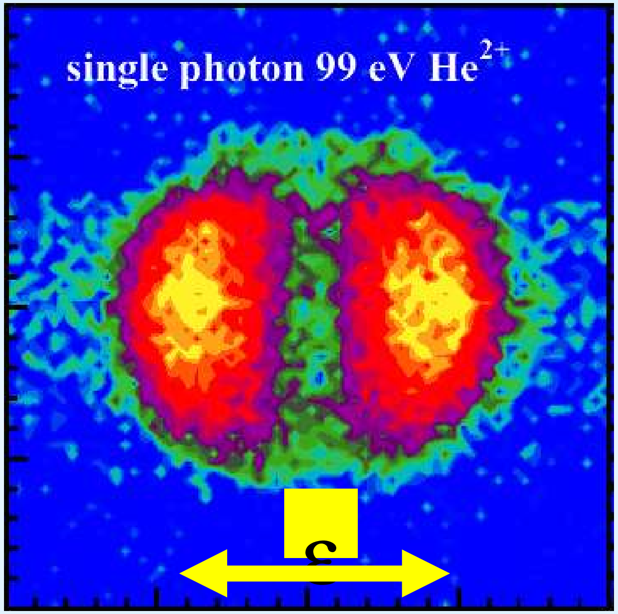
  - *- Pump-Probe: The Molecular Movie*

Double Ionization Fields

photo absorption

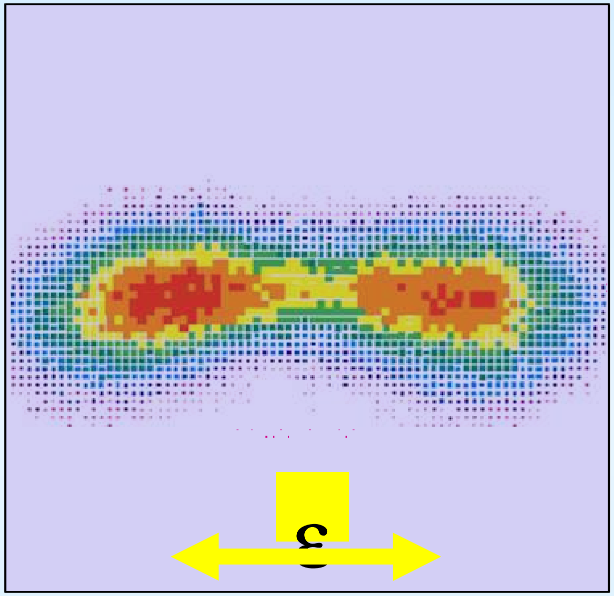


multi-photon ionization

not existent at all:
always many photons

53 for helium!

tunnelling ionization



what about
two active electrons

Double Ionization

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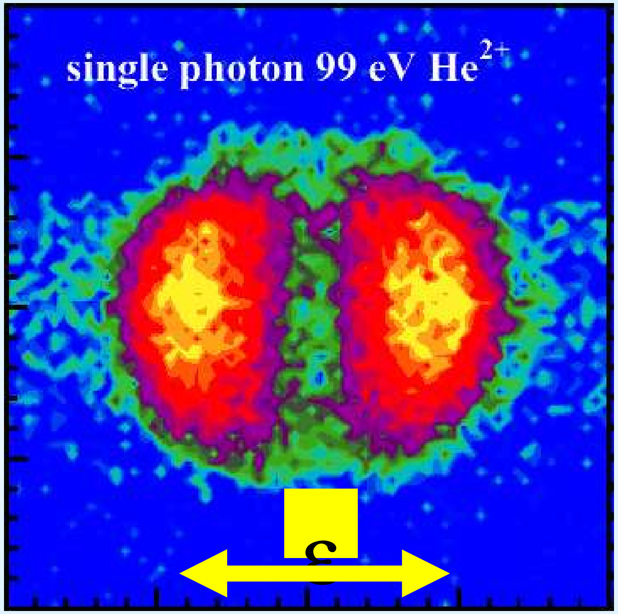
photo absorption

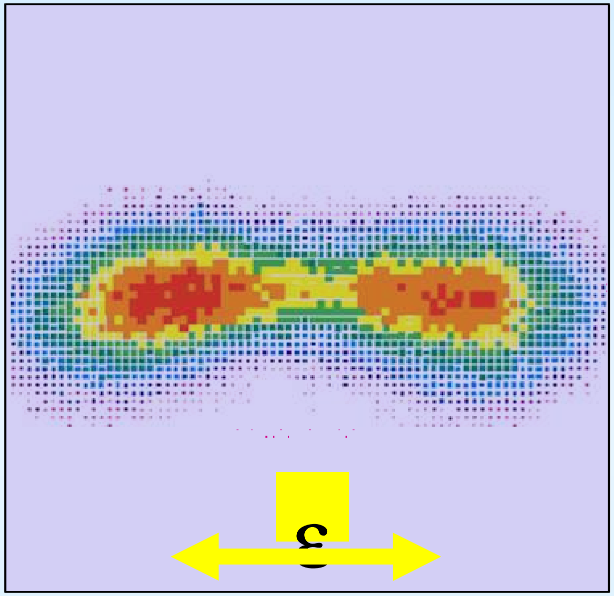
multi-photon ionization

tunnelling ionization



FEL

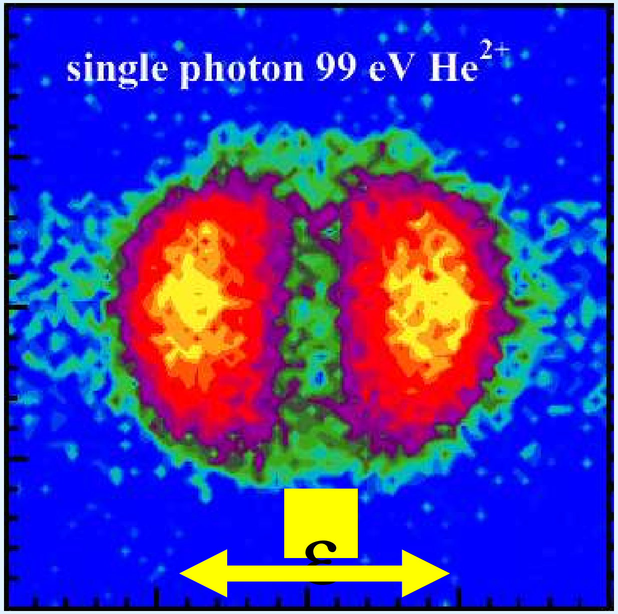
few photons interact with two electrons



what about two active electrons

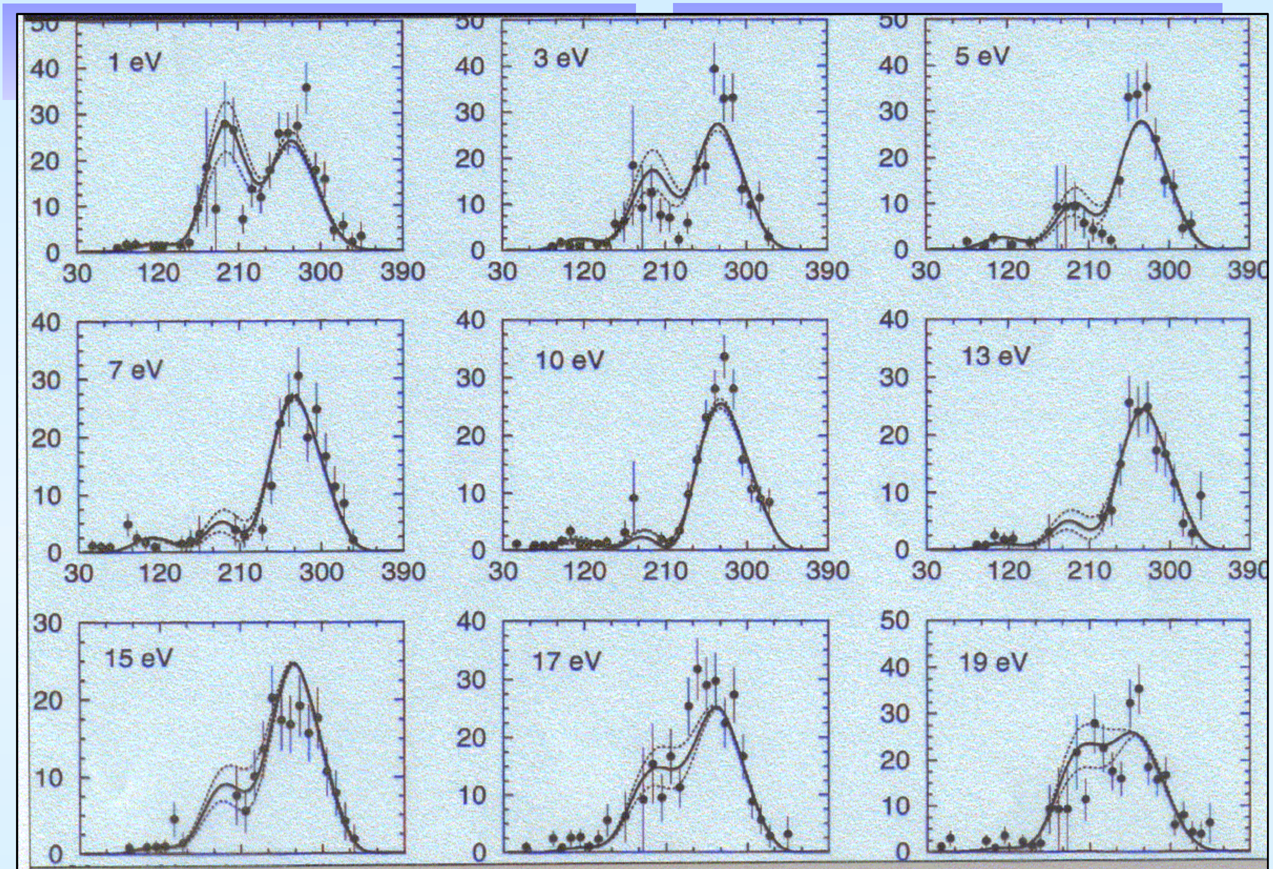
Double Ionization: 1 hν

photo absorption



- complete experiments
- reliable predictions

"well understood"



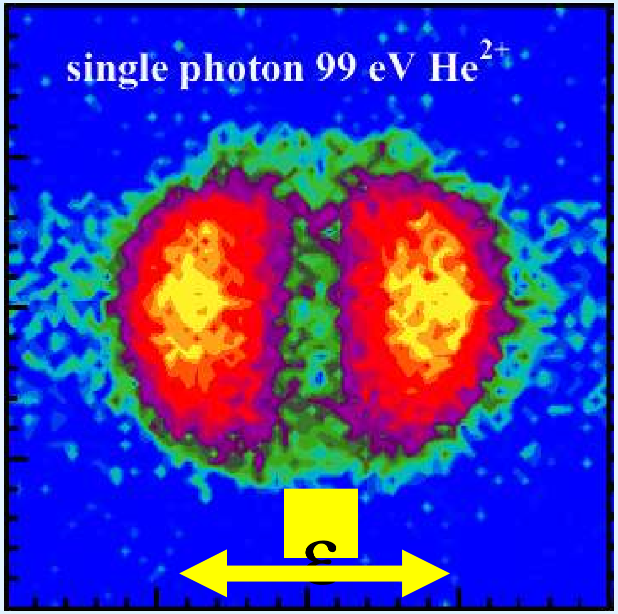
Experim.: *Bräuning, Dörner et al.*; Becker; Reddish; Huetz,...

Theory: *Selles, Malegat, Kazansky*; Bray; Shakeshaft; Robicheaux; .

two active electrons

Double Ionization: 1 hν

photo absorption



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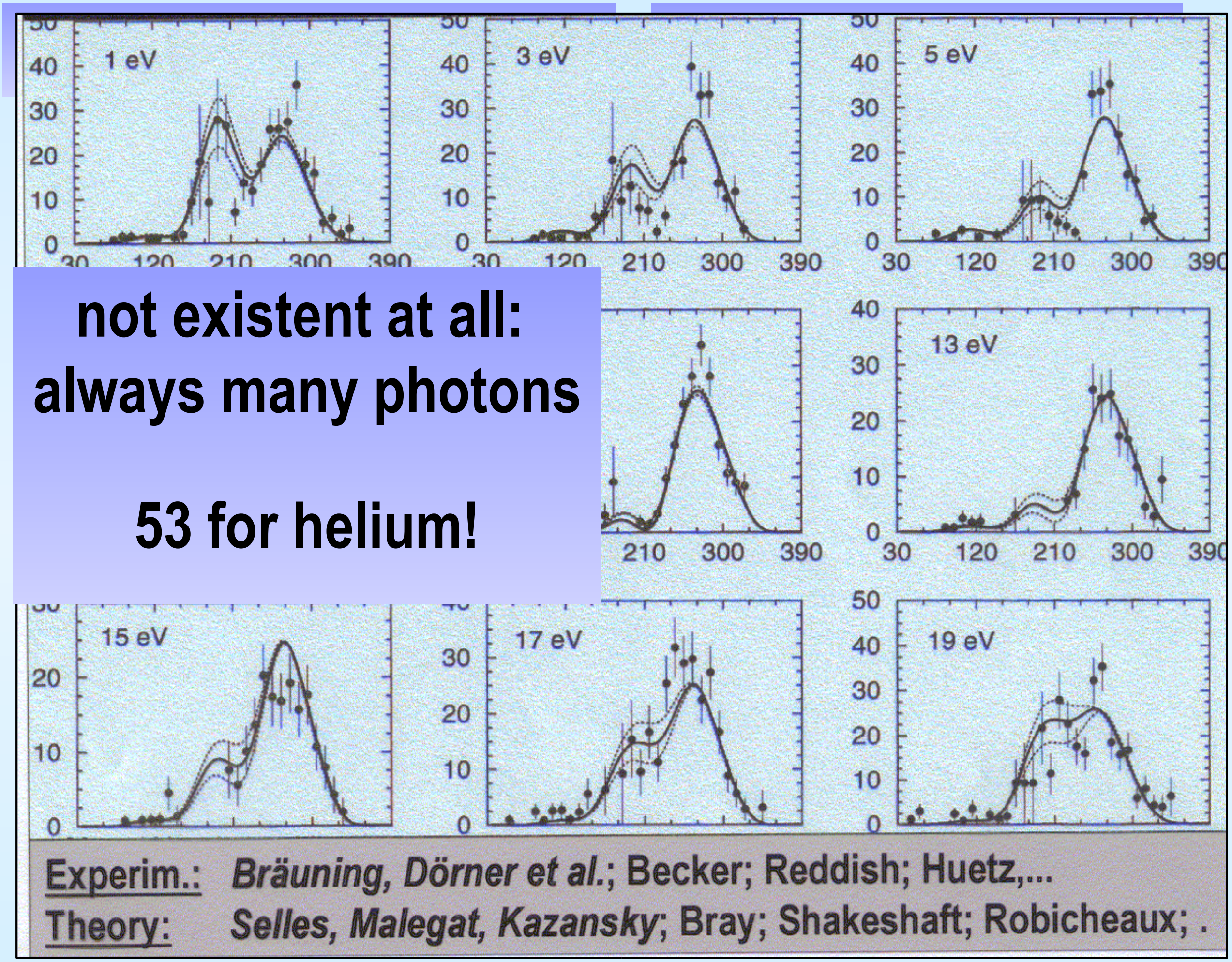

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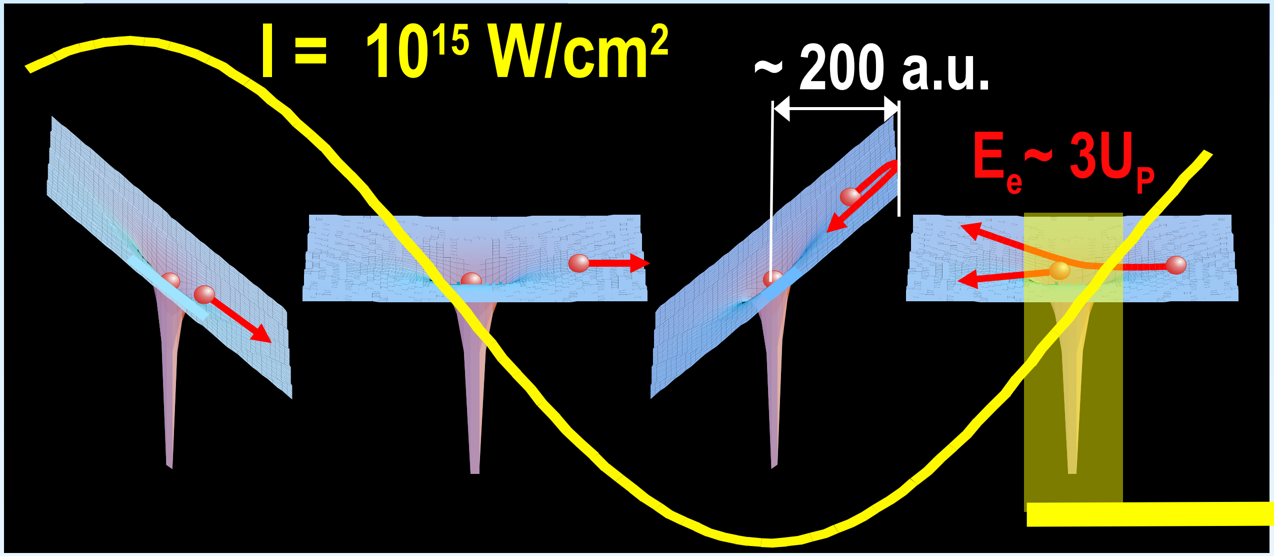
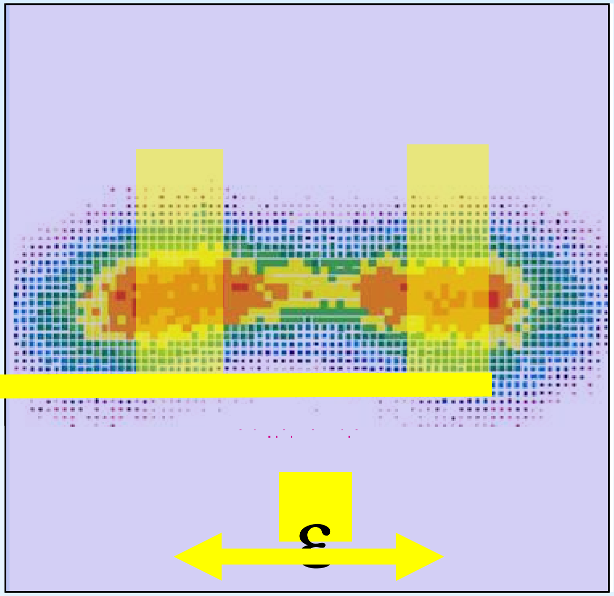
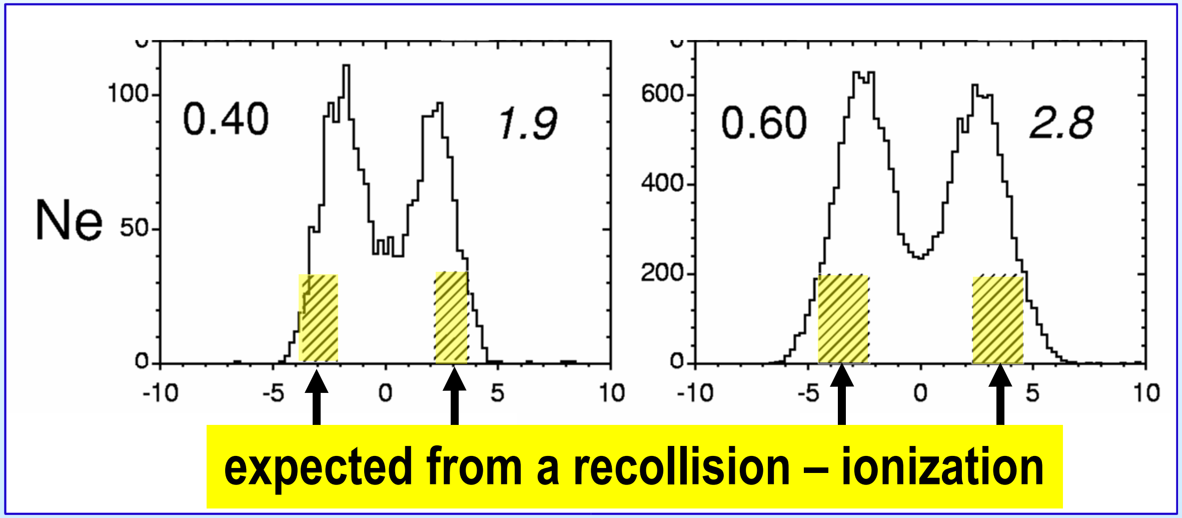
two active electrons

Double Ionization: n· hν



tunnelling ionization

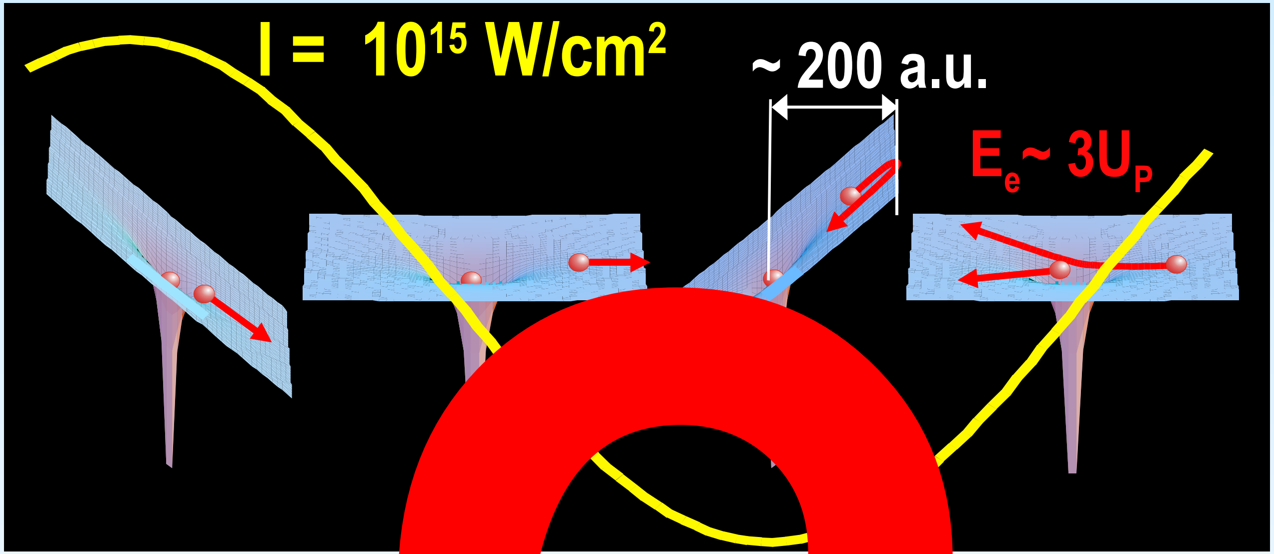




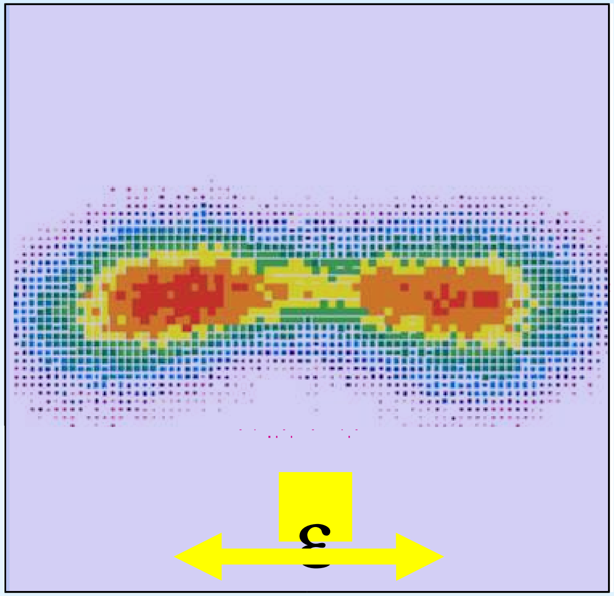
expected from a recollision – ionization

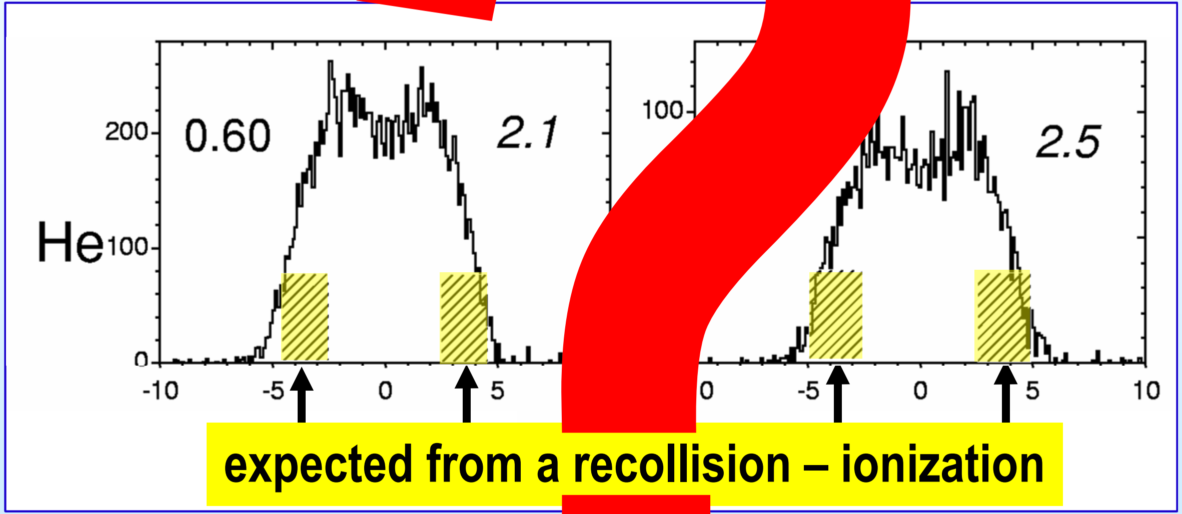
two active electrons

Double Ionization: n· hν



tunnelling ionization

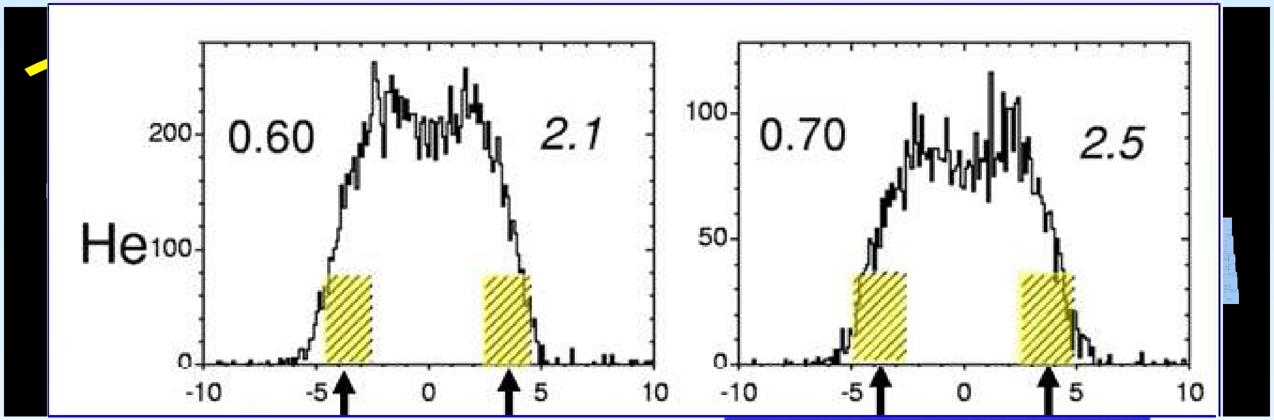


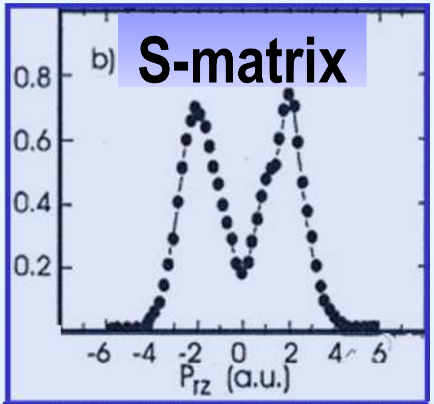


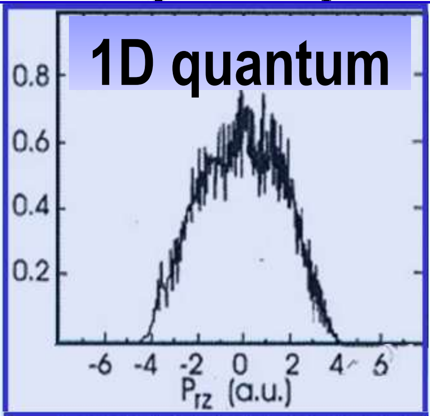
expected from a recollision – ionization

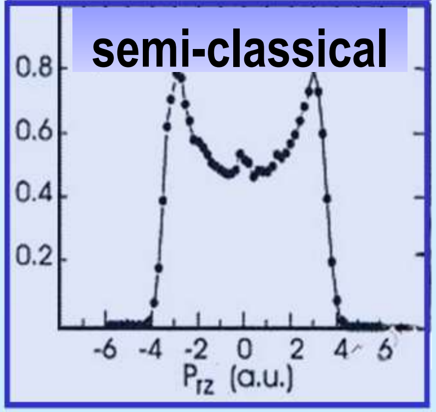
tw ctive electrons

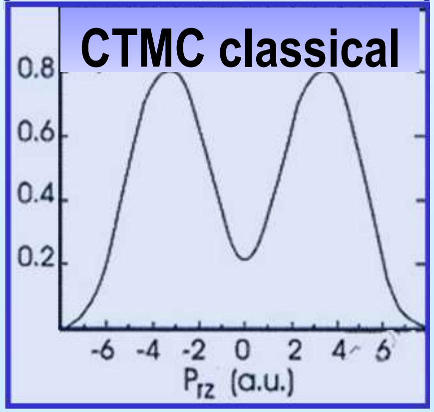
Double Ionization: $n \cdot h\nu$



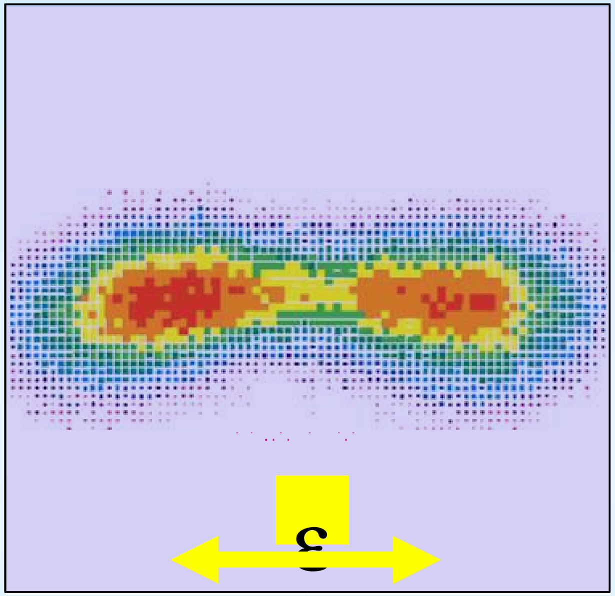

S-matrix


1D quantum


semi-classical


CTMC classical

tunnelling ionization



- differential experiments
- many model approaches

“not understood”

ons

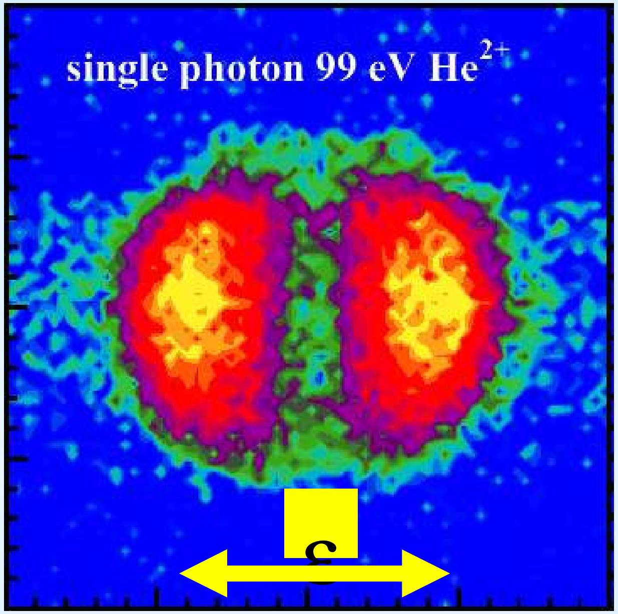
Double Ionization: n· hν

Max-Planck-Institut
für Kernphysik

photo absorption

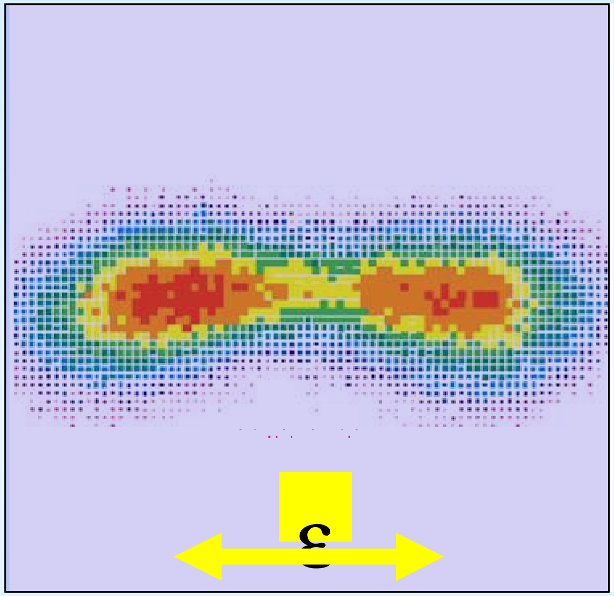
multi-photon ionization

tunnelling ionization



not existent at all:
always many photons

53 for helium!



- complete experiments
- reliable predictions

“well understood”

- differential experiments
- many model approaches

“not understood”

two active electrons

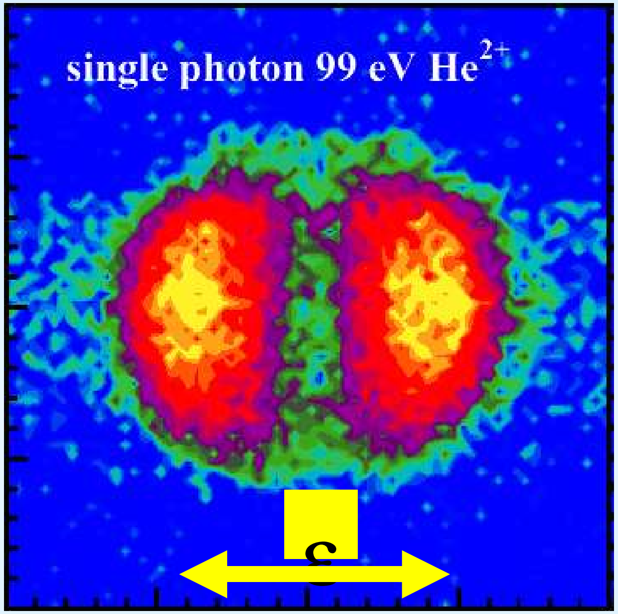
Double Ionization: n· hν

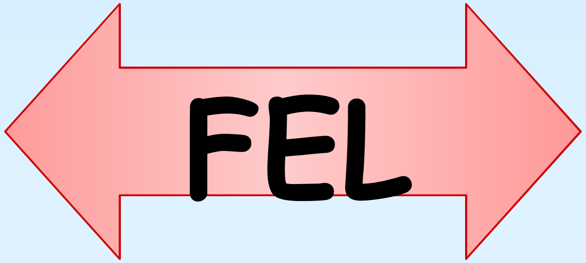
Max–Planck–Institut für Kernphysik

photo absorption

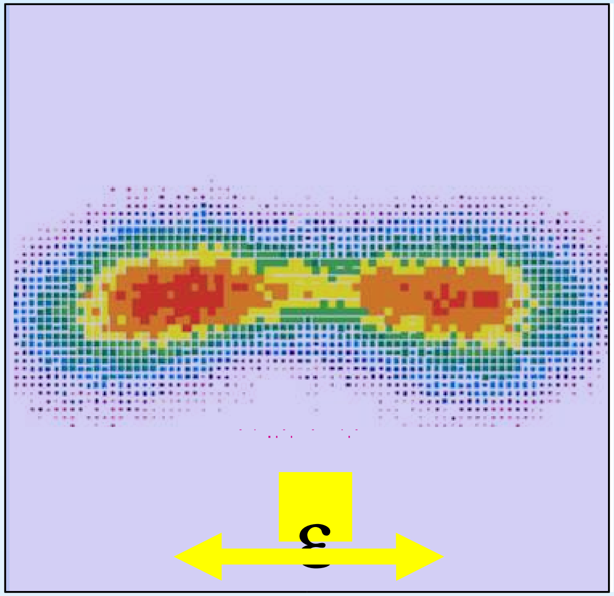
multi-photon ionization

tunnelling ionization





few photons interact with few electrons



- complete experiments
- reliable predictions

"well understood"

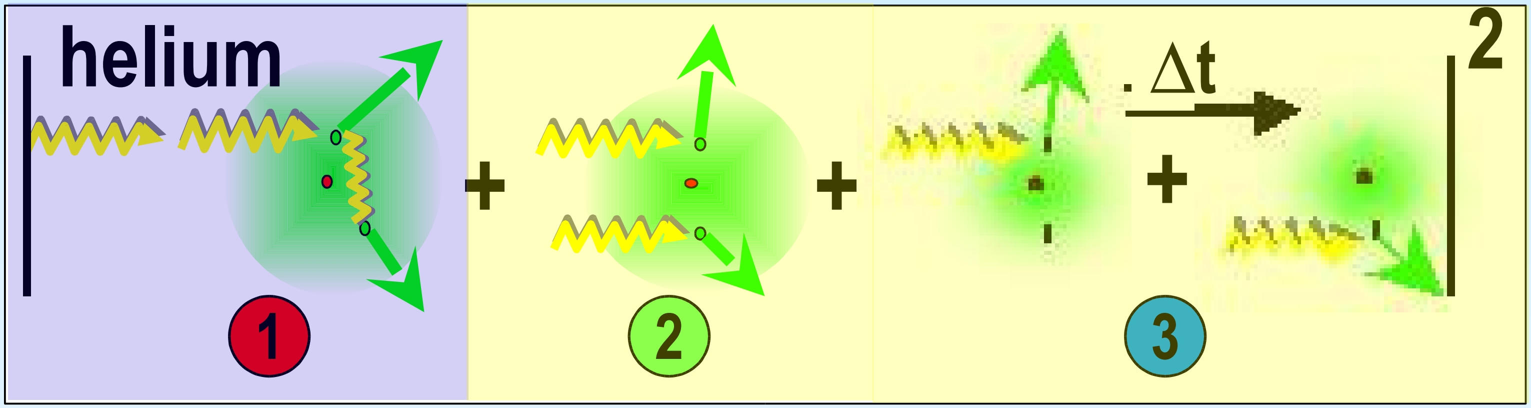
- complete experiments
- few, defined photons !
- $U_p = I/\omega^2$ is small !
- reliable predictions ?!

- differential experiments
- many model approaches

"not understood"

two active electrons

Few Photons at the FEL!



“no correlated al”

uncorrelated ntial”

Few Photons at the FEL!

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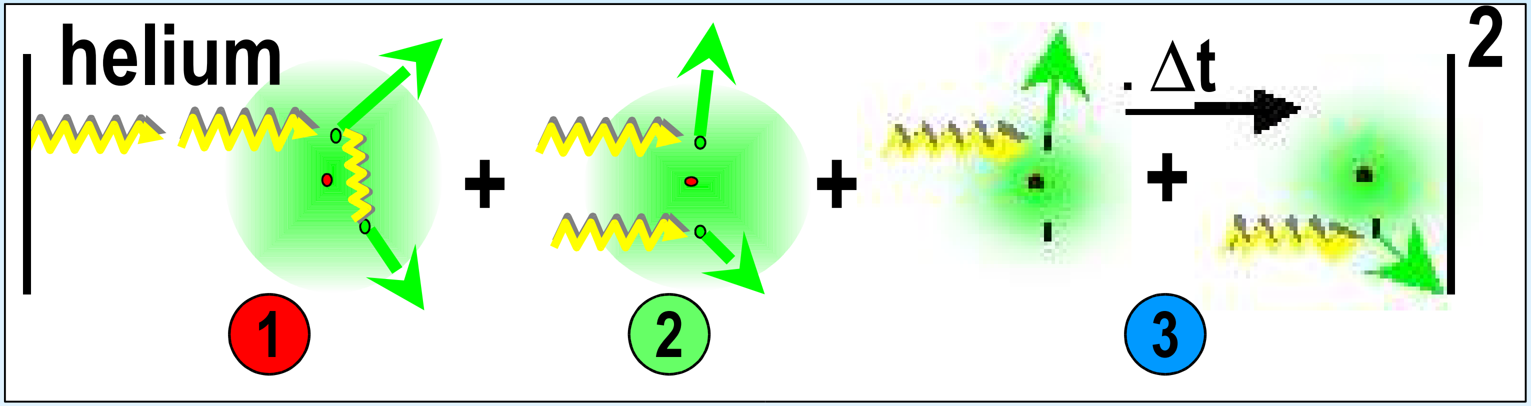
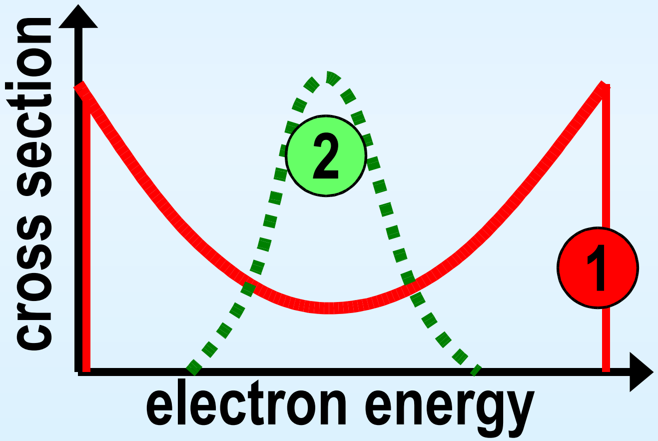
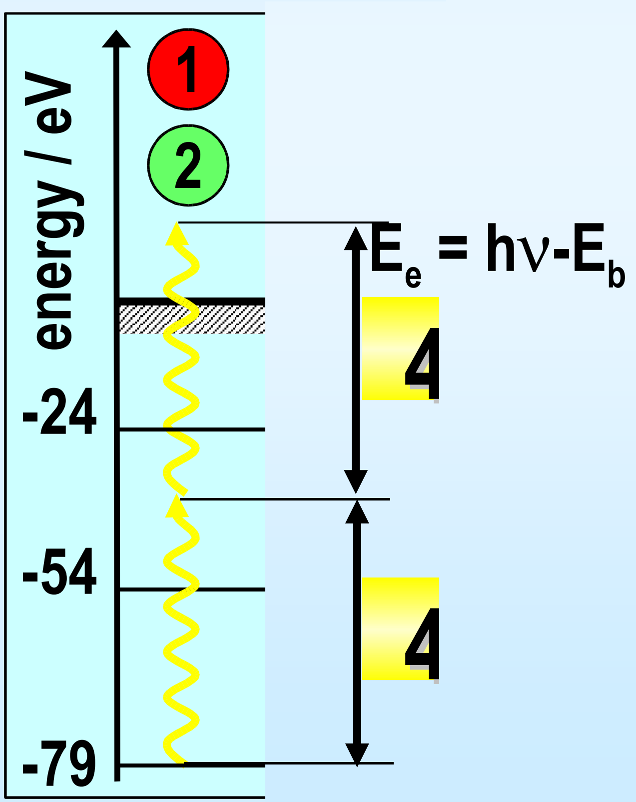
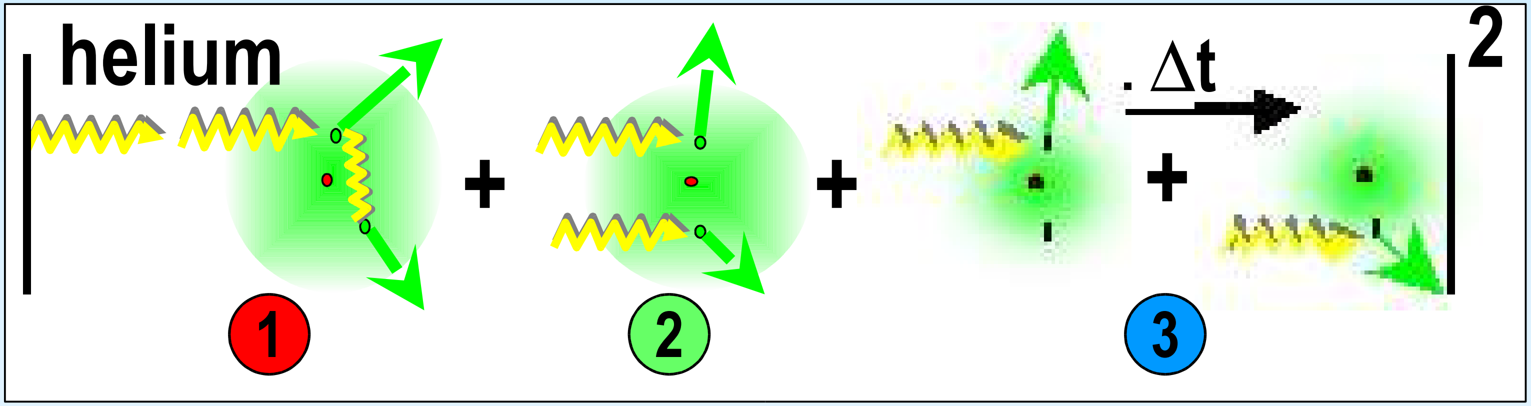


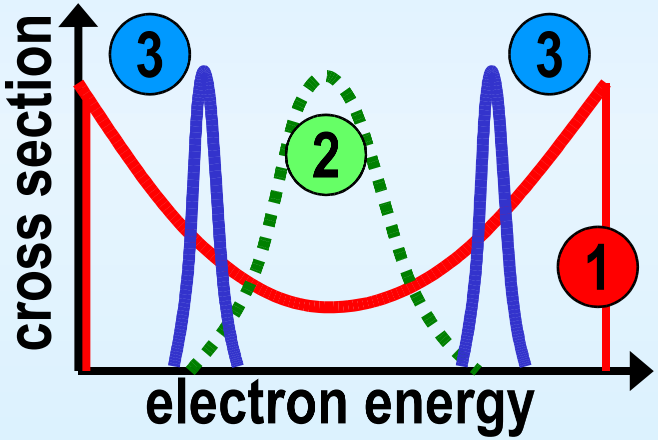


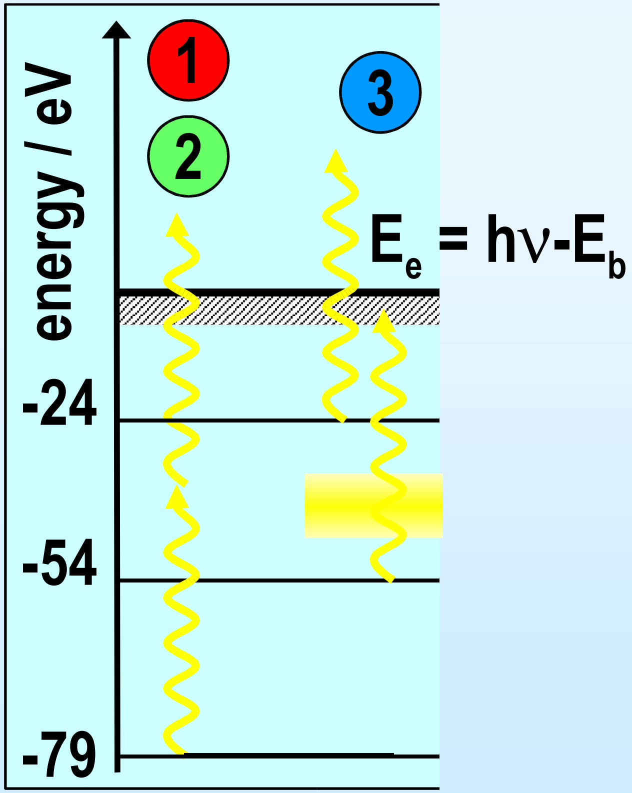


energy sharing between electrons?

Few Photons at the FEL!

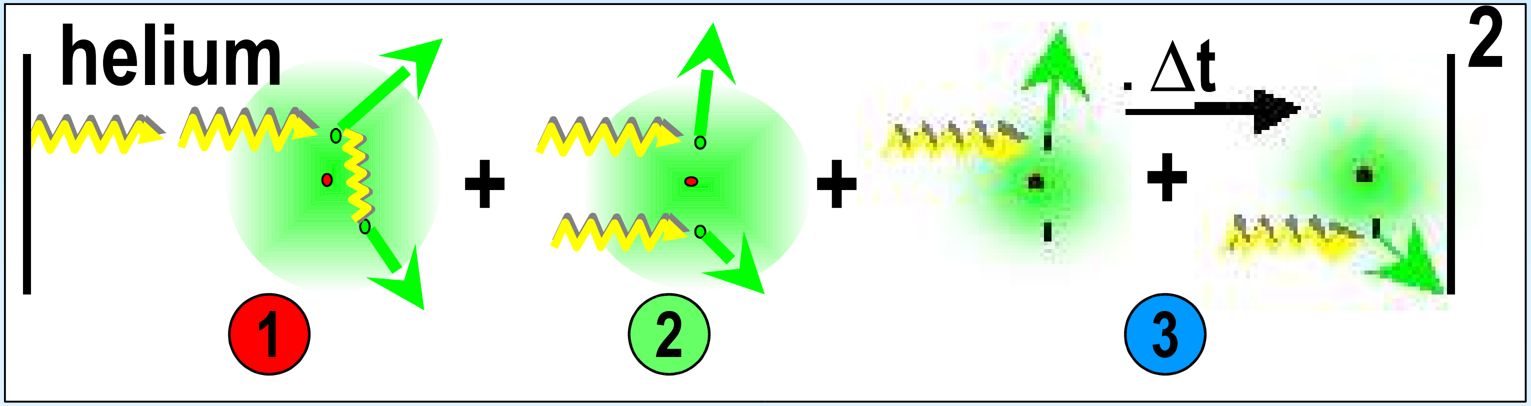


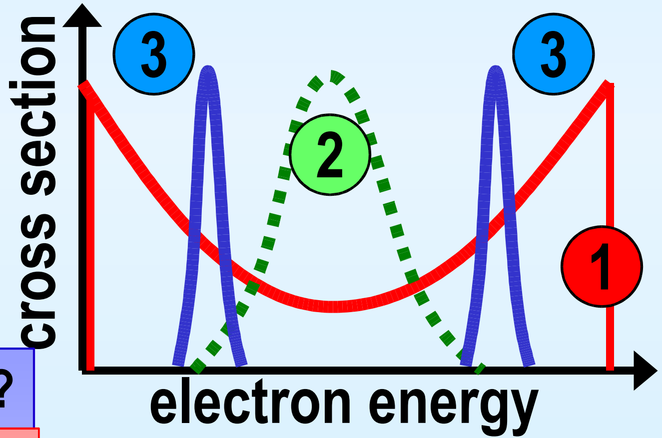


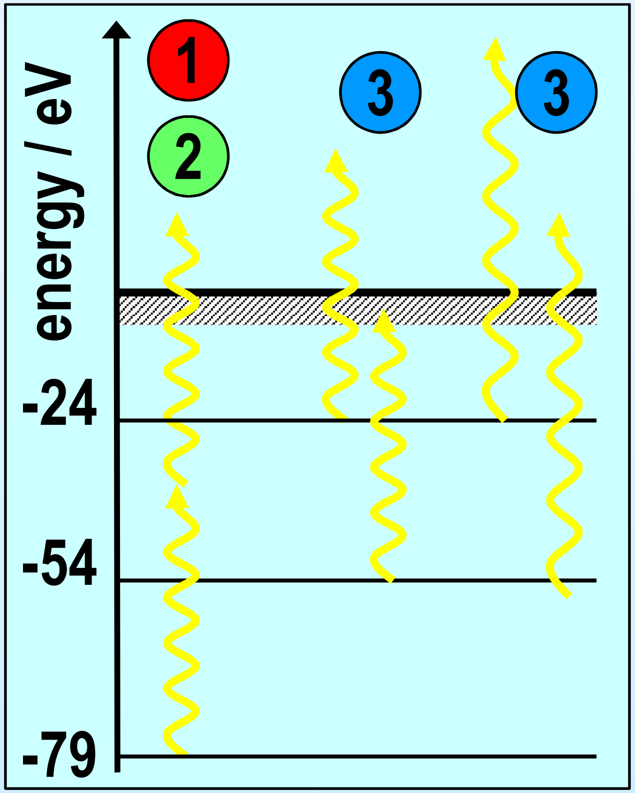


intensity dependence:
electronic relaxation time

Few Photons at the FEL!







- intensity dependence ?

main goal of first experiments

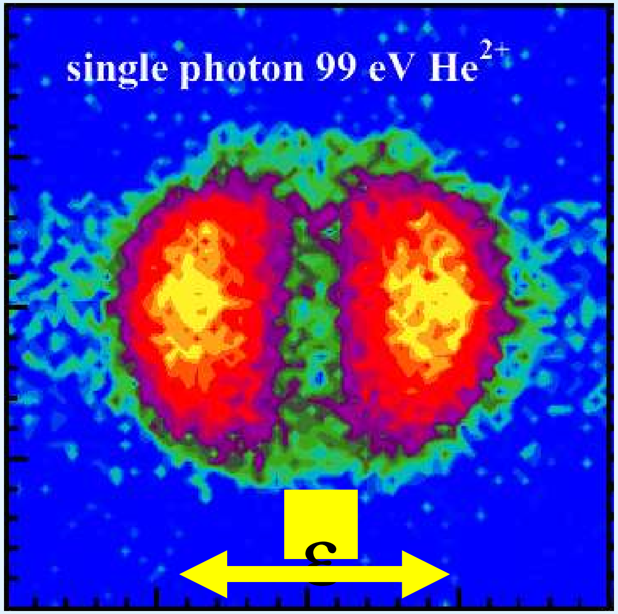
many questions

Double Ionization: n· hν

photo absorption

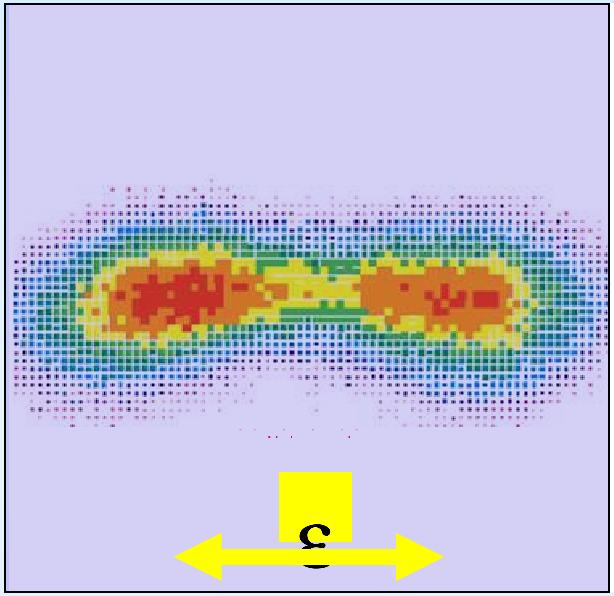
multi-photon ionization

tunnelling ionization



FEL

few photons interact with few electrons



- complete experiments
- reliable predictions

"well understood"

- complete experiments
- few, defined photons !
- $U_P = I/\omega^2$ is small !
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- differential experiments
- many model approaches

"not understood"

two active electrons

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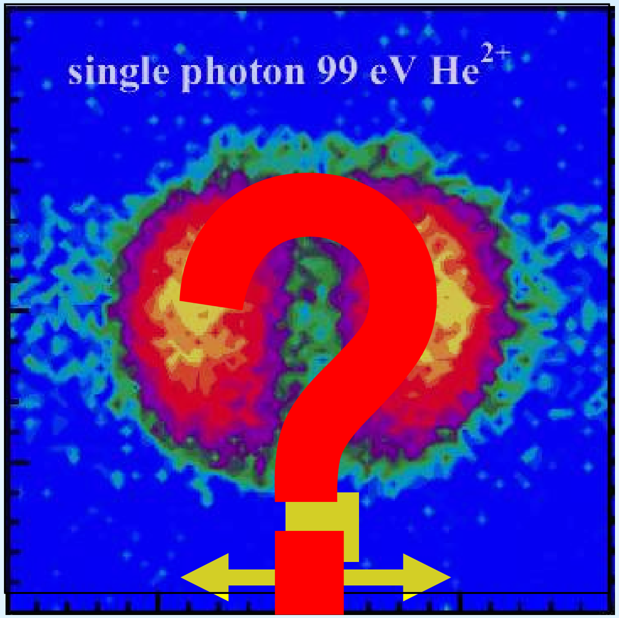
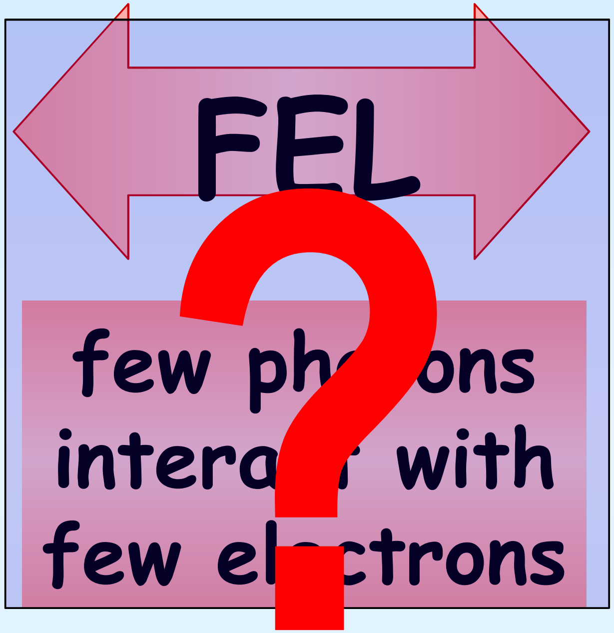
  - *- Pump-Probe: The Molecular Movie*

Multiple Ionization

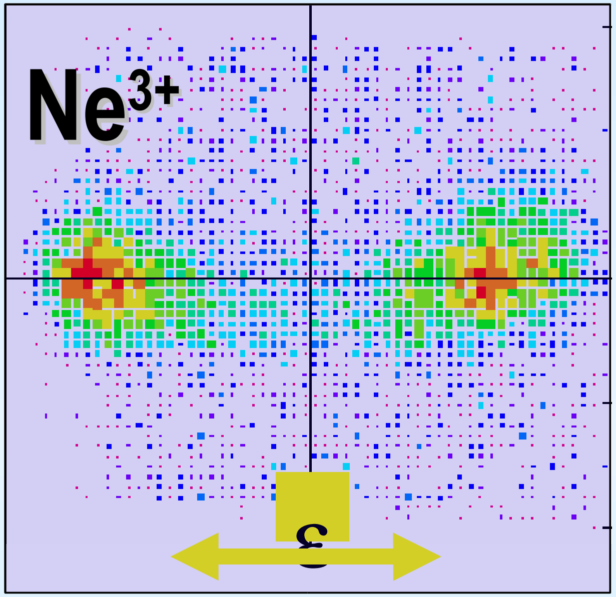
photo absorption	multi-photon ionization	tunnelling ionization





FEL

few photons
interact with
few electrons



- no fully diff. data

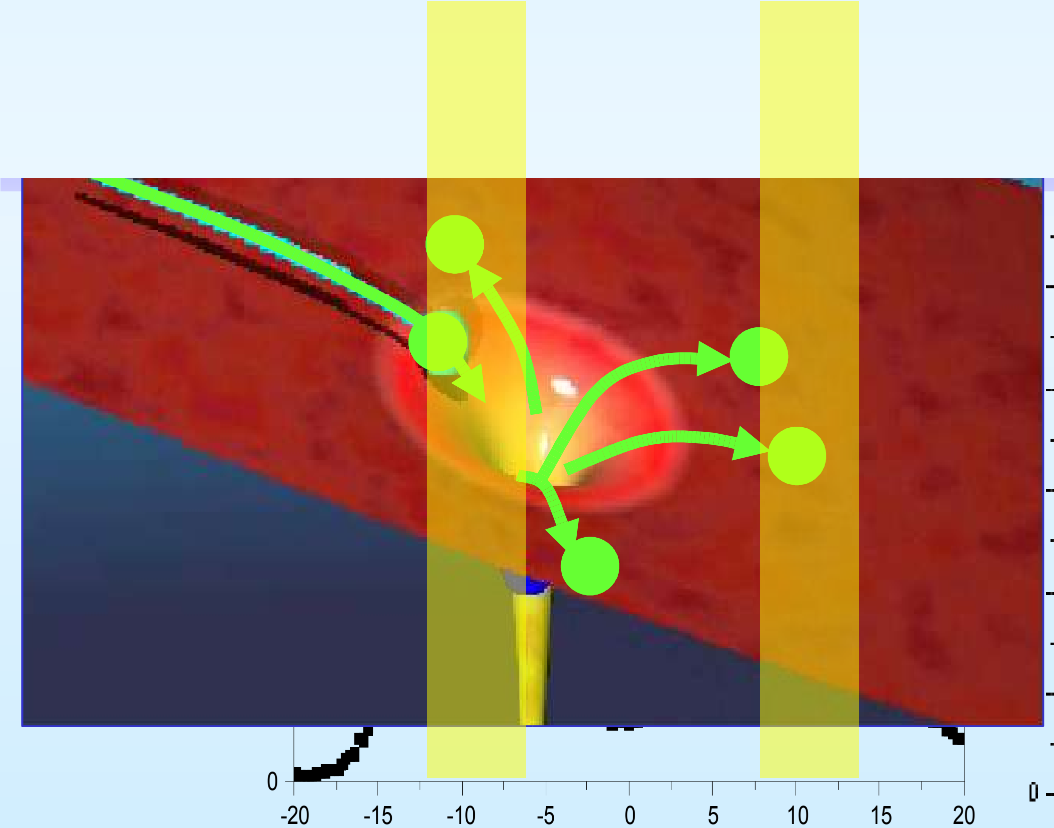
- no differential data

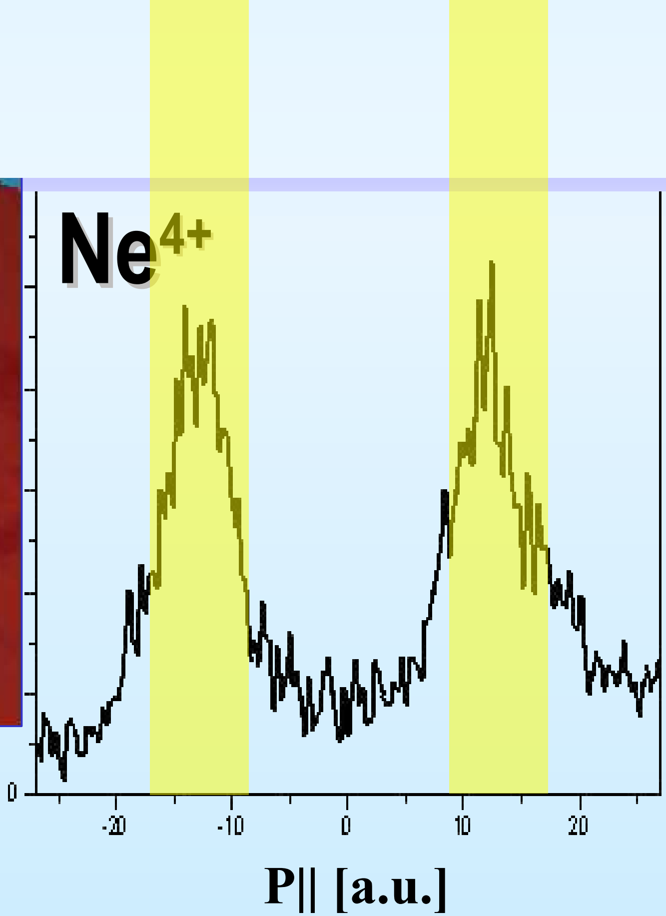
- total yields
- first differential data

Surprises!

Structure Dependence

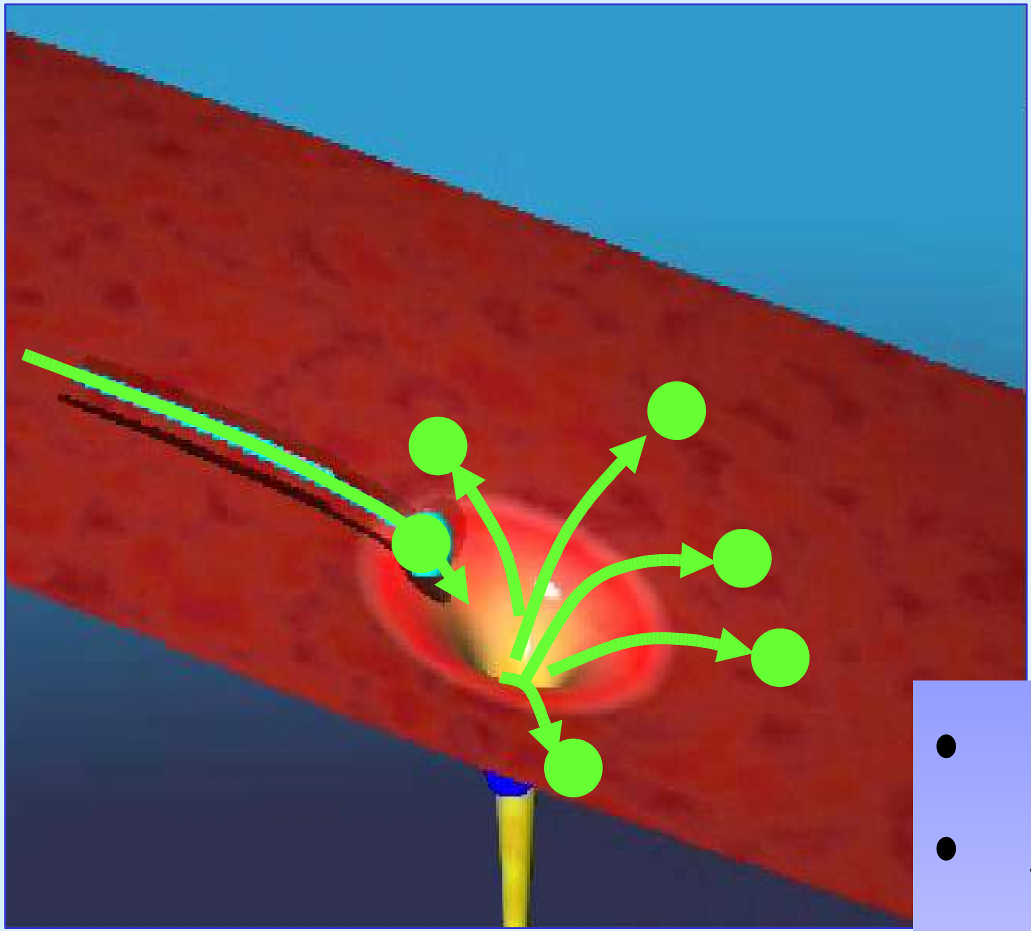
produced at well defined phases in the field!

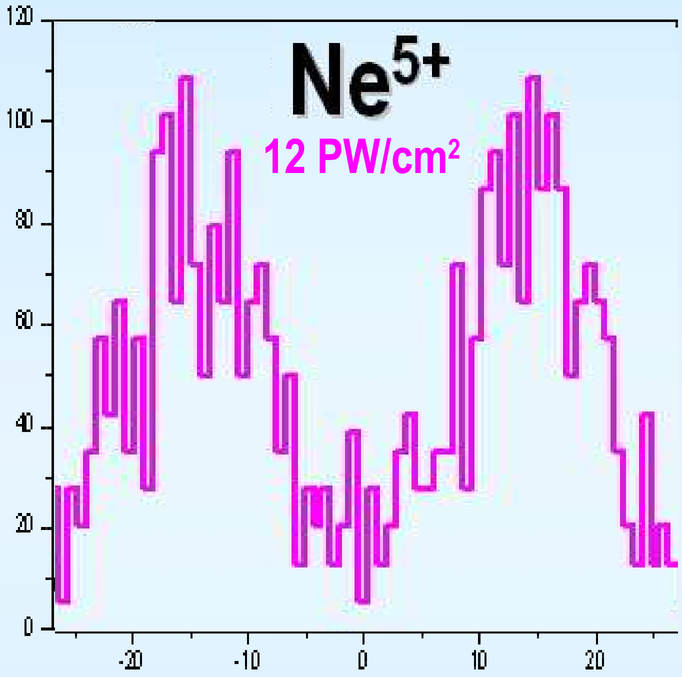




A. Rudenko et al., PRL 93 (2004) 253001

Structure Dependence





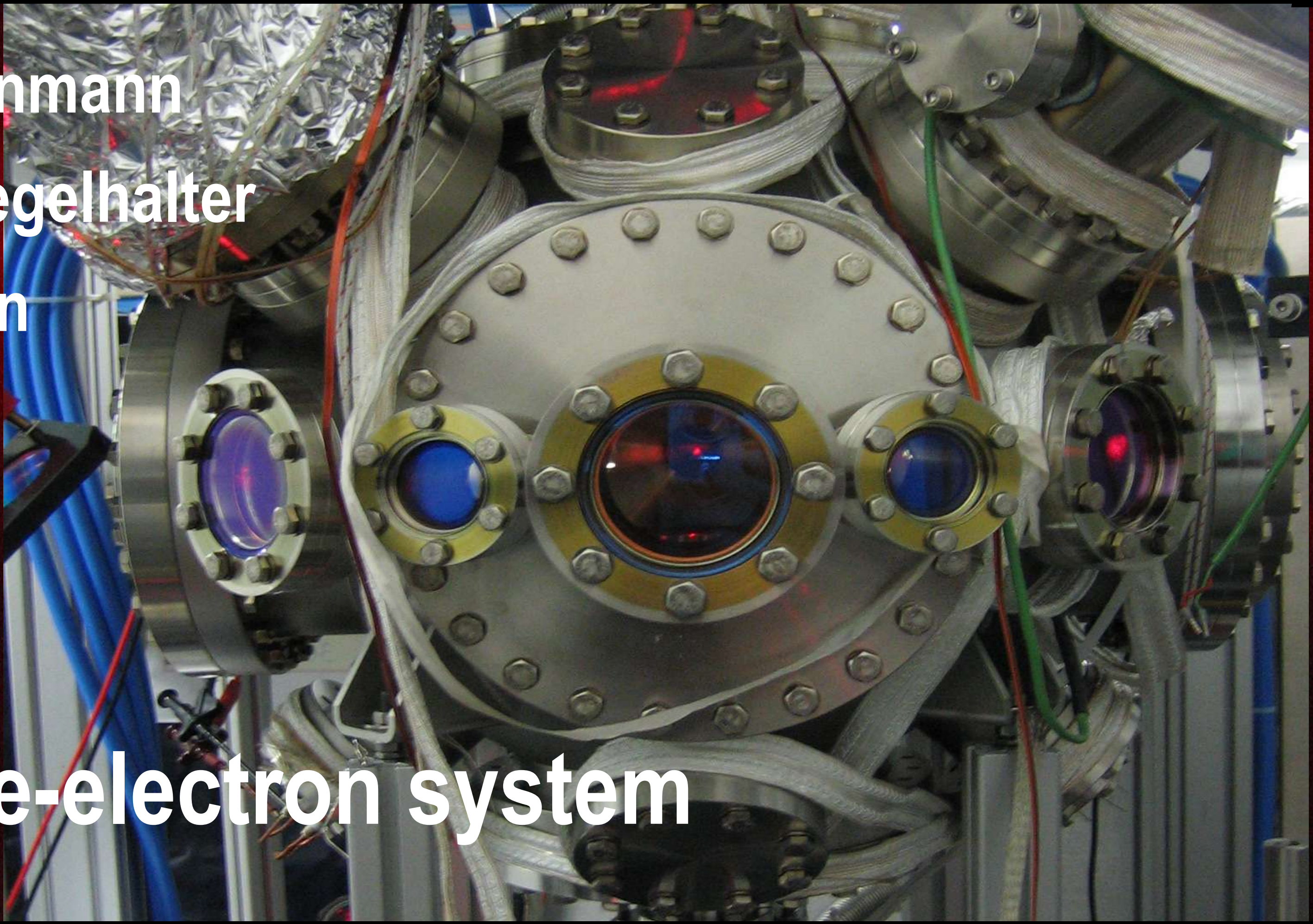
W. Becker, P. Corkum (in preparation)

A. Rudenko et al., PRL 93 (2004) 253001

- *field assisted,*
- *strongly correlated,*
- *400-attosecond*

many-electron transition

Reaction Microscope



J. Steinmann

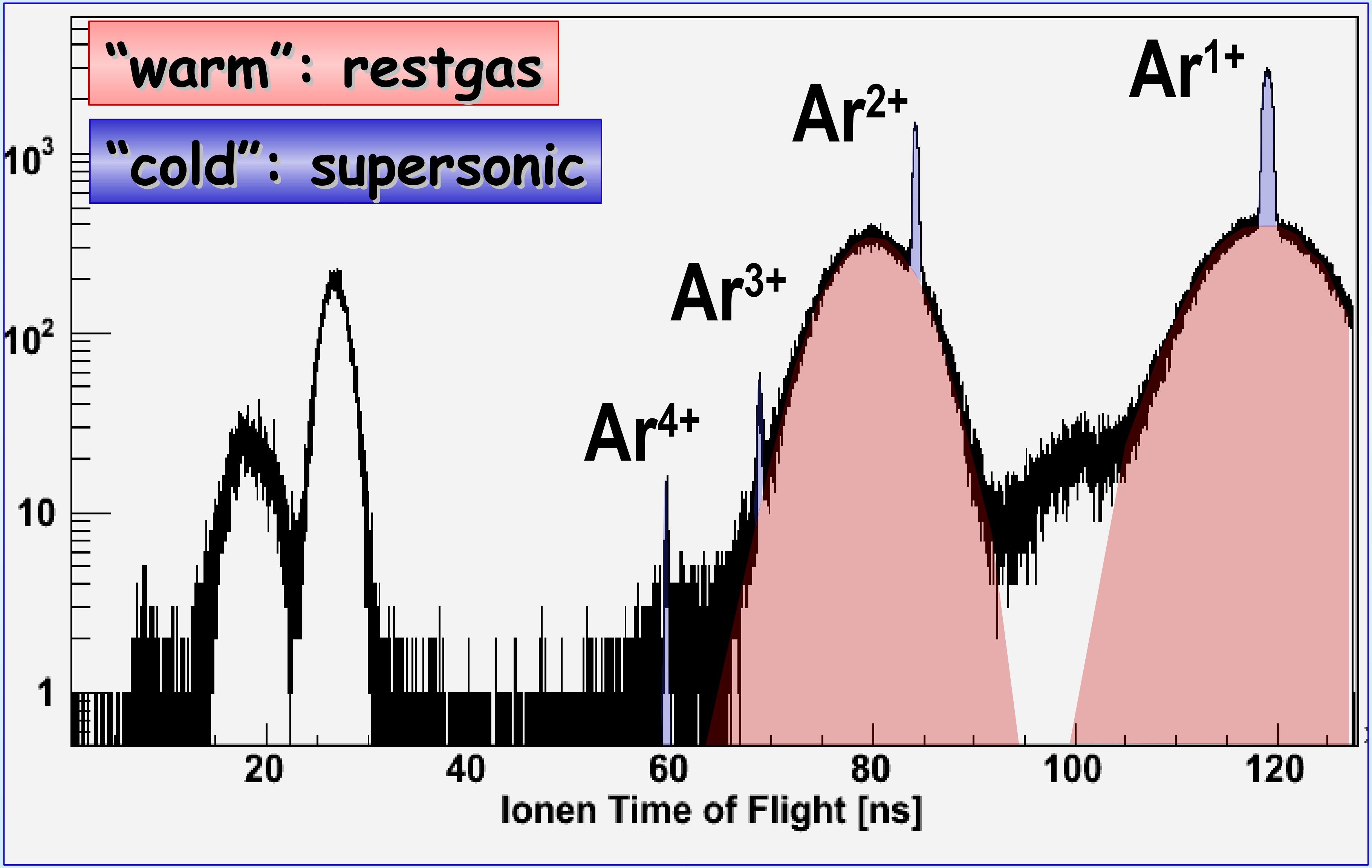
F. Spiegelhalter

A. Dorn

- three-electron system

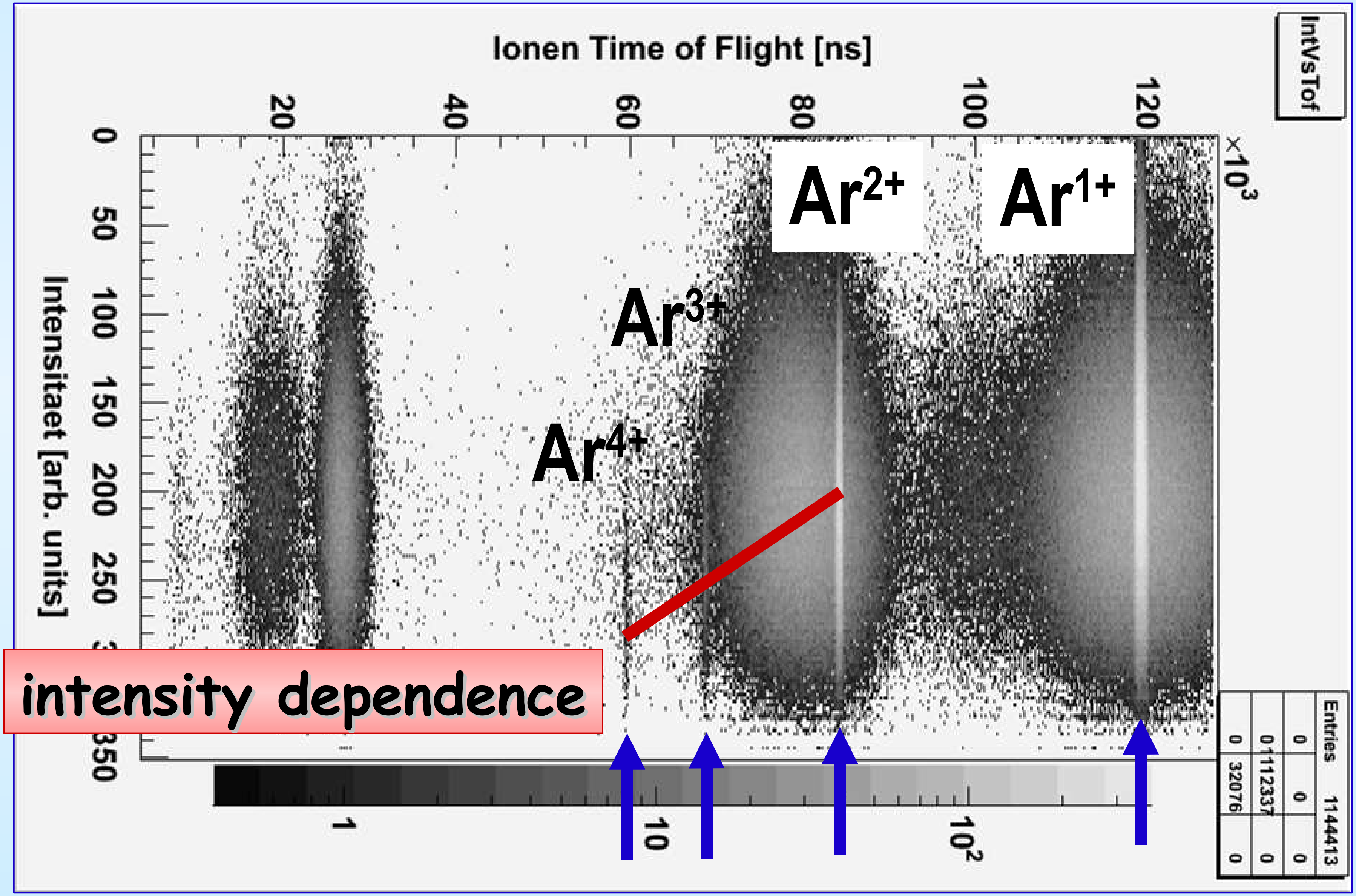
A Lithium MOT!

First Results from DESY



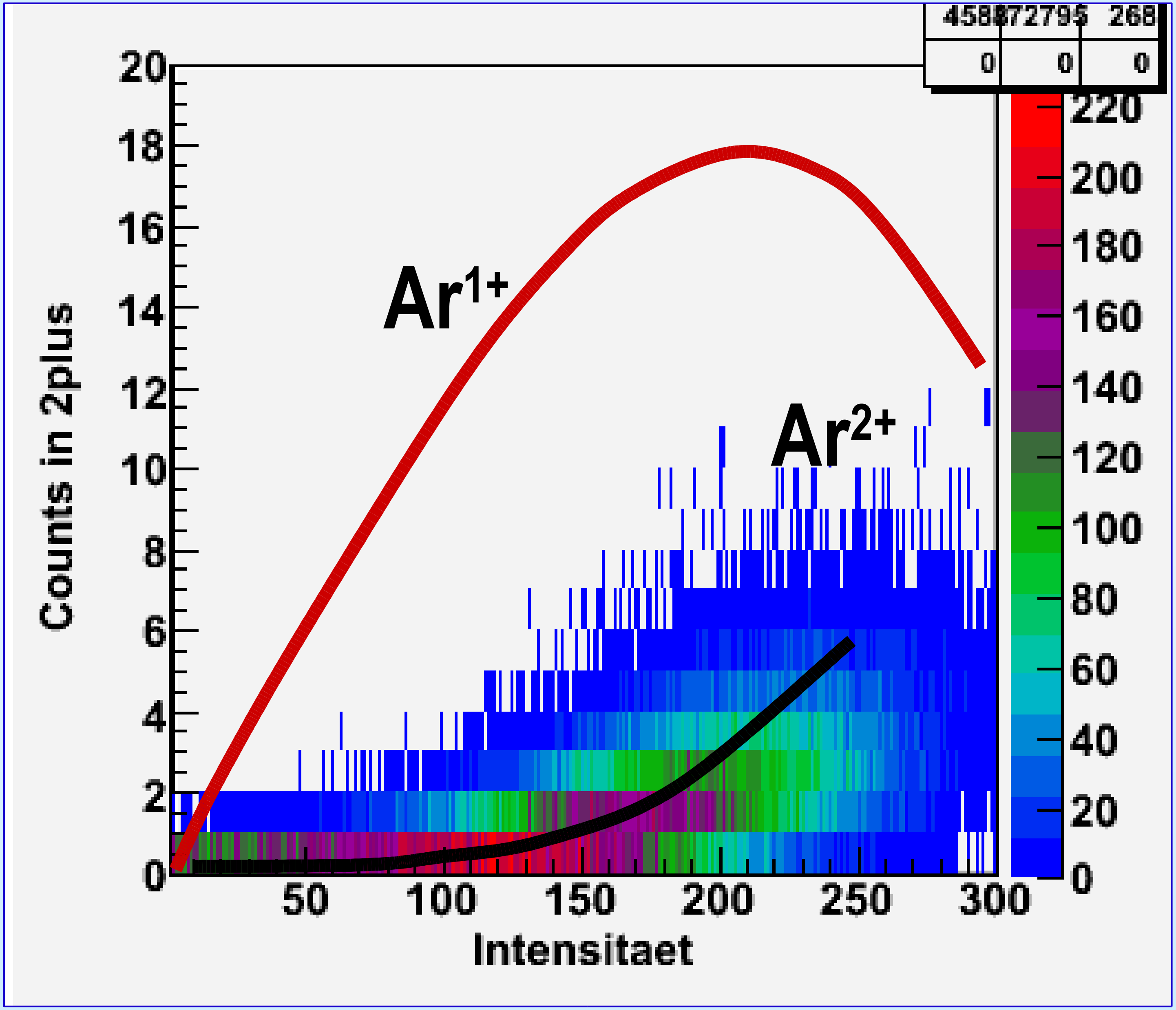
Intensity Dependence!

Max-Planck-Institut für Kernphysik



intensity dependence

Intensity Dependence!



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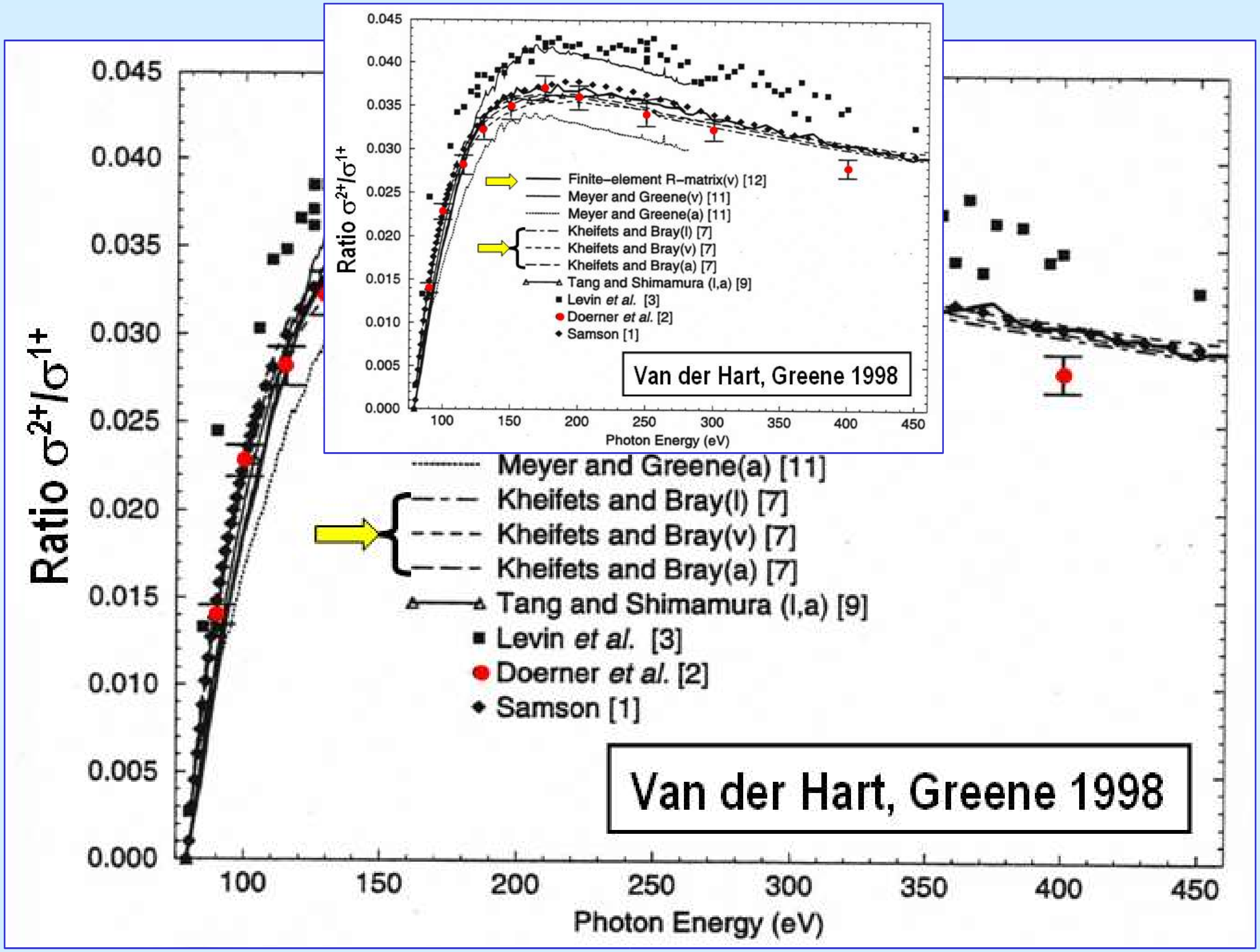
  - *- Attosecond Correlation*
  - *- Pump-Probe: The Molecular Movie*

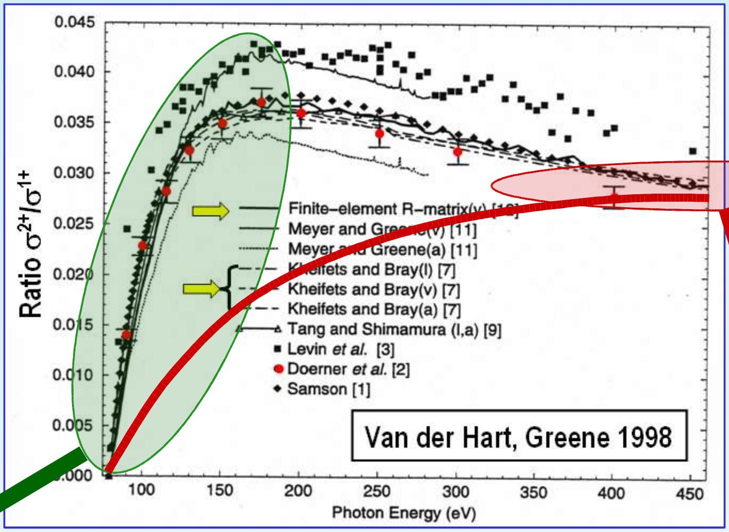
Electron Correlation:

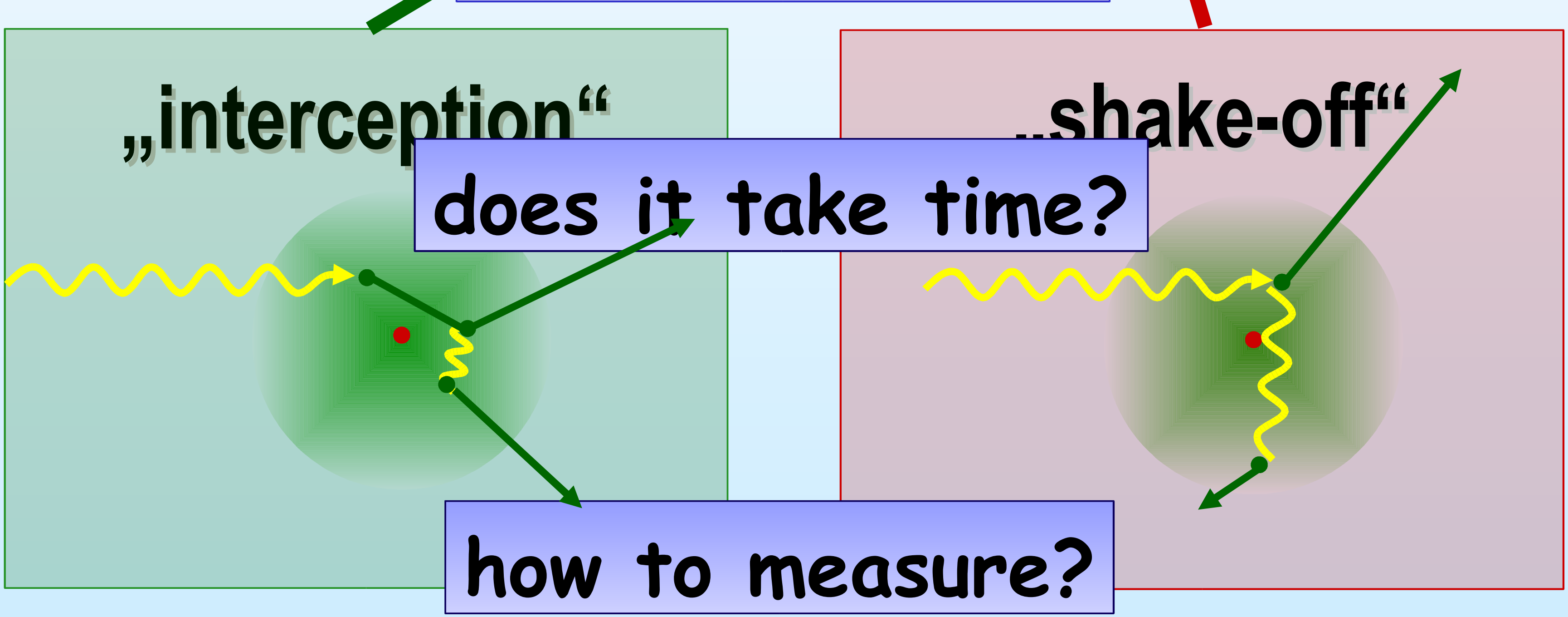
Max-Planck-Institut für Kernphysik



Electron Correlation:

Max-Planck-Institut für Kernphysik





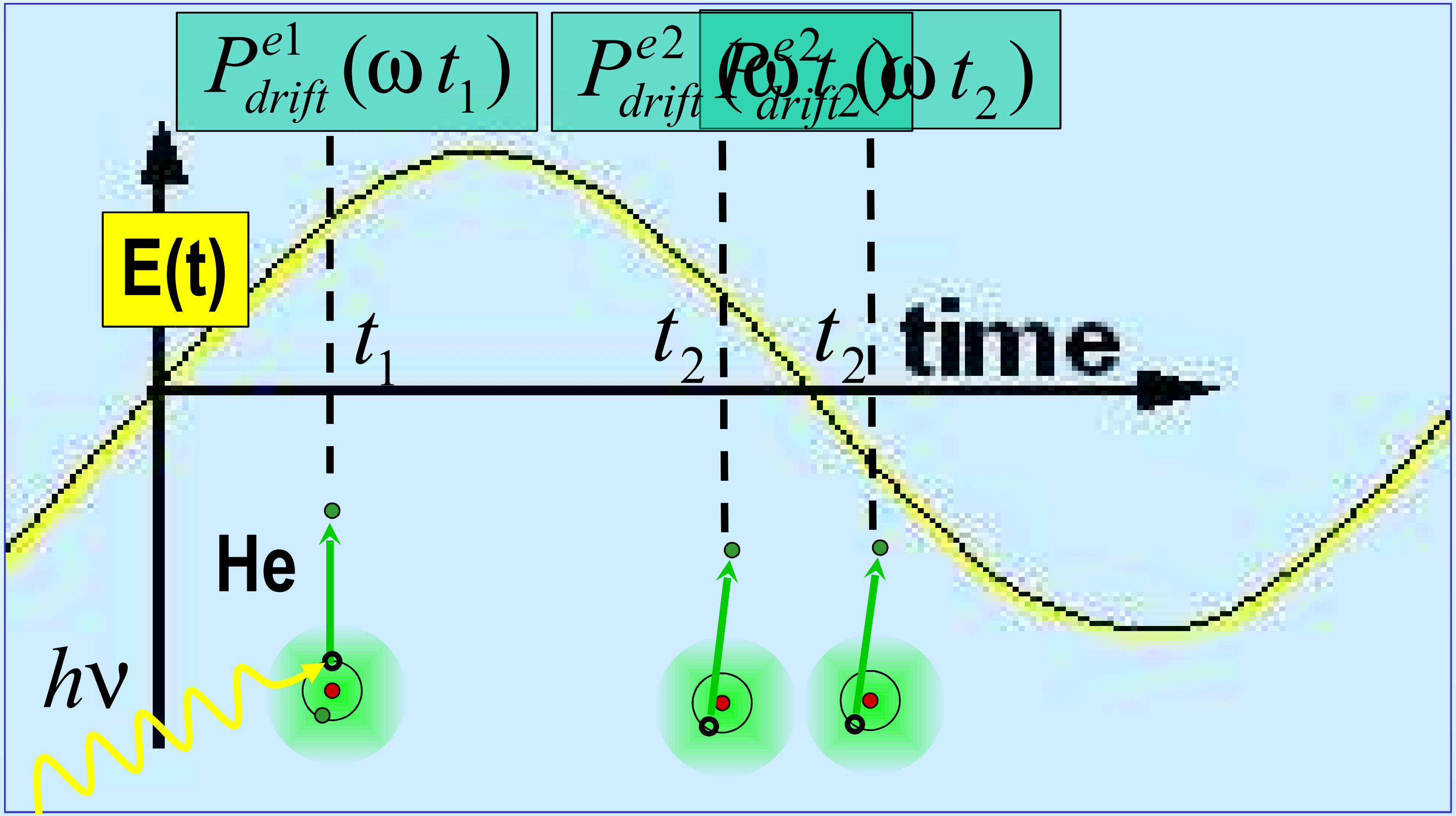
does it take time?

how to measure?

Electron Correlation:

Max-Planck-Institut für Kernphysik

Δt ~ 80 as. . . Heisenberg??



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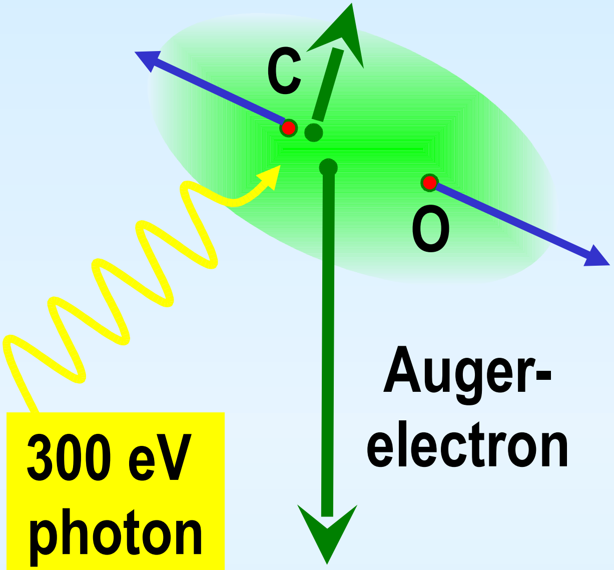
Dissociation Dynamics

Max-Planck-Institut für Kernphysik

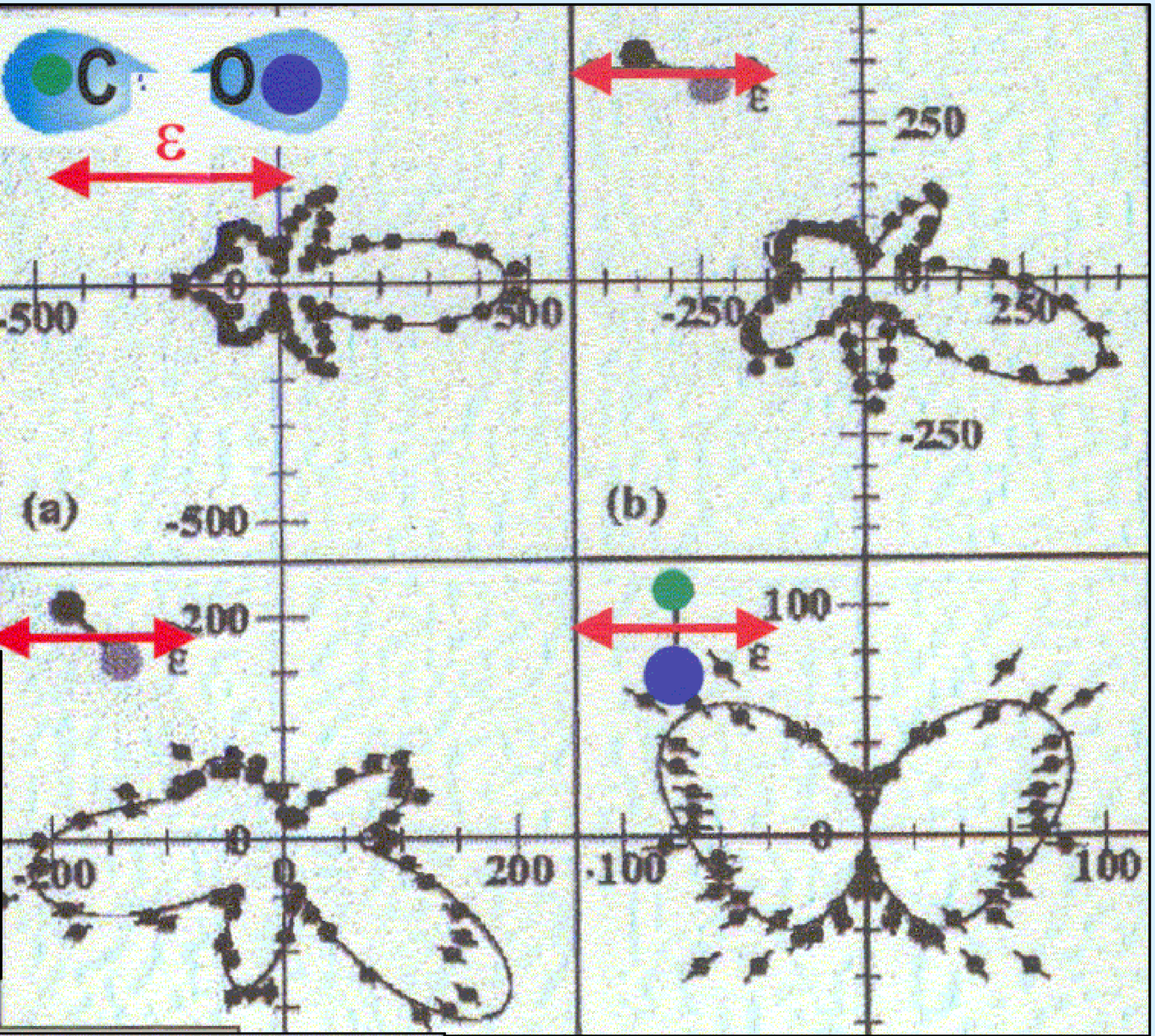


10 eV photo-electron

Auger-electron

300 eV photon

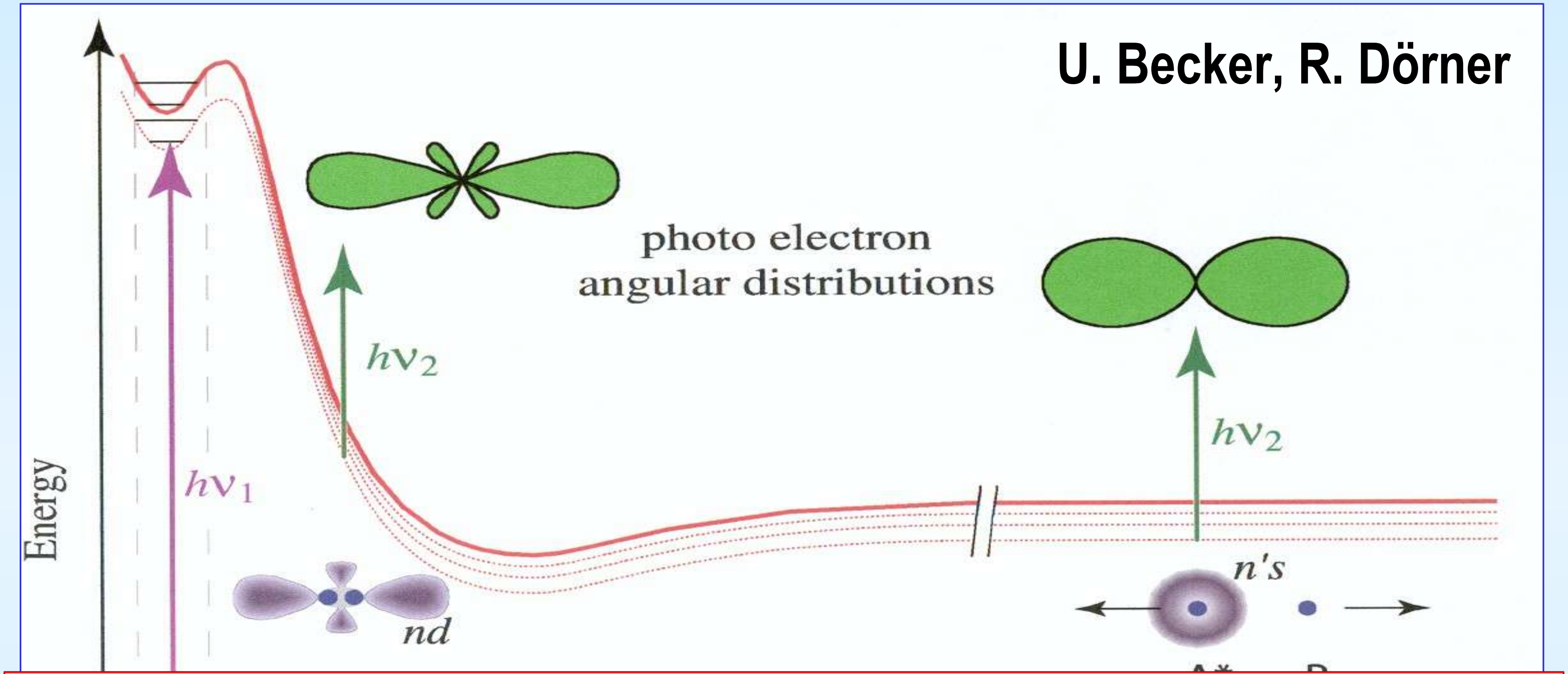
- E. Shigemasa et al., RRL 74 (1995)
- F. Heiser,.., U. Becker PRL 79 (1997)



- Launders, Dörner PRL 87 (2001)

Dissociation Dynamics

Max-Planck-Institut für Kernphysik

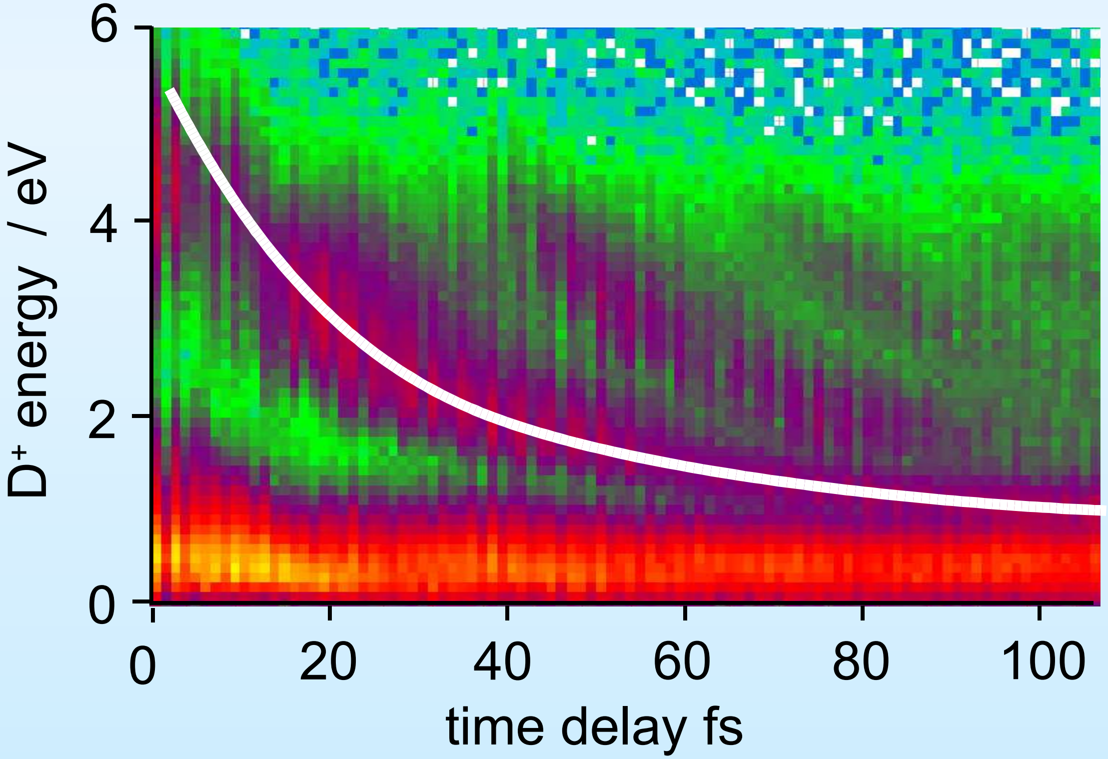


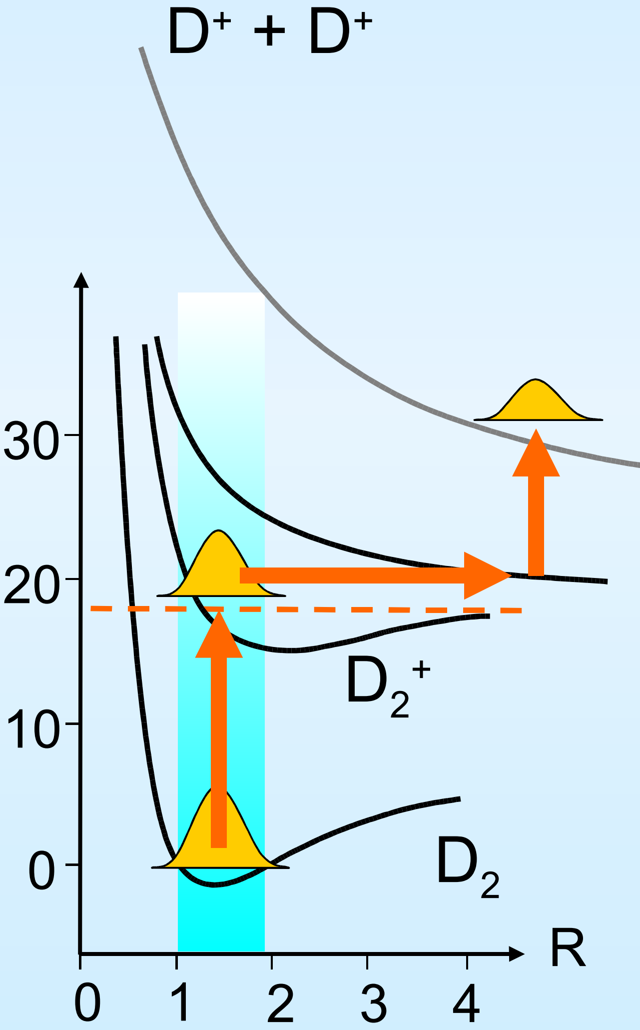
U. Becker, R. Dörner

"Snapshots" of the time-evolution of intra-molecular potentials

"Movie" of the dissociation reaction

8 fs Pump-Probe with D_2





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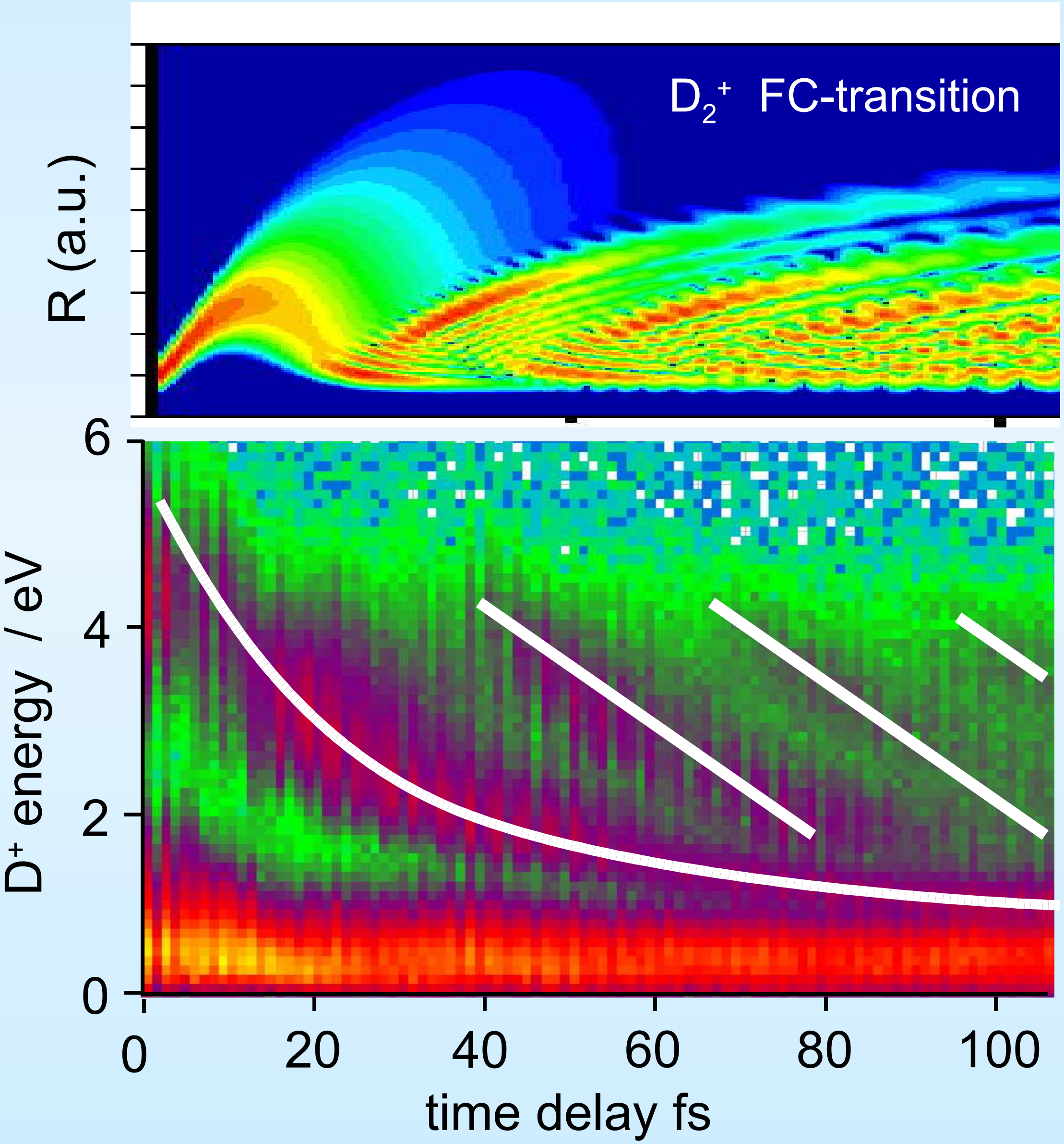
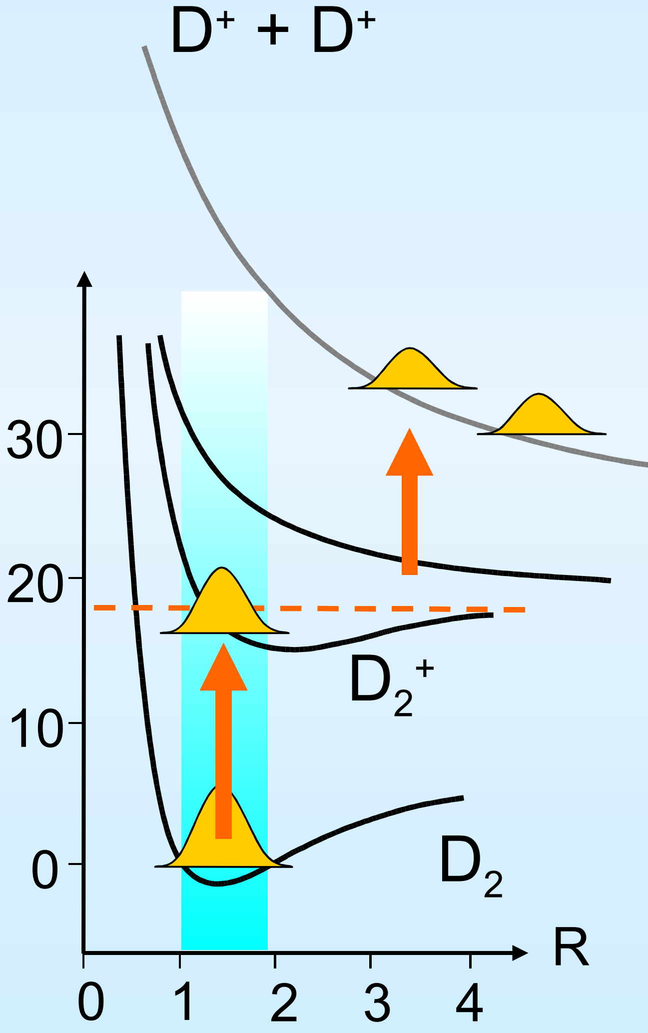
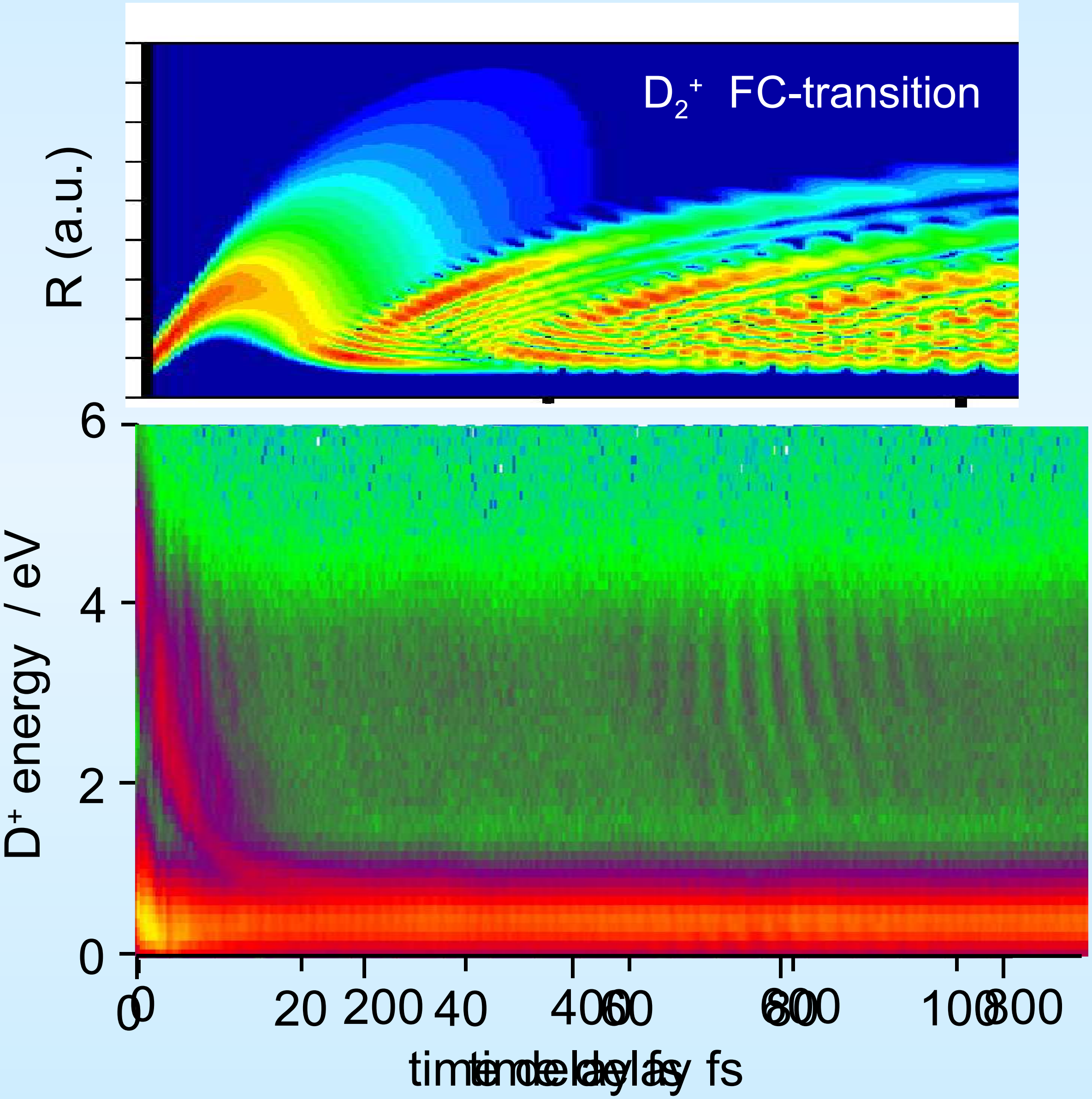
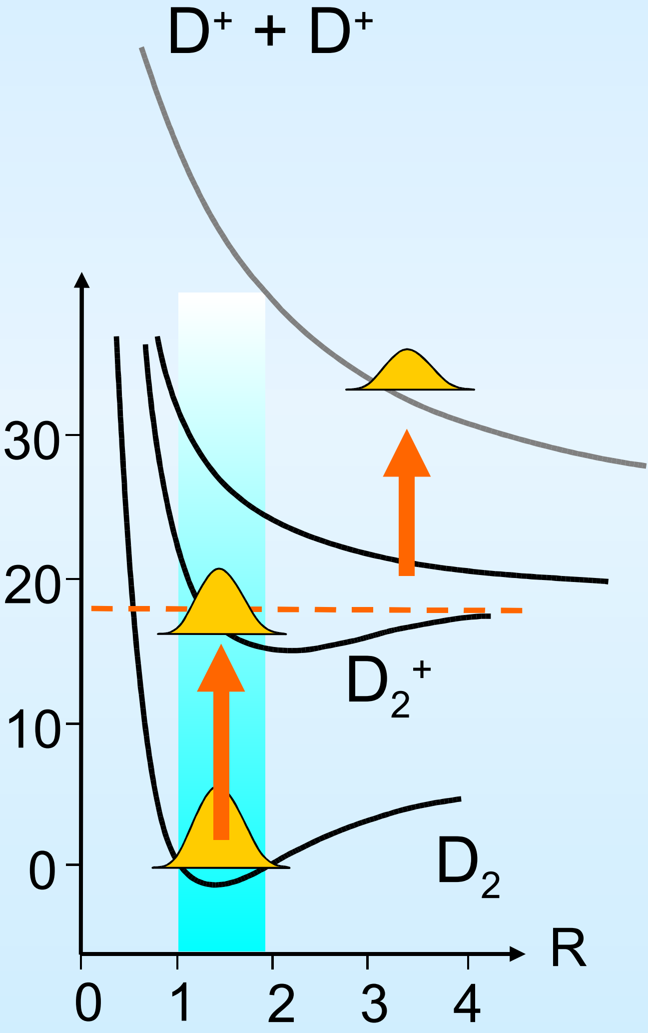




8 fs Pump-Probe with D_2

Max-Planck-Institut für Kernphysik





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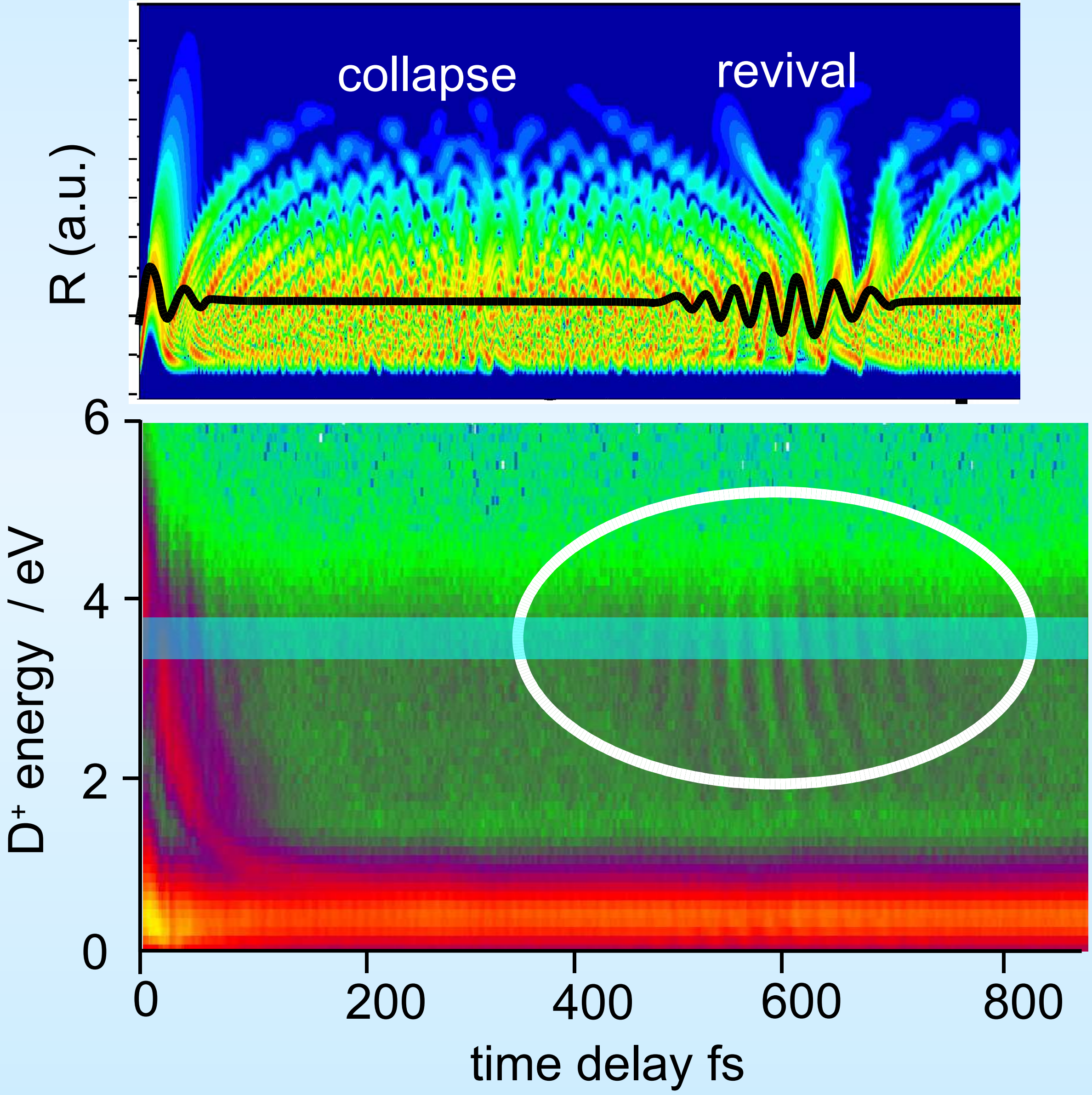

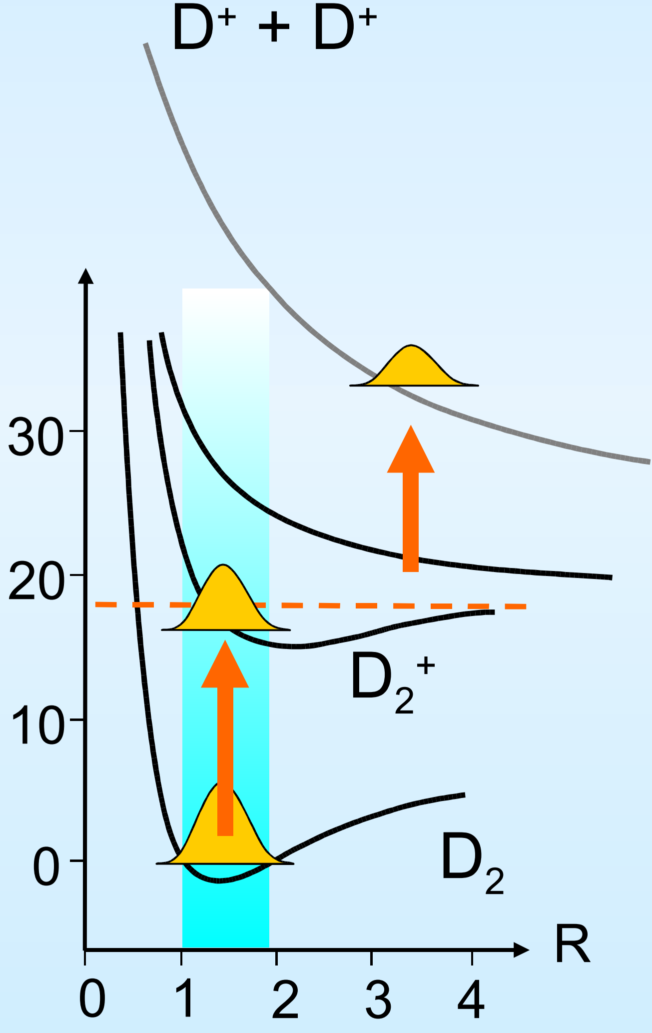

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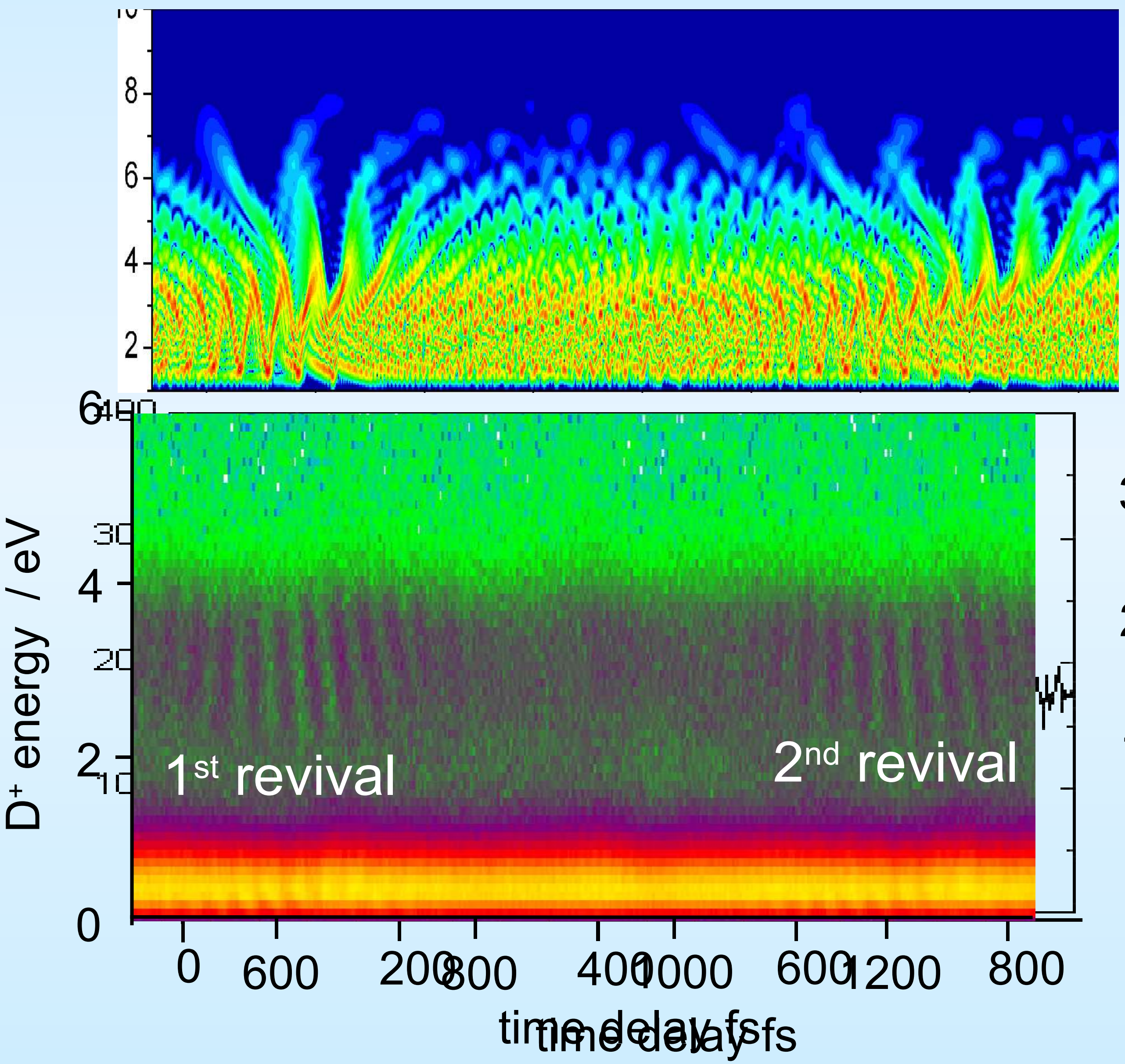

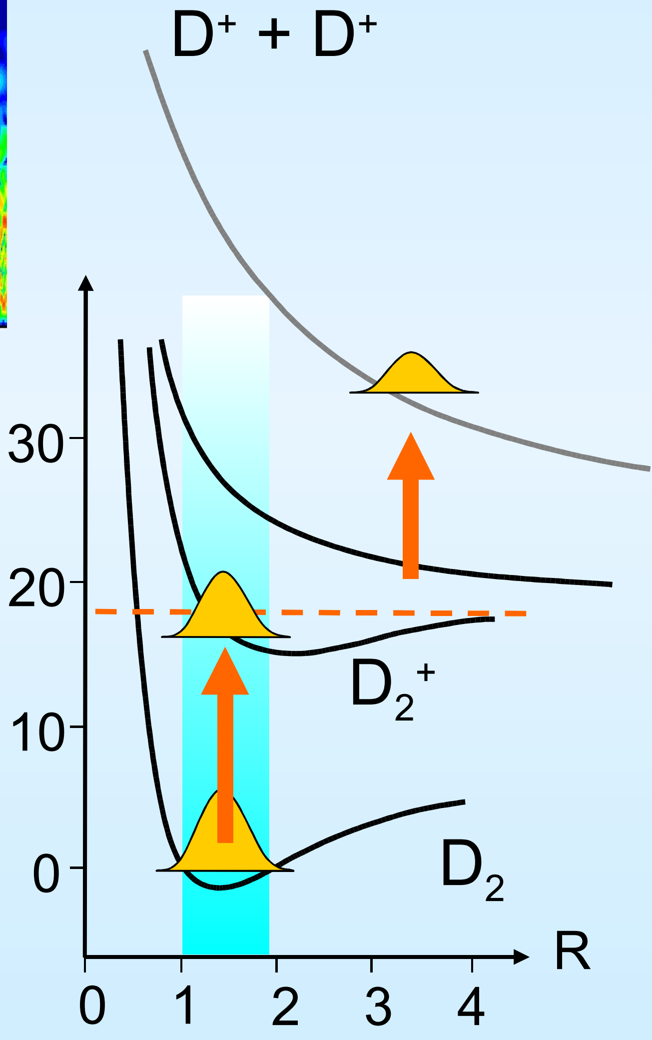

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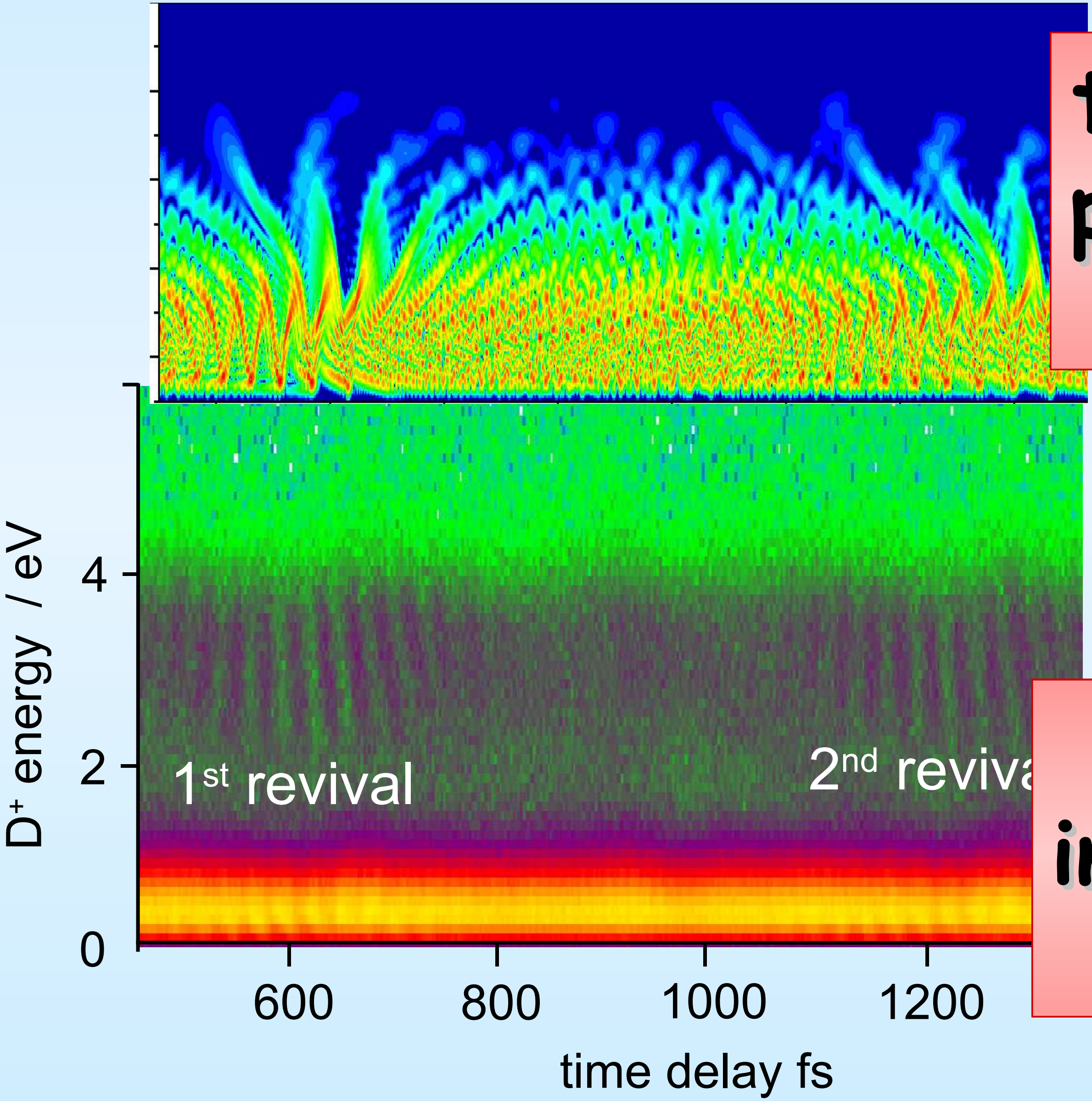

time resolved photo electron spectroscopy

follow the intra-molecular potentials !

End