

# Sampling electron dynamics in atoms in real time with sub-femtosecond resolution

Matthias Uiberacker  
Prof. Ferenc Krausz



Max-Planck-Institut  
für Quantenoptik  
Garching, Germany

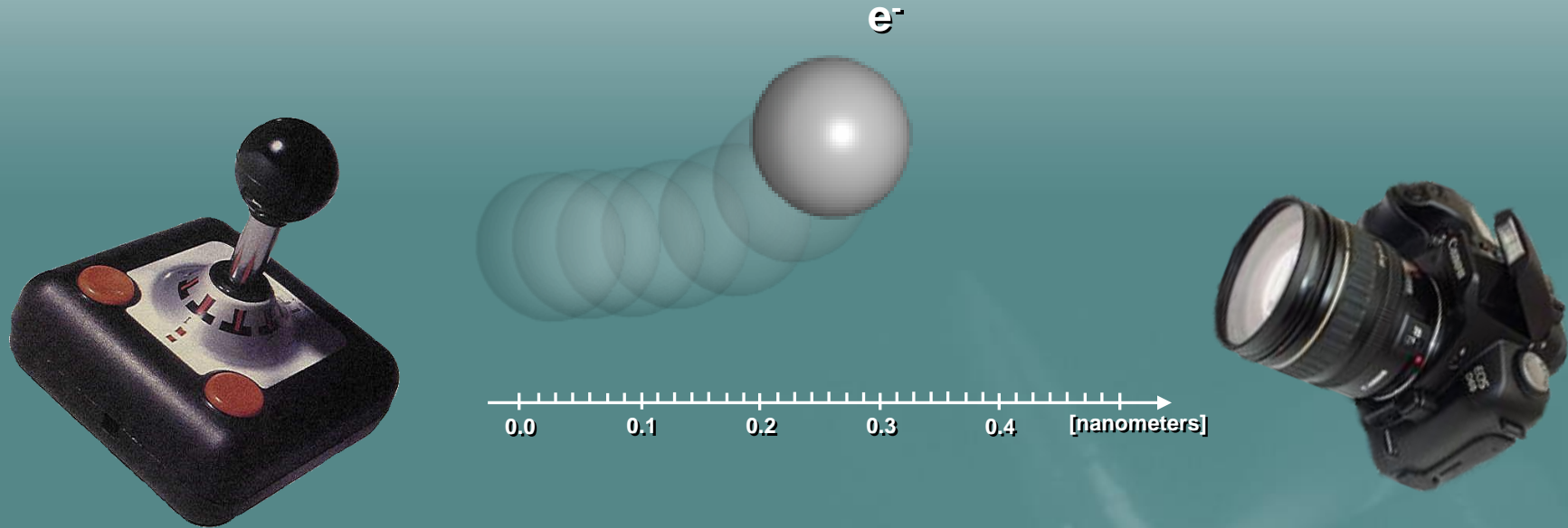


Dept. f. Physik, Ludwig-  
Maximilians-Universität  
München, Germany



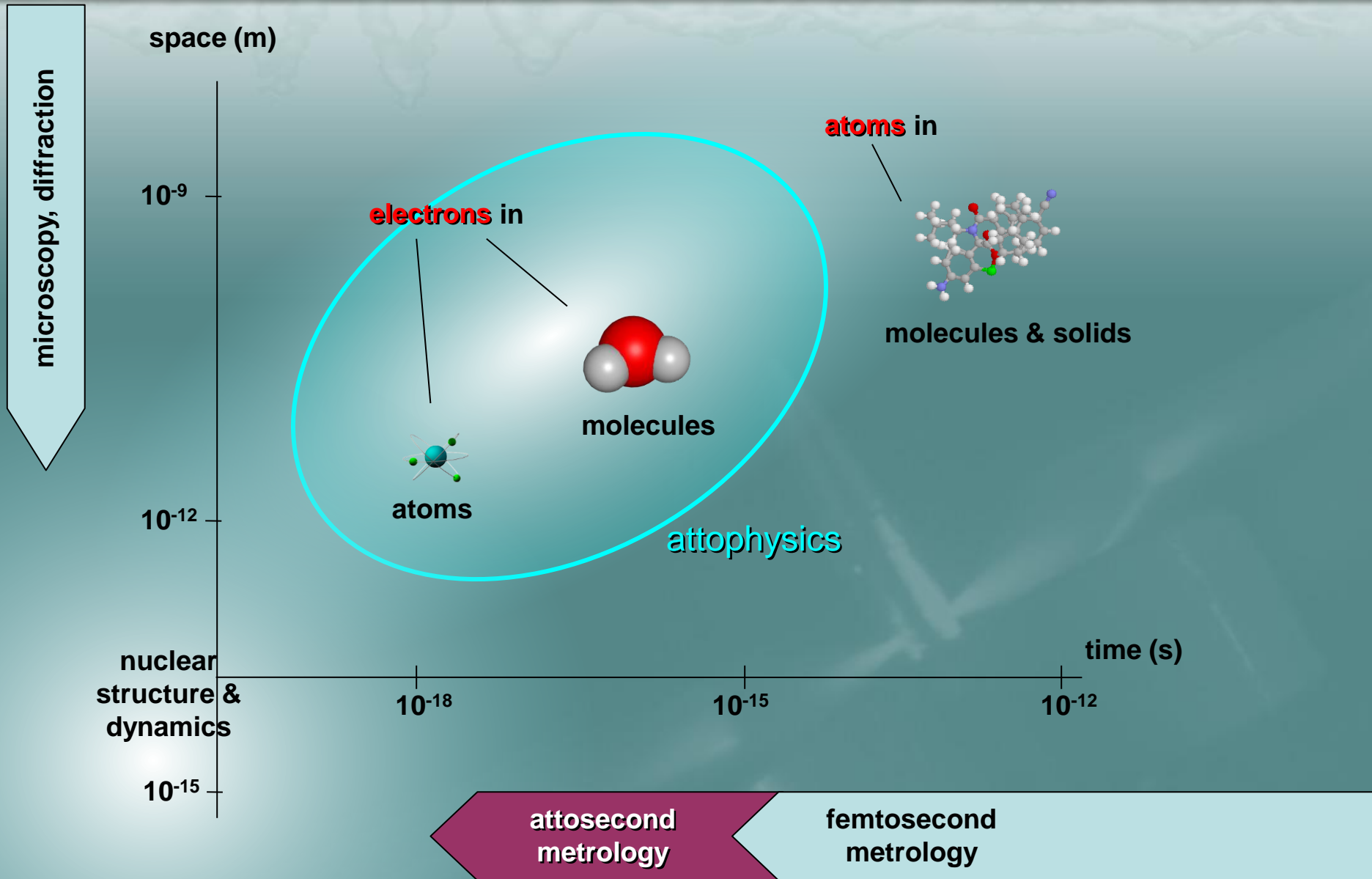
Institut für Photonik  
Technische Universität Wien  
Wien, Austria

# attosecond physics aims at gaining insight into the motion of electrons on atomic scales



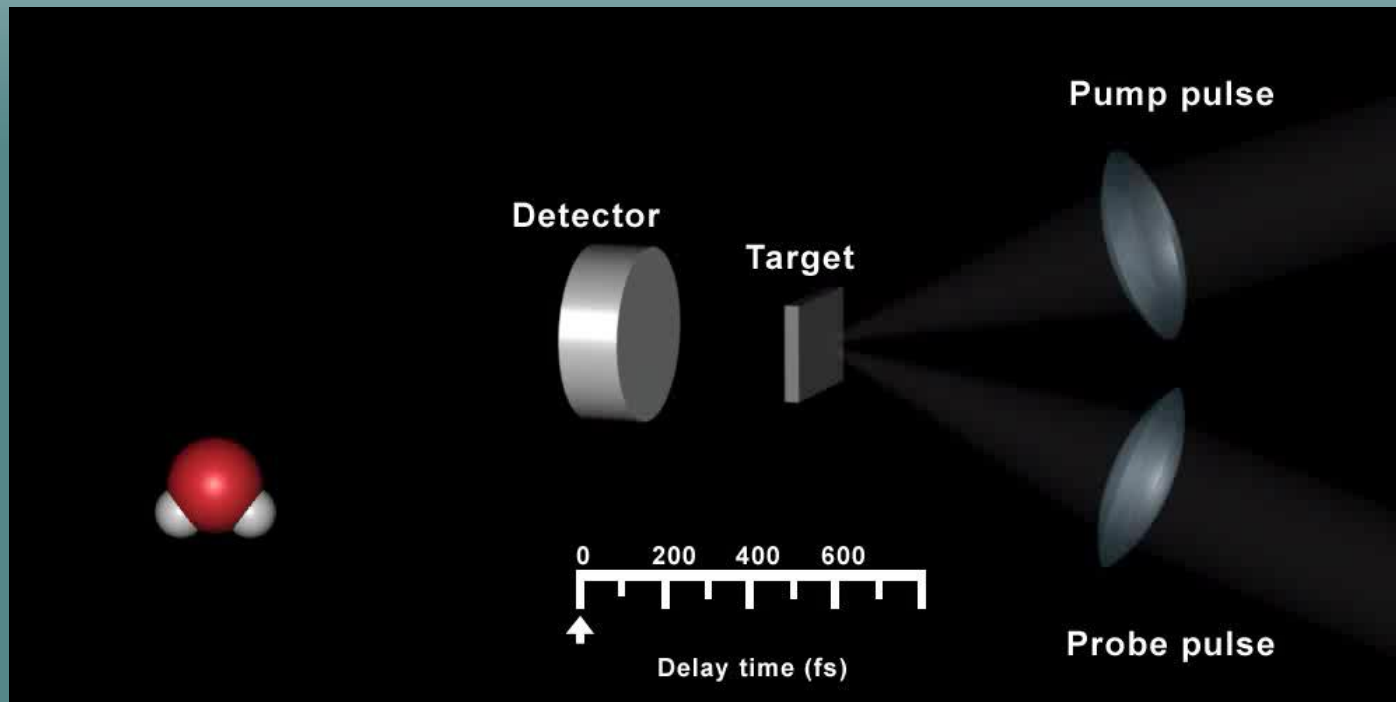
**direct control** & **real-time observation**  
of electronic motion in atoms, molecules, solids and plasmas

# the microcosm: imaging in space and time



# high-speed photography of microscopic processes:

## time-resolved pump-probe spectroscopy

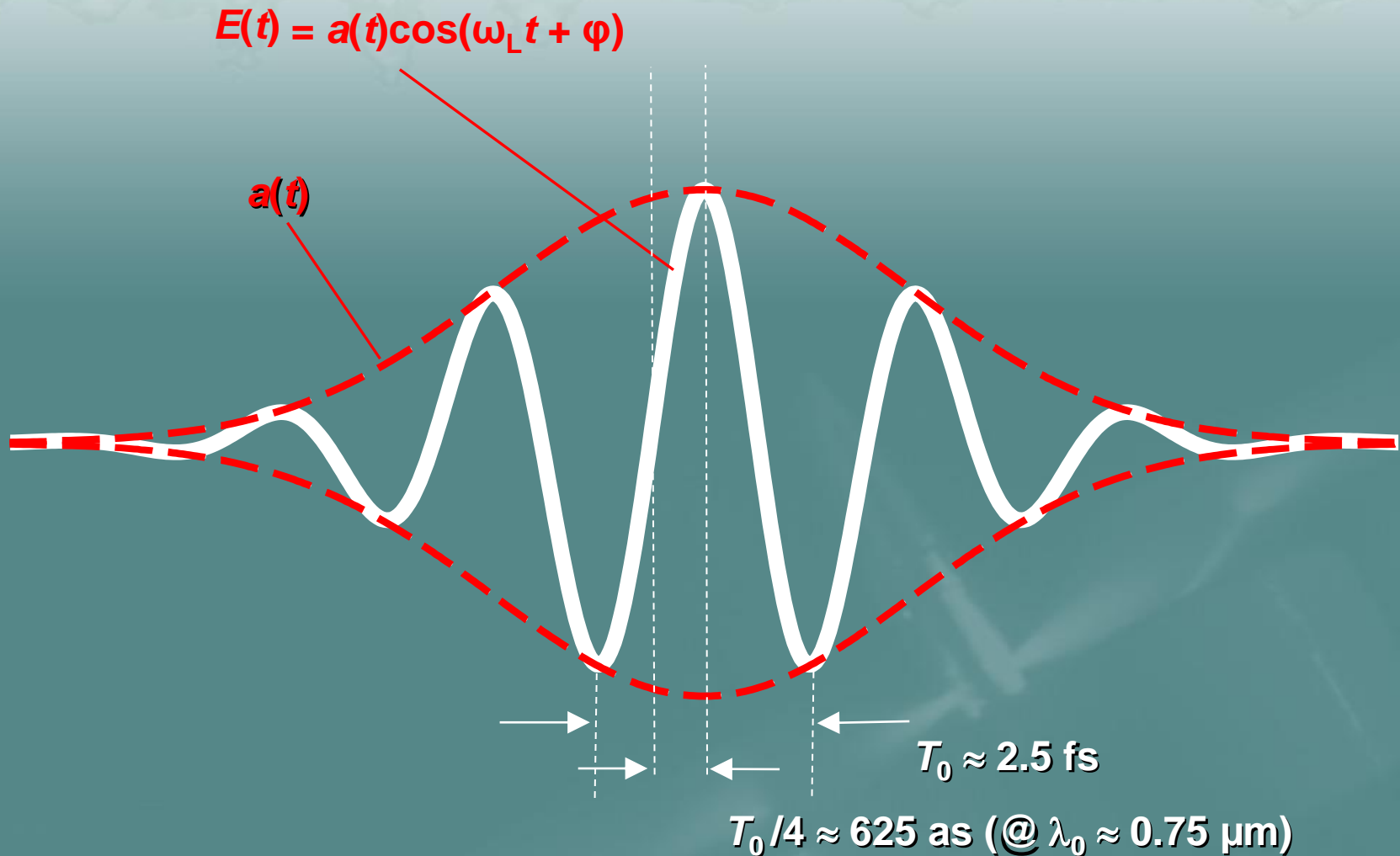


# a sampling system with sub-fs resolution

- utilizing pump/probe techniques
- pump pulse and probe pulse need to be short enough to freeze the motion of electrons
- ultrashort visible laser pulses are close to the wavecycle-limit of pulse duration (1-3fs).
- ..but, can be used to produce shorter (sub-fs) xuv pulses (high harmonic generation)
- efficiency for 2 sub-fs xuv pulses is not sufficient yet
- what to do?



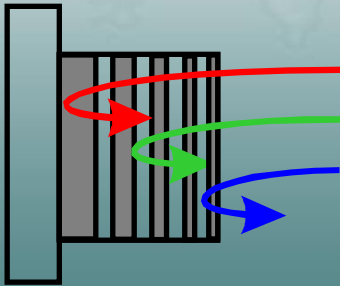
# using the electric field of laser pulses for probing with sub-fs resolution



- requires stabilization and control of carrier-envelope phase
- in combination with a weak sub-fs xuv pulse -> pump/probe measurements with sub-fs resolution!

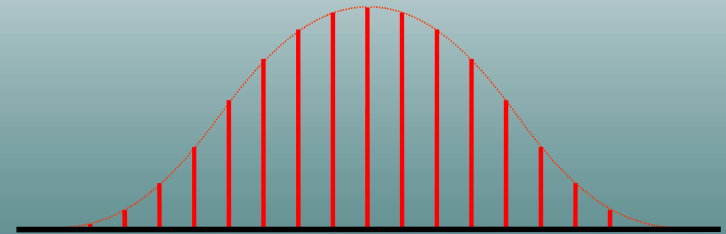
# waveform-controlled few-cycle light opens the door to steering & capturing electrons on an attosecond timescale

ultrabroadband dispersion  
control with chirped multilayers

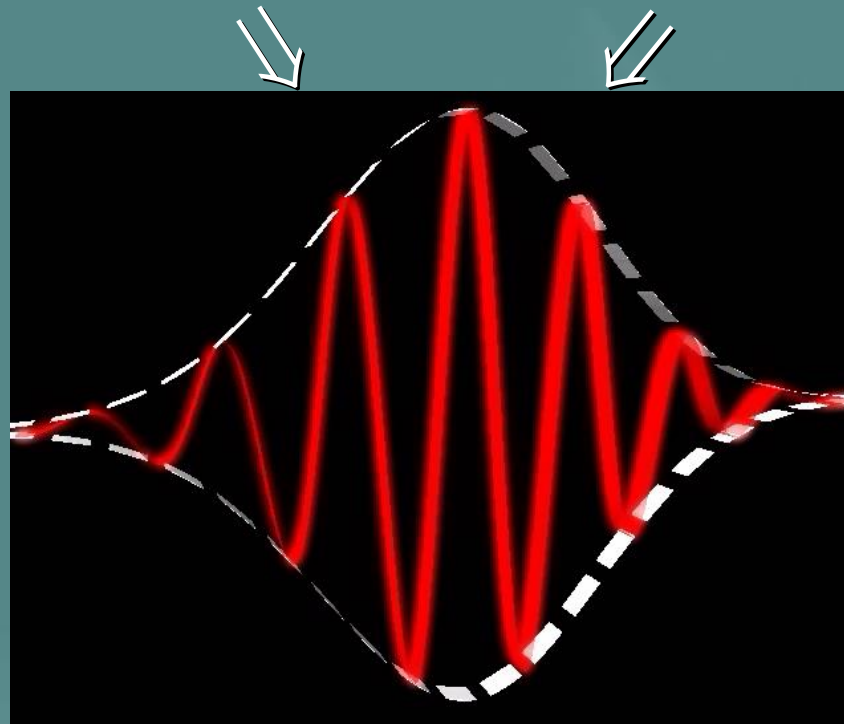


R. Szipöcs, K. Ferencz, Ch. Spielmann, F. Krausz  
*Opt. Lett.* **19**, 201 (1994)

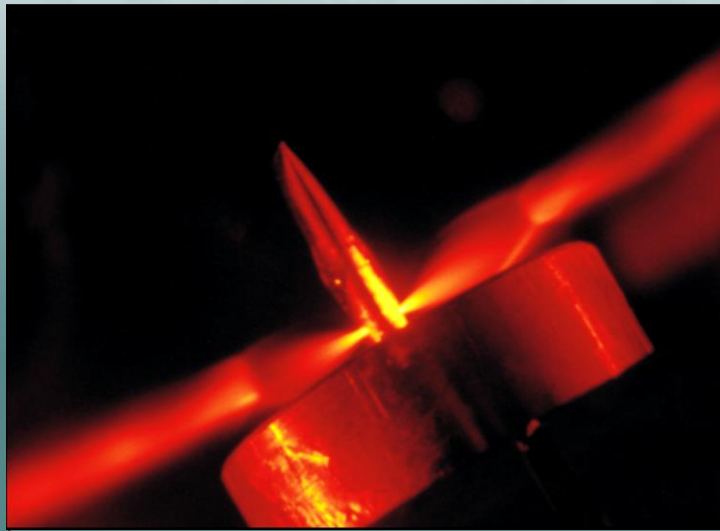
stabilization of the  
frequency comb of a mode-locked laser



T. W. Hänsch et al., 1997, 1999  
H. Telle et al. *Appl. Phys. B* **69**, 327 (1999)  
D. Jones et al., *Science* **288**, 635 (2000)



# xuv/x-ray radiation from strongly driven atoms



few-femtosecond,  
few-cycle  
laser pulse  
 $\lambda_L \approx 750 \text{ nm}$   
 $T_p = 5 - 6 \text{ fs}$   
 $W_p = 0.2 - 0.4 \text{ mJ}$

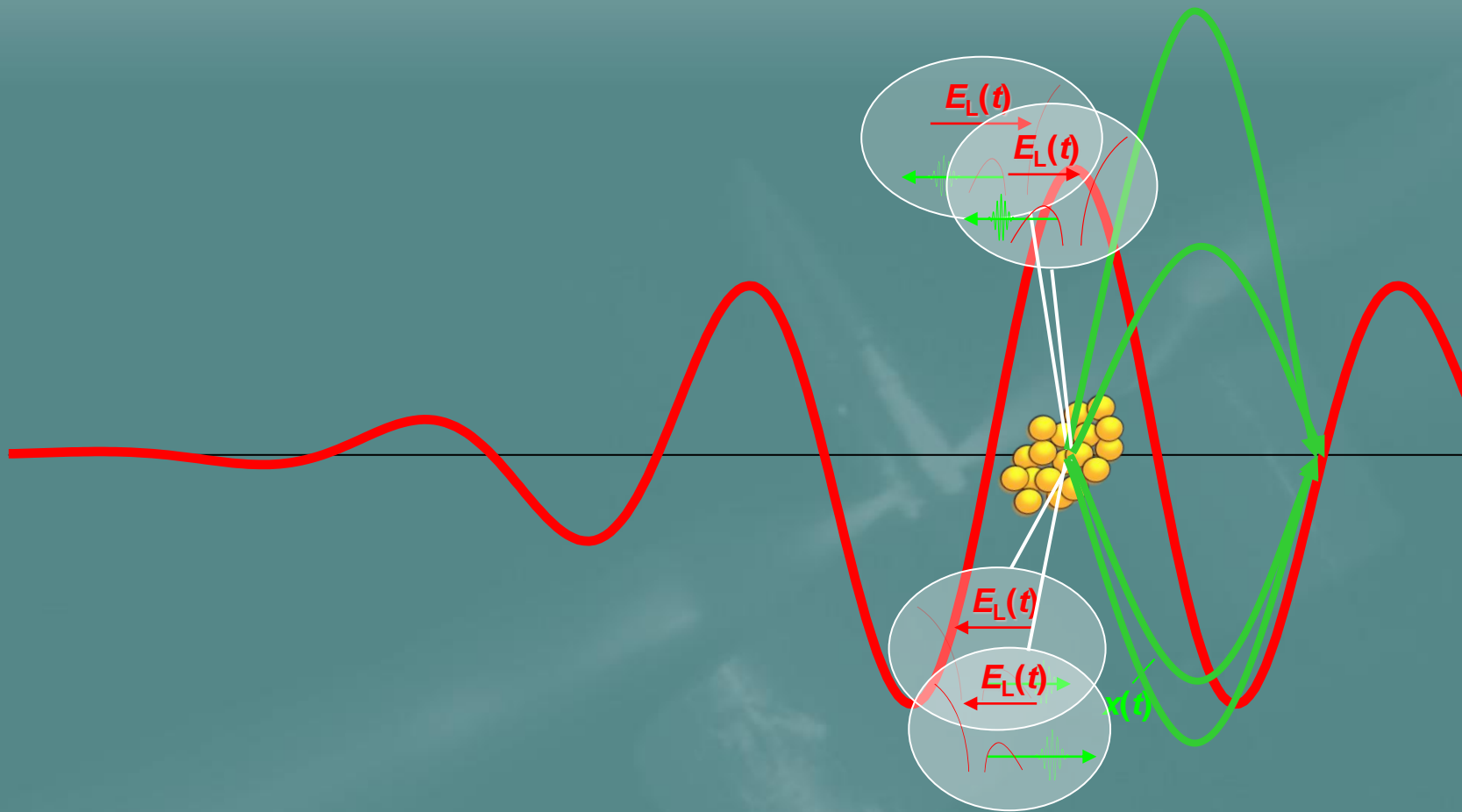
↑  
Ne gas

phase-stabilized  
electric field

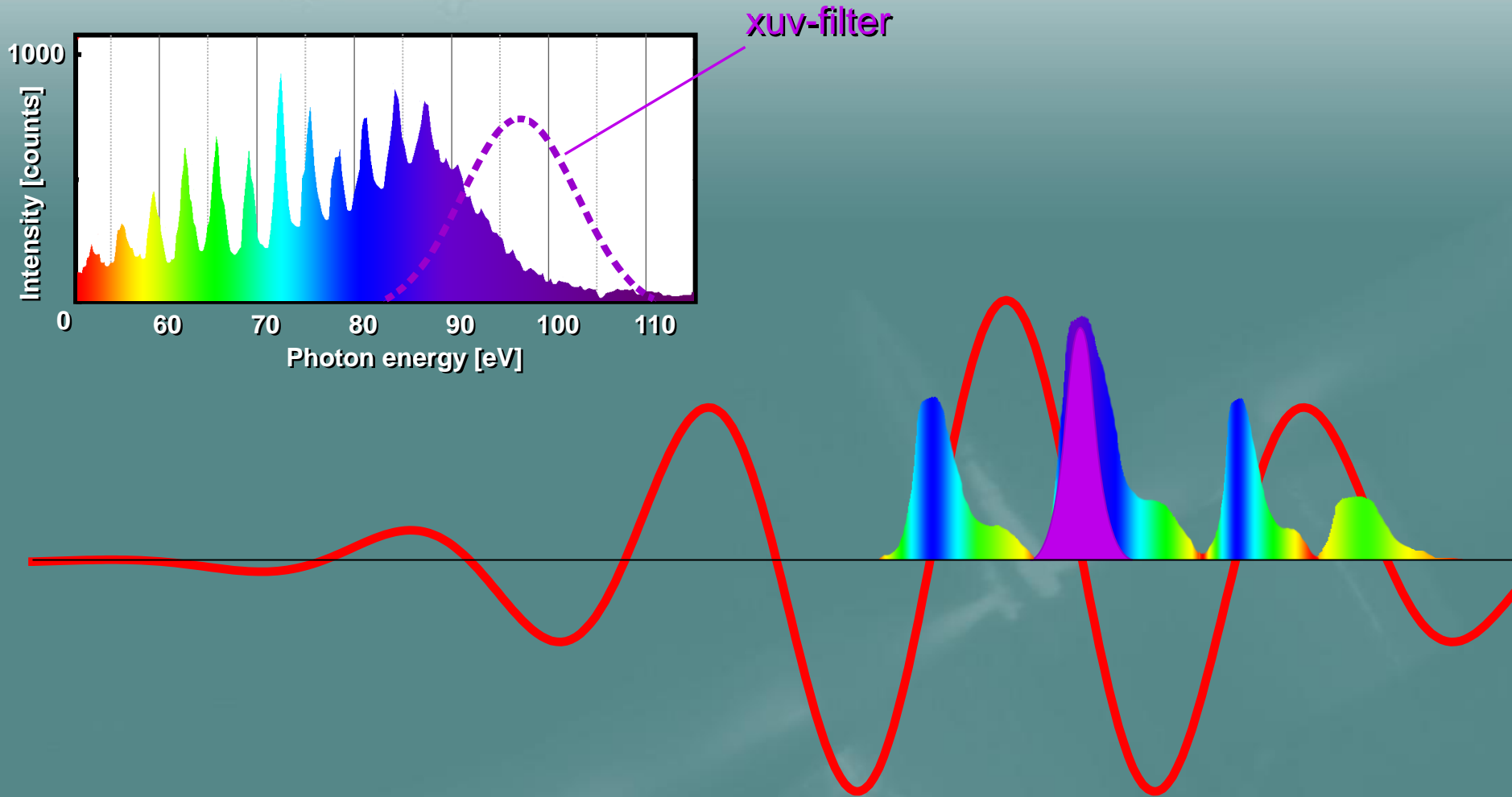
# steering bound electrons with controlled light fields: the generation of a sub-femtosecond pulse



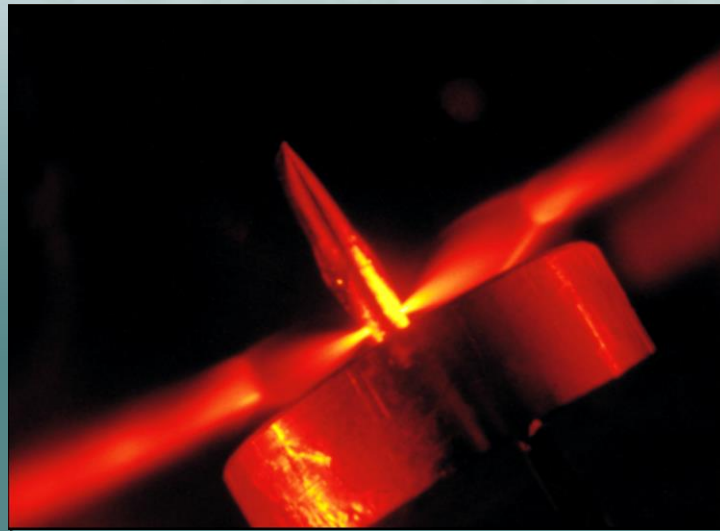
# xuv/x-ray radiation from strongly driven atoms



# sub-femtosecond xuv/x-ray pulse generation

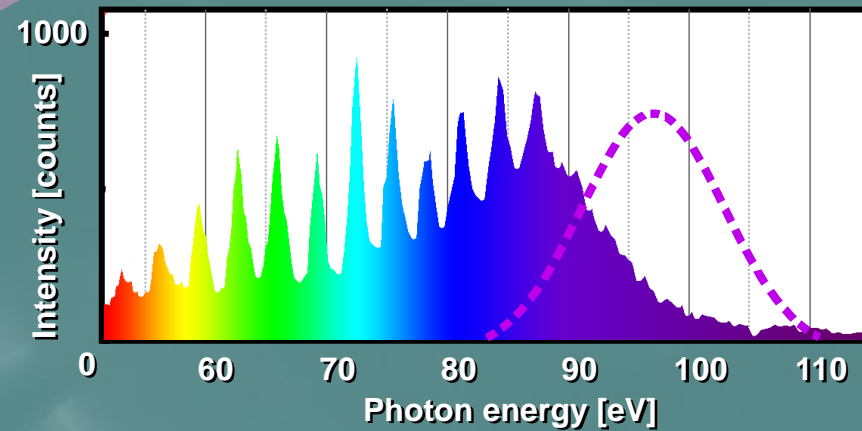
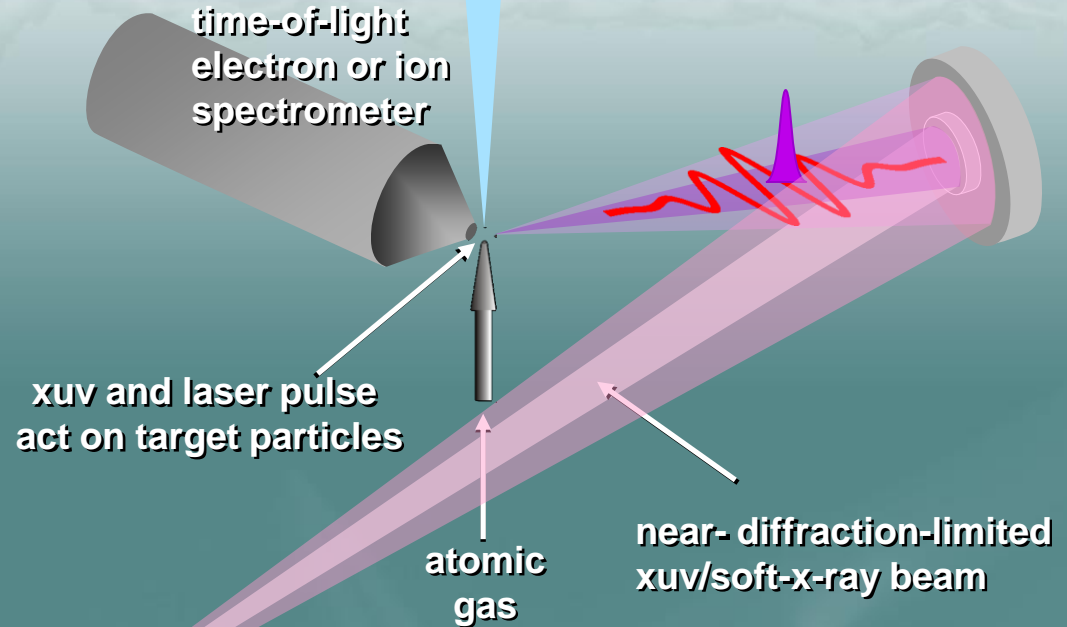


# attosecond pulse generation and detection



few-femtosecond,  
few-cycle  
laser pulse  
 $\lambda_L \approx 750 \text{ nm}$   
 $T_p = 5 - 7 \text{ fs}$   
 $W_p = 0.3 - 0.5 \text{ mJ}$

Ne gas



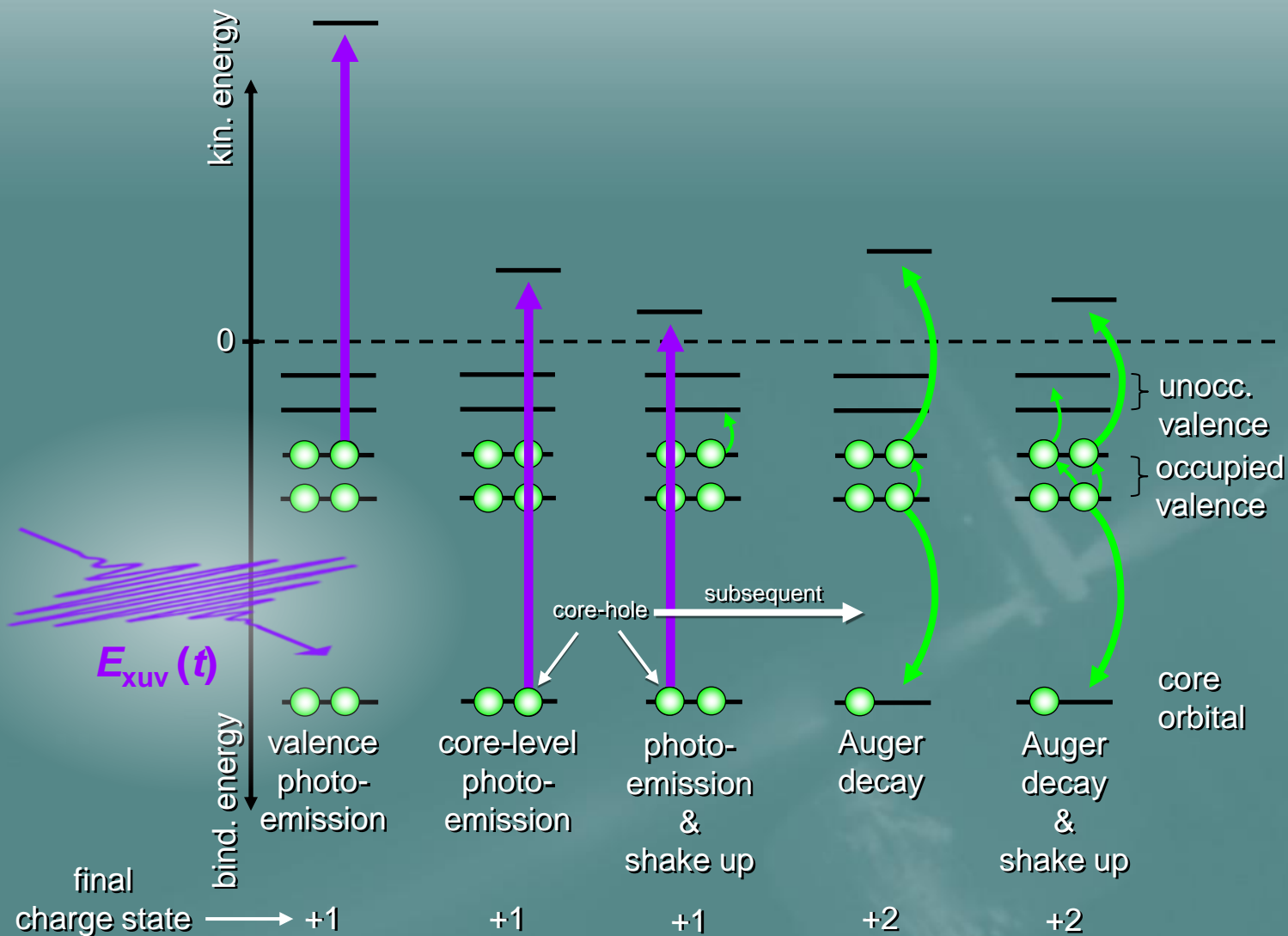
Drescher *et al.*, *Science* **291**, 1923 (2001)

Hentschel *et al.*, *Nature* **414**, 509 (2001)

Kienberger *et al.*, *Science* **297**, 1144 (2002)

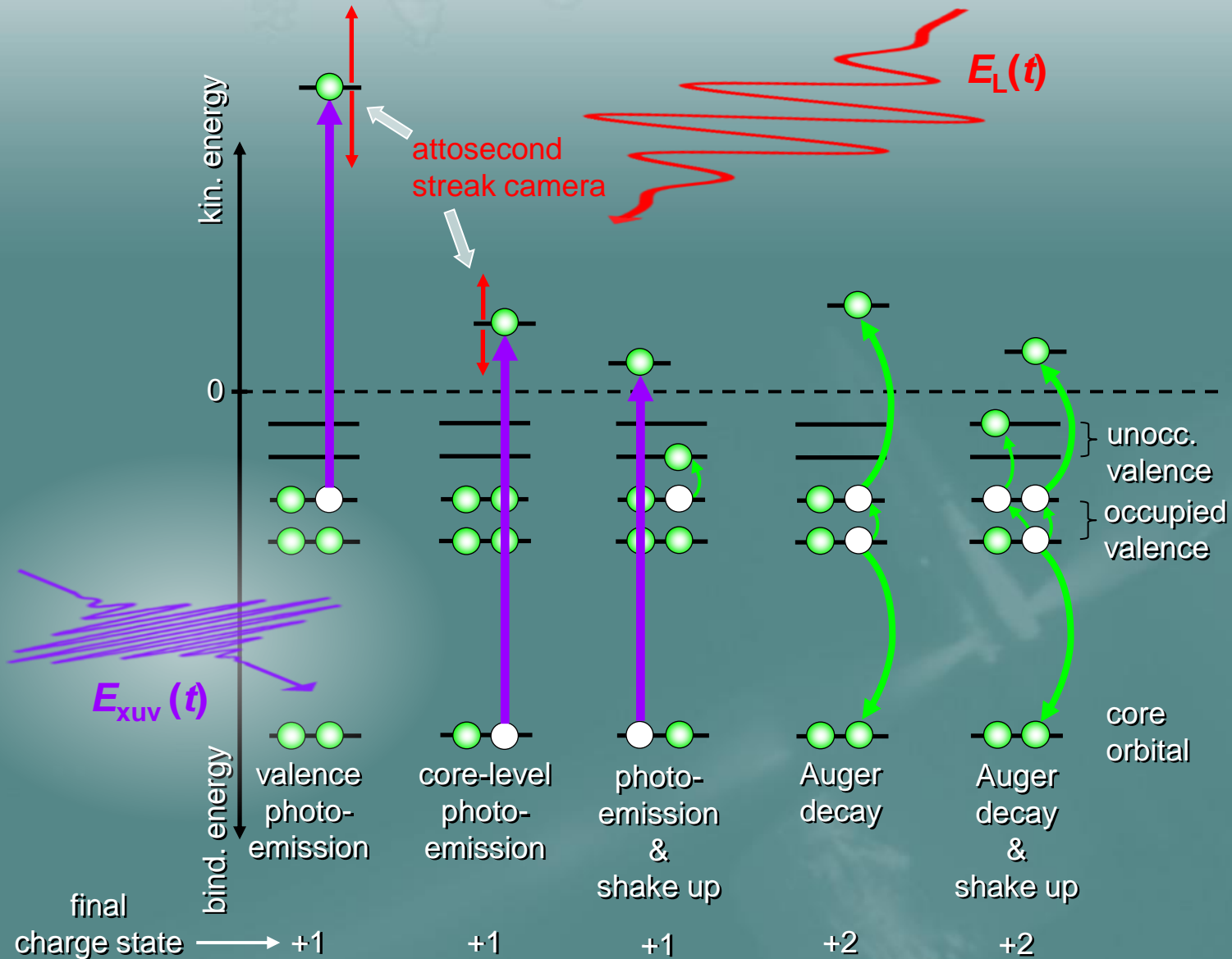
# triggering electronic transitions inside atoms

by irradiation with **xuv-light** pulses

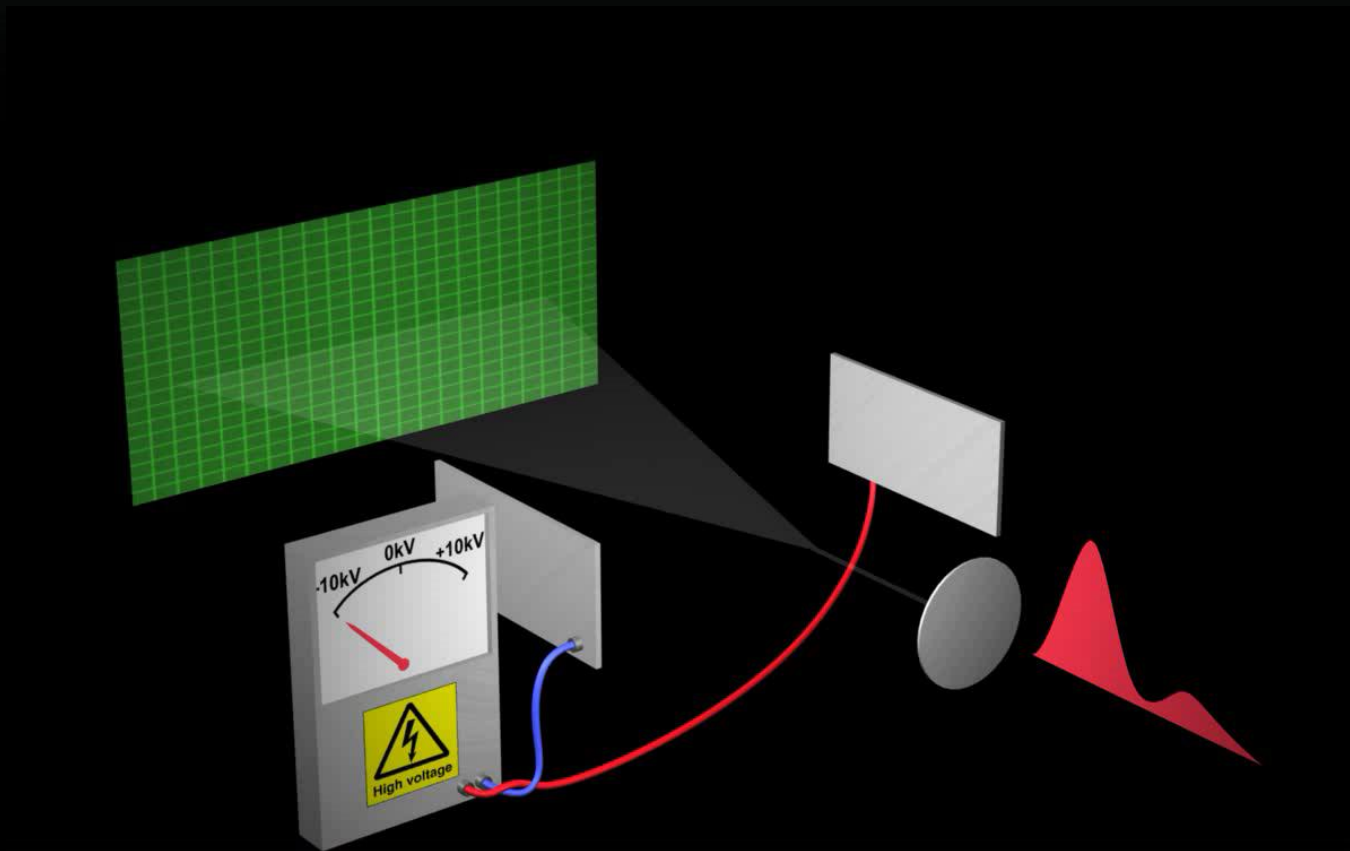


# probing electronic transitions inside atoms

by means of strong-field-induced free-free transitions: **streaking**

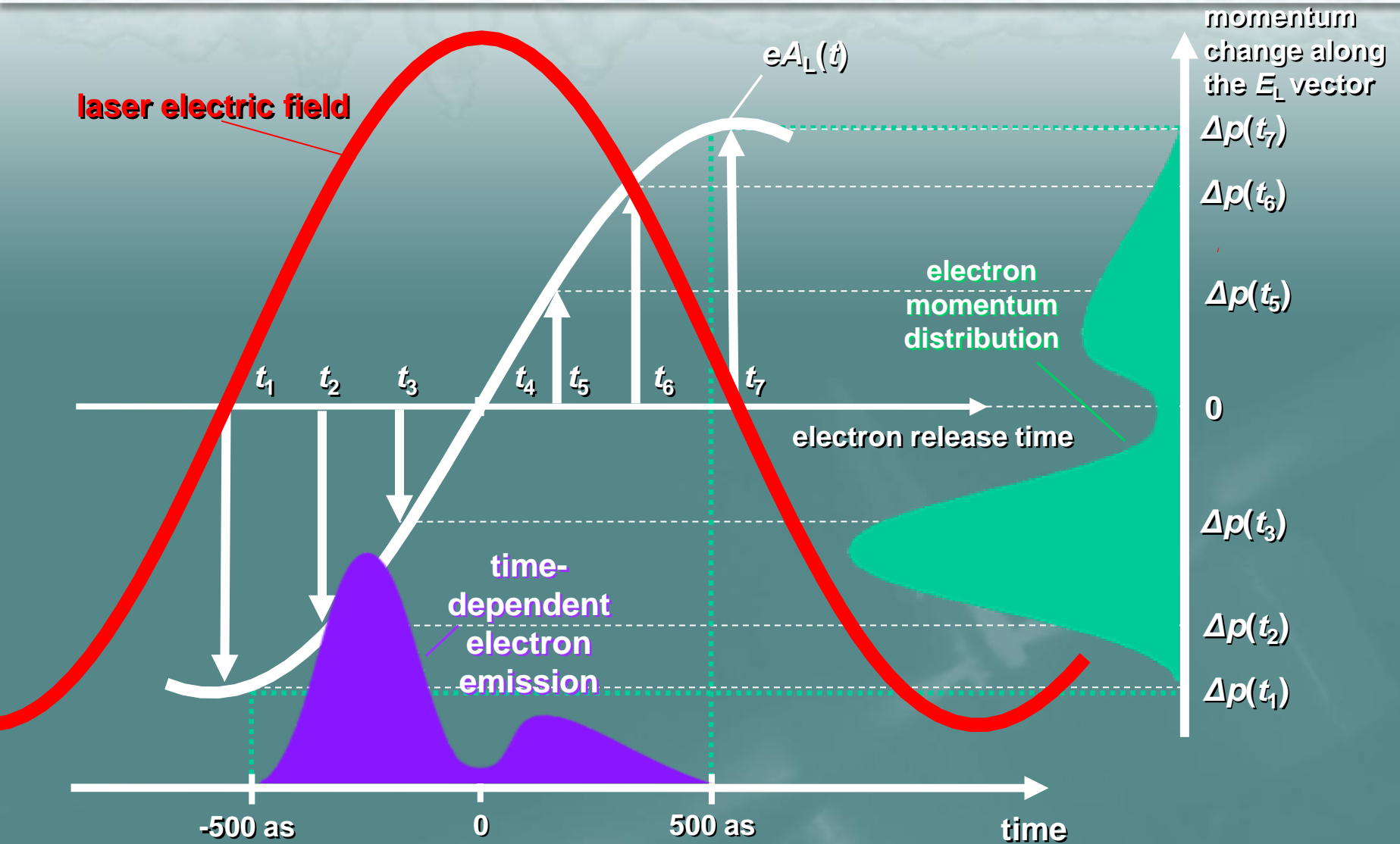


# electron-optical streak camera



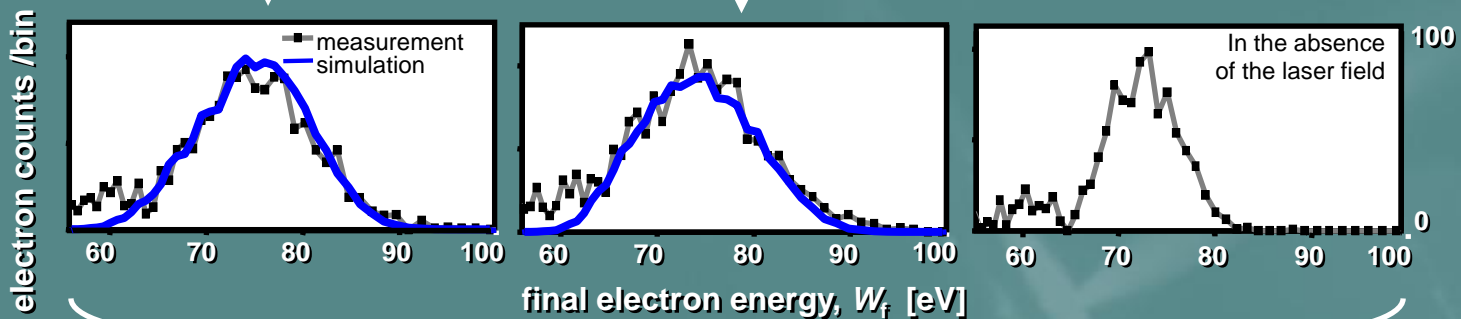
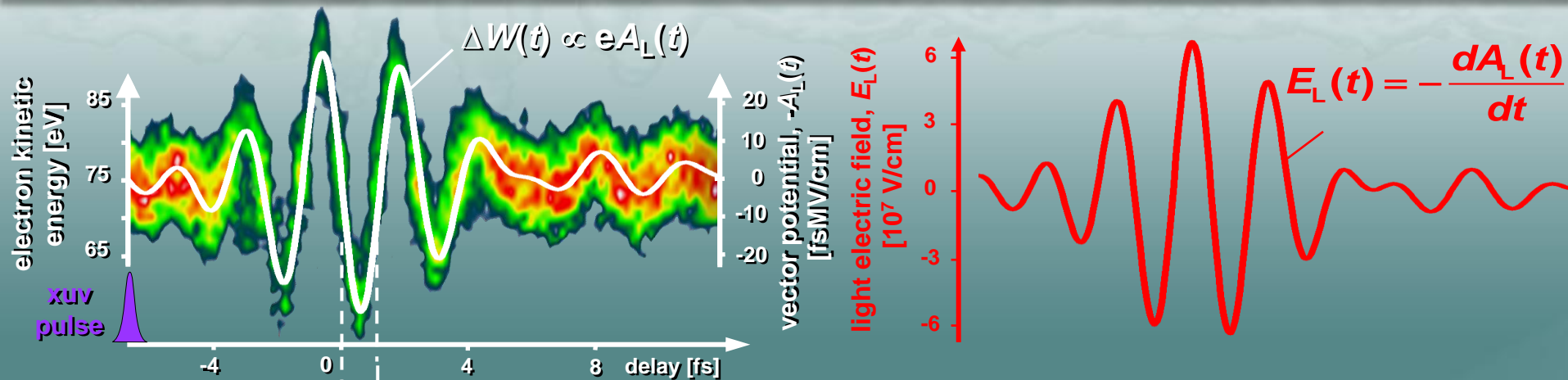
resolution ~ 100 femtoseconds

# mapping time to momentum

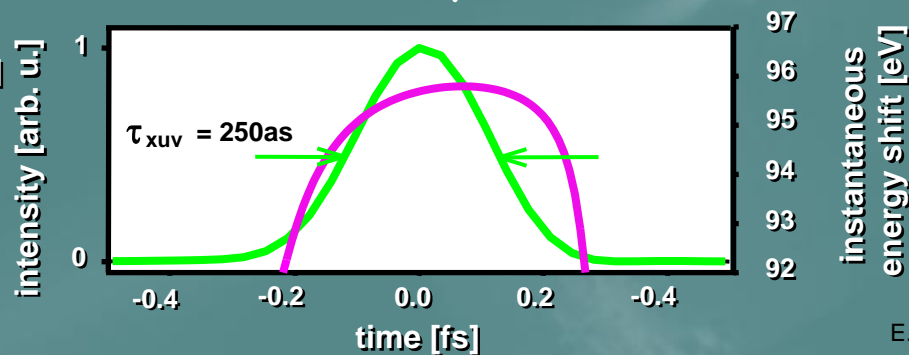


## optical-field-driven streak camera

# attosecond streak camera: complete measurement of a few-cycle light wave *and* a sub-fs xuv pulse

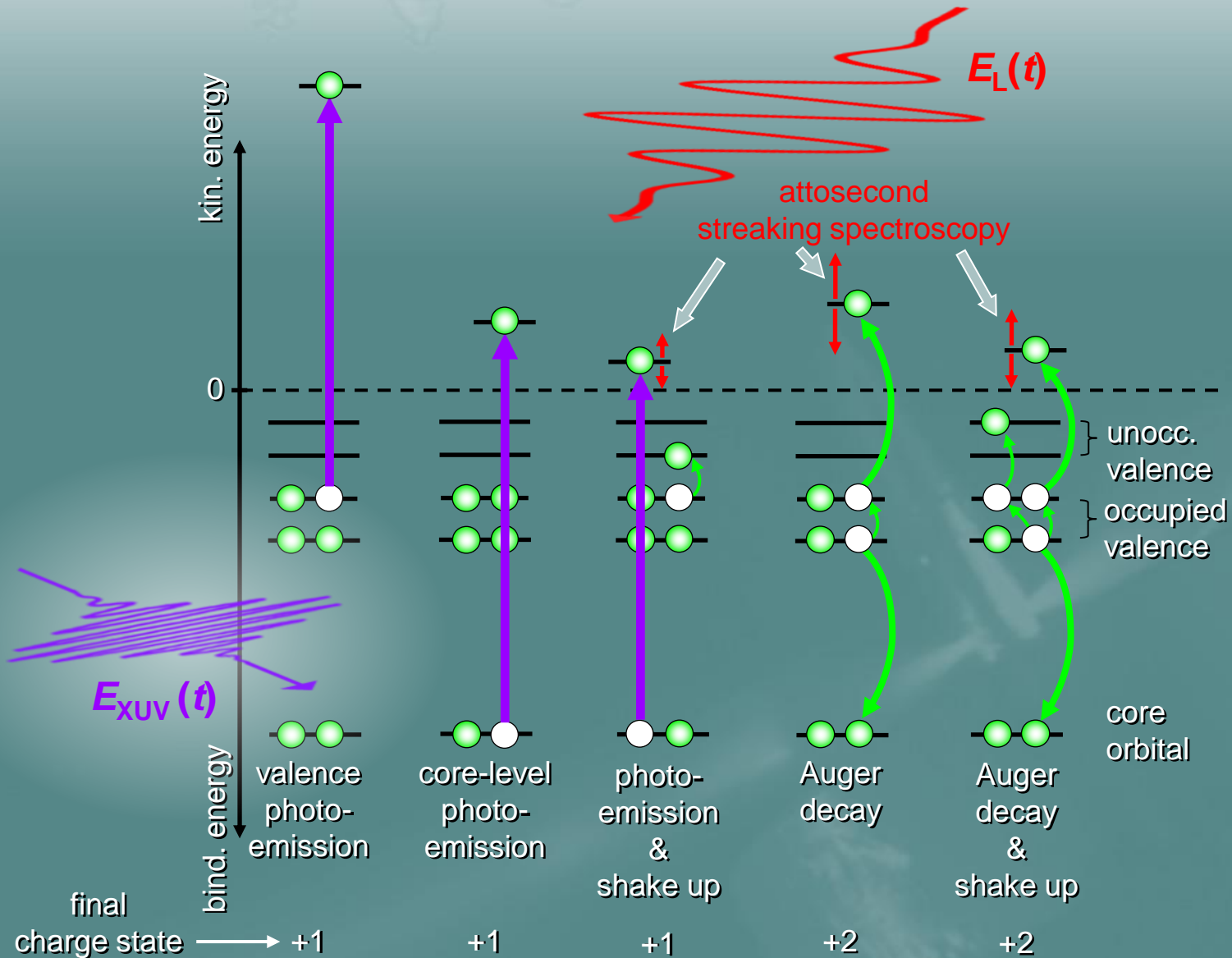


single 250-attosecond xuv pulse @ 95 eV

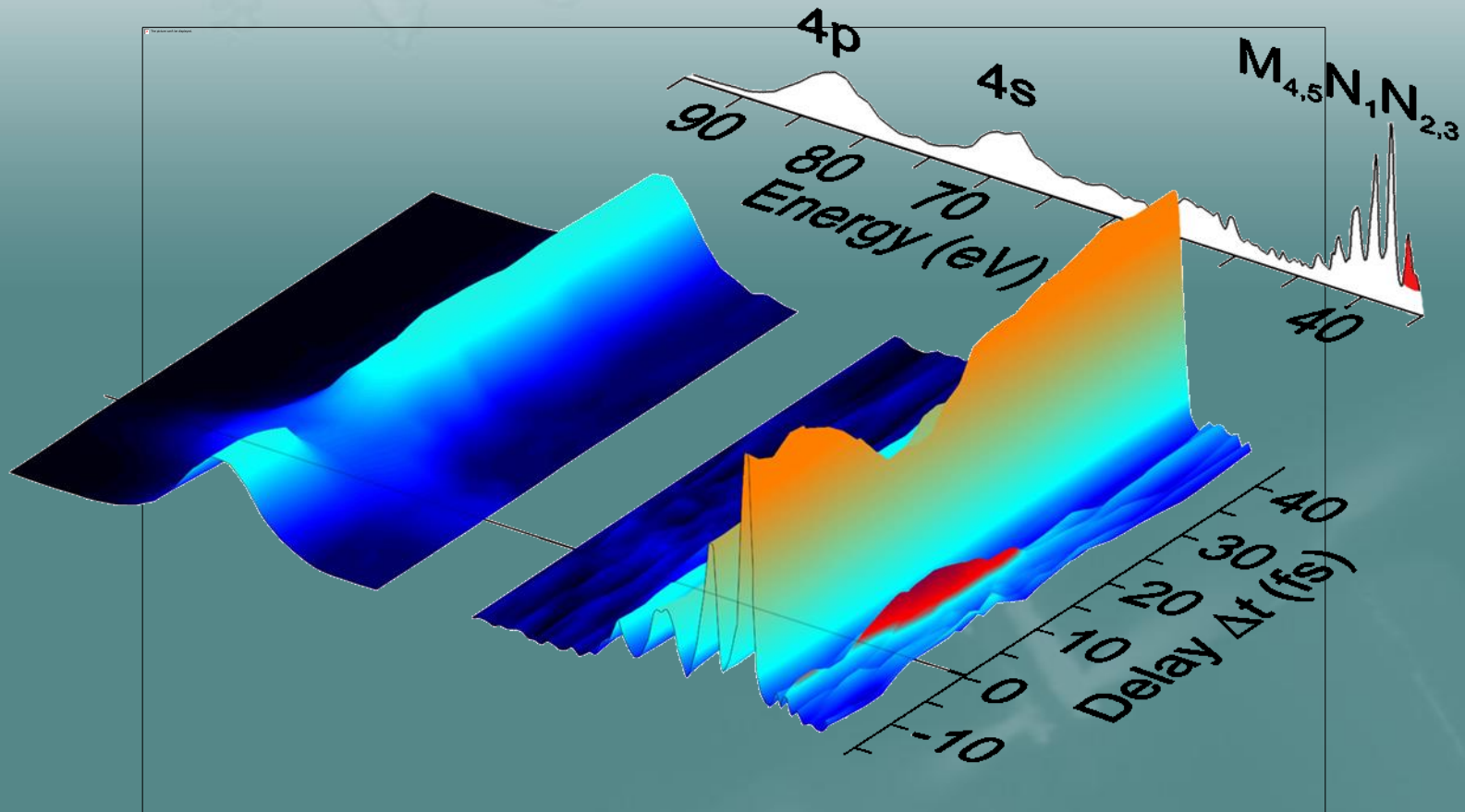


# probing electronic transitions inside atoms

by means of strong-field-induced free-free transitions: **streaking**



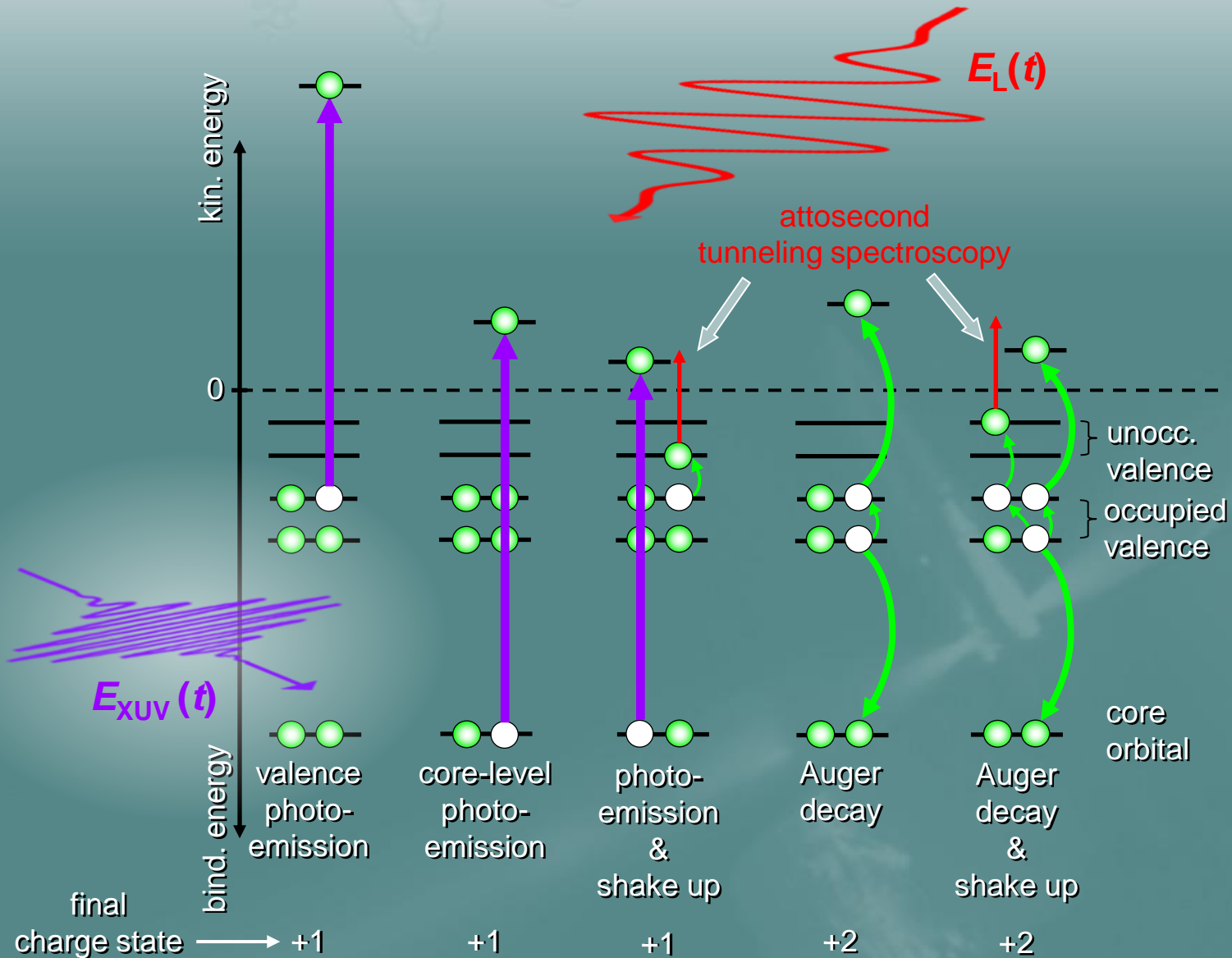
# streaked electron spectra following core-hole excitation in krypton by a sub-fs xuv pulse



tracing core-hole decay directly in time  
lifetime of M-shell (3d) vacancy in Krypton:  $\tau_h = 7.9 \pm 1$  fs

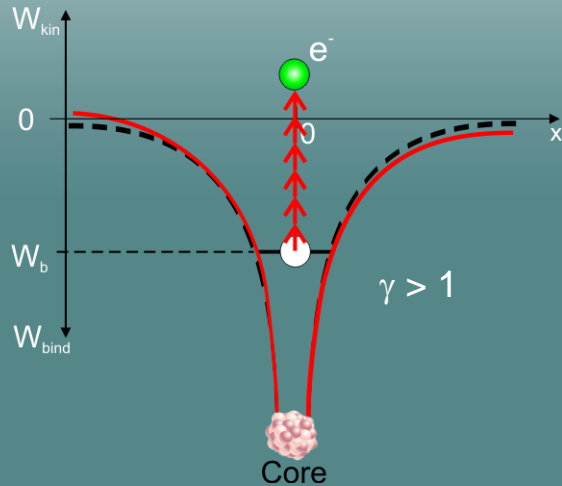
# probing electronic transitions inside atoms

by means of strong-field-induced bound-free transitions: **tunneling**



# multiphoton versus tunneling ionization: the Keldysh theory

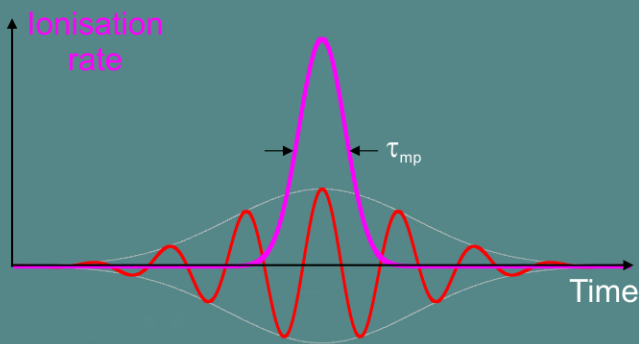
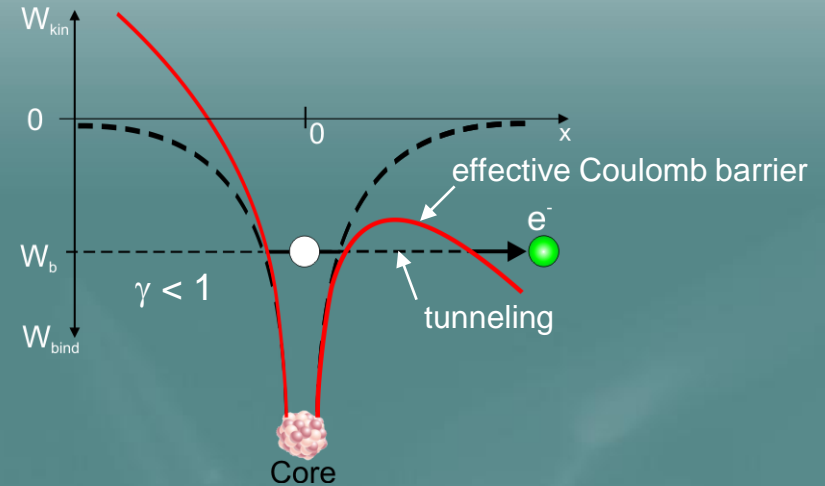
multiphoton ionization:  
due to absorption of many photons



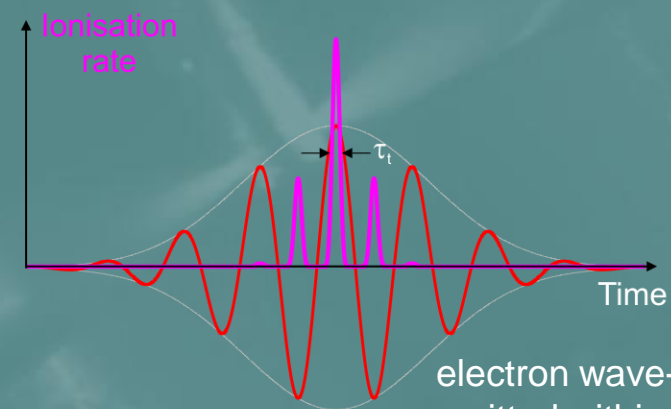
Keldysh parameter:  

$$\gamma = \frac{\omega_L \sqrt{2mW_b}}{eE_L}$$

tunneling ionization:  
due to suppression of Coulomb potential

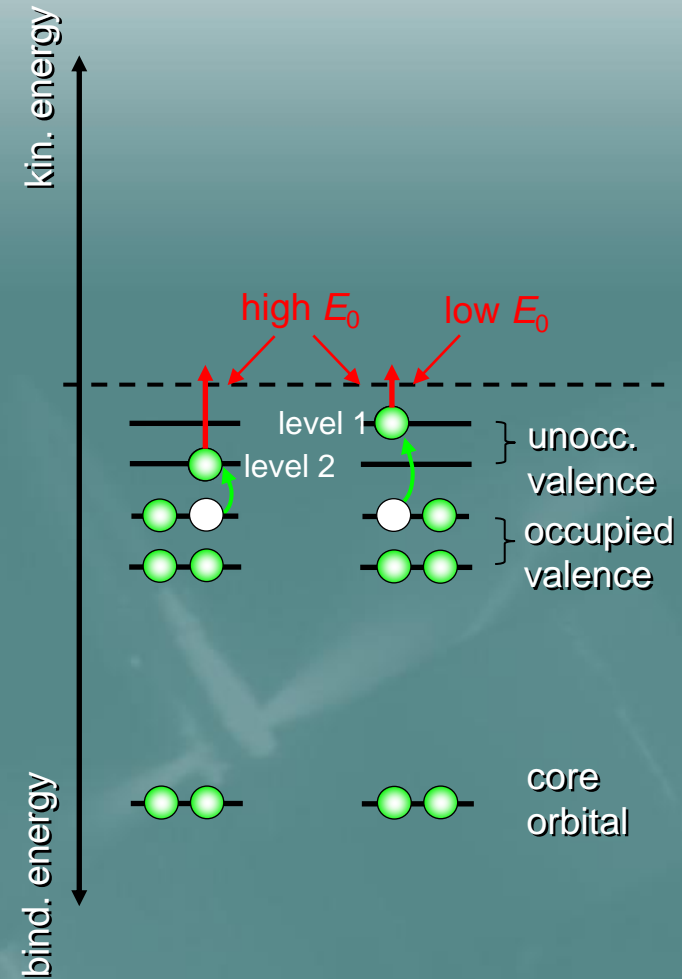
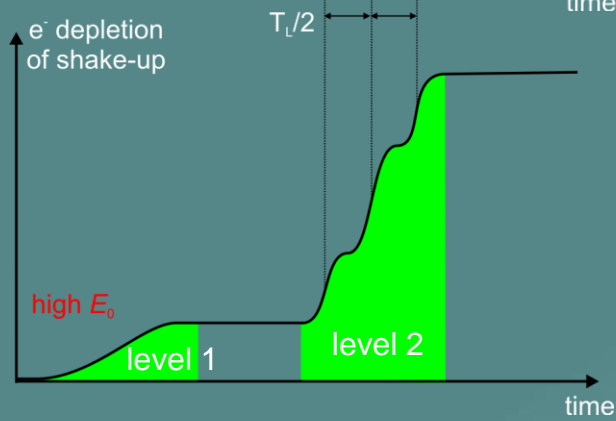
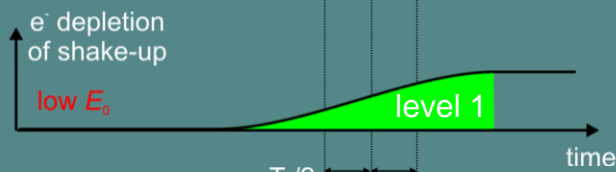
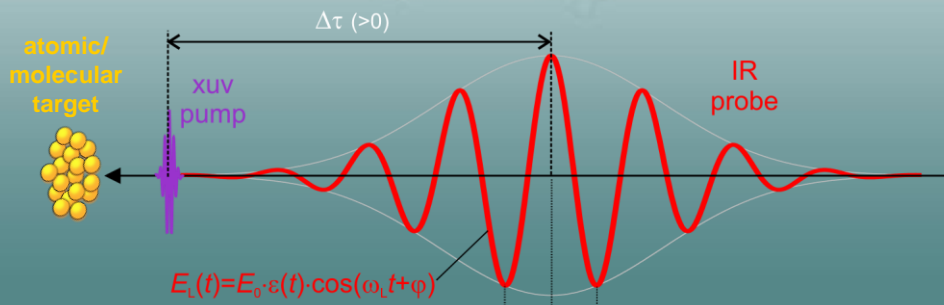


electron emission  
within a time  $\tau_{mp}$  shorter  
than the pulse duration



electron wave-packets  
emitted within a time  $\tau_t$   
shorter than the half-period  
of the laser

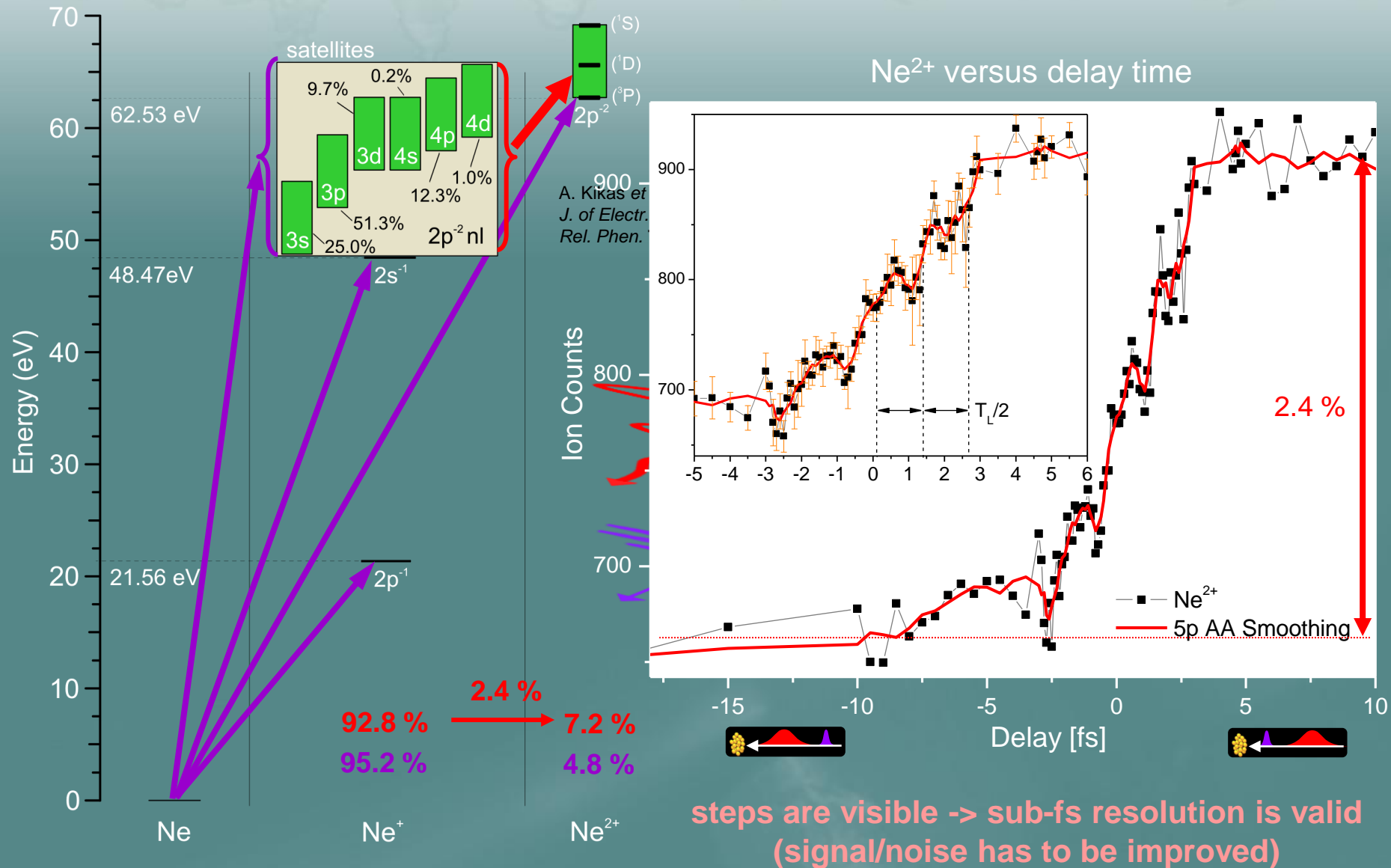
# time evolution of probing – ionization with a few-cycle pulse



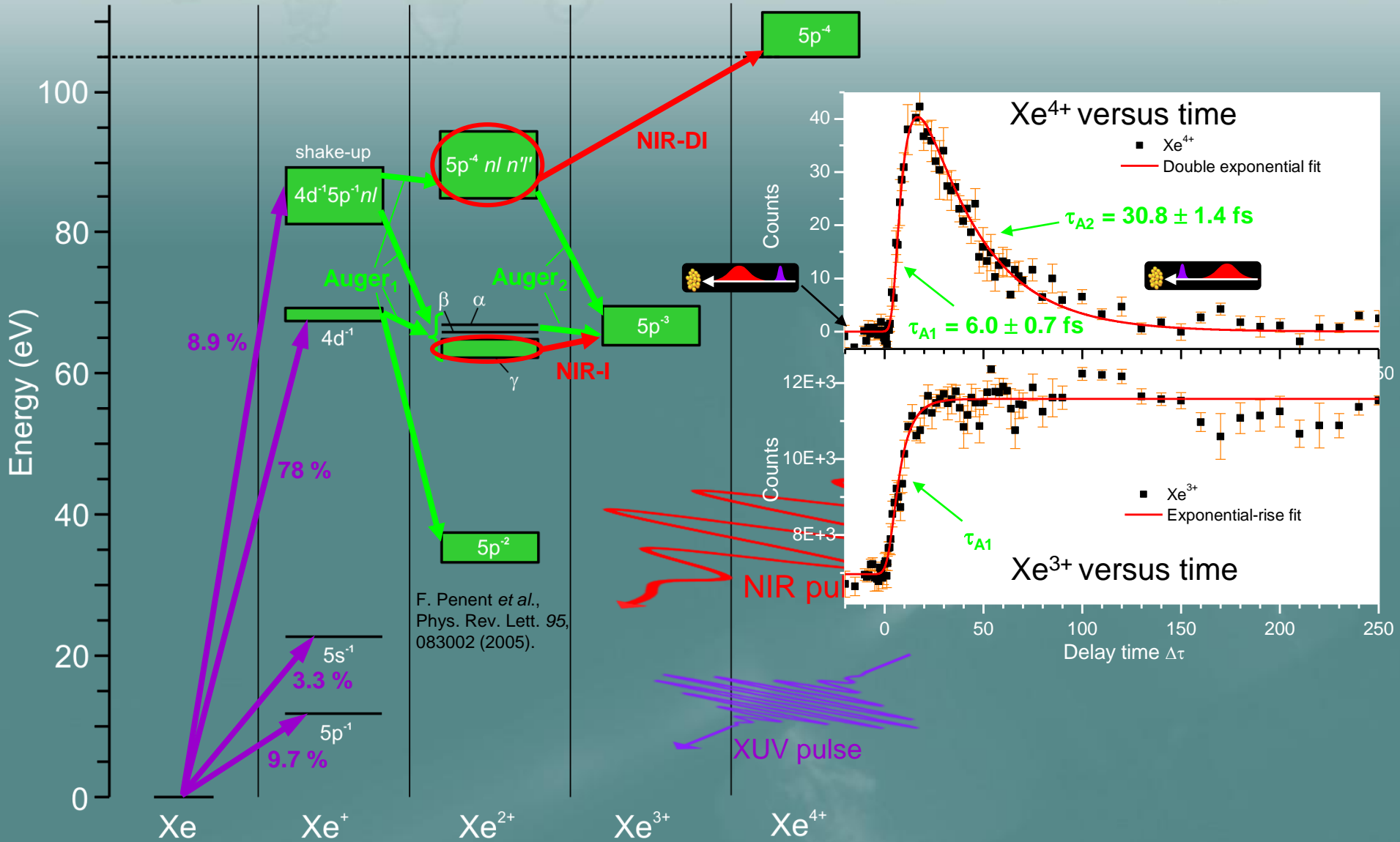
# attosecond tunneling spectroscopy

first experiments in neon and xenon

# testing the sub-fs resolution with neon



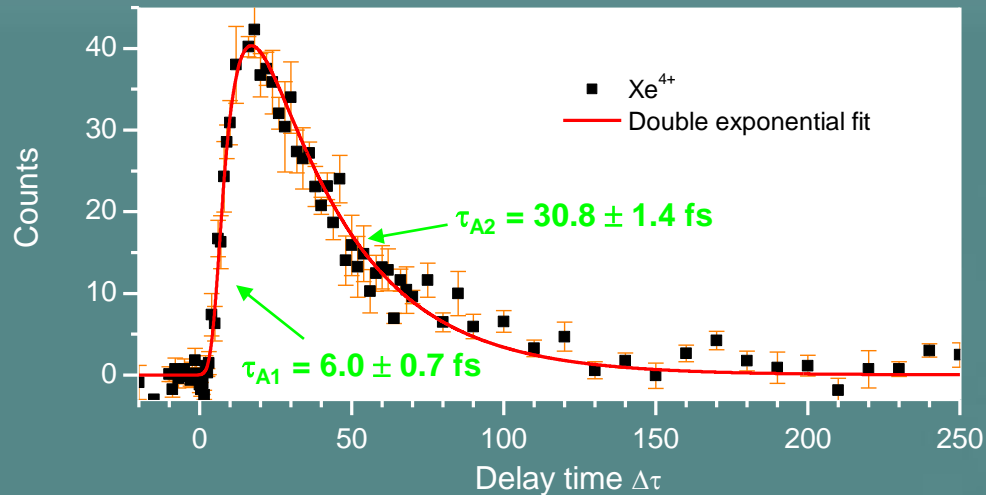
# xenon energy levels – illustration of dynamics



# resolving electron dynamics in xenon

this experiment:

time-integral  
frequency-resolved  
experiments:



..to be published

$$\tau_{A1} (4d_{3/2}) = 6.3 \pm 0.2 \text{ fs}$$

$$\tau_{A1} (4d_{5/2}) = 5.9 \pm 0.2 \text{ fs}$$

$$\tau_{A2} > 23 \text{ fs}$$

F. Penent, Phys. Rev. Lett. 95, 083002 (2005).

## coworkers & collaborators

### postdoctoral:

A. Apolonski

A. Baltuska

A. Cavalieri

T. Fuji

E. Goulielmakis

R. Kienberger

J. Seres

M. Uiberacker

V. Yakovlev

### PhD candidates:

N. Ishii

T. Metzger

J. Rauschenberger

M. Schultze

C. Theisset

A. Verhoef

### xuv optics & atomic spectroscopy:

Th. Uphues, U. Kleineberg, U. Heinzmann  
Univ. Bielefeld, Germany

M. Drescher

Univ. Hamburg, DESY, Germany

### light phase control:

Ch. Gole, R. Holzwarth, T. Udem, T. W. Hänsch  
Univ. Munich - MPQ Garching, Germany

### & measurement:

G. Paulus, H. Walther

A&M Univ. Texas, USA / MPQ Garching

Ch. Lemell, J. Burgdörfer, A. Scrinzi  
Vienna Univ. Techn., Austria

### metrology:

P. B. Corkum, M. Yu. Ivanov

NRC Canada, Ottawa, Canada

### molecular spectroscopy:

M. Kling, M. Vrakking

AMOLF, Amsterdam, Netherlands

M. Lezius, K. Kompa

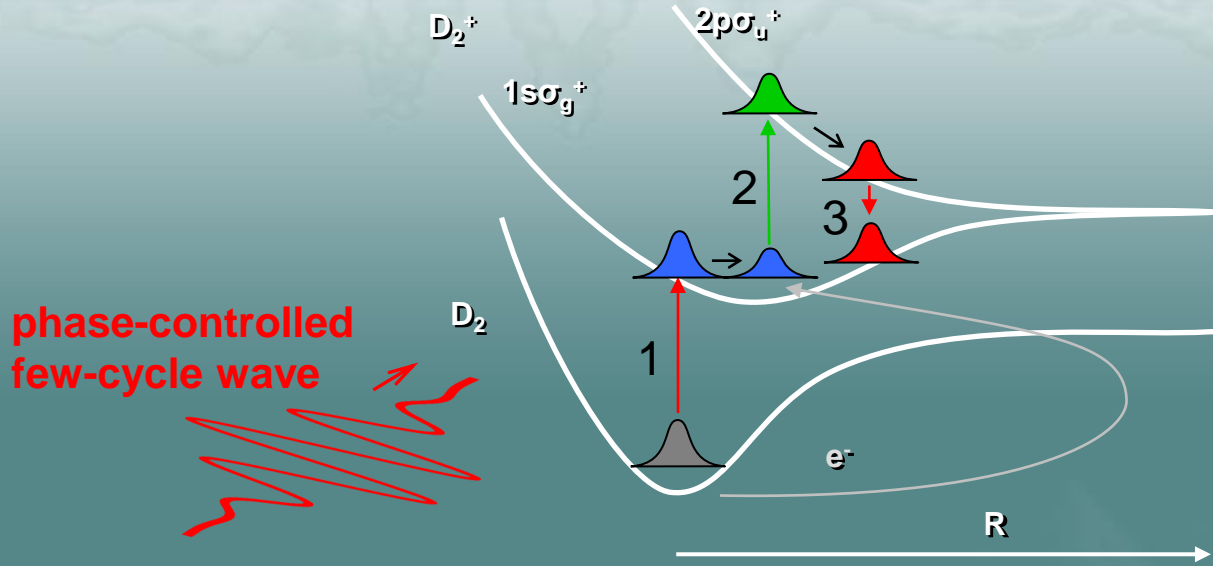
MPQ Garching, Germany

graphics: **Barbara Ferus**

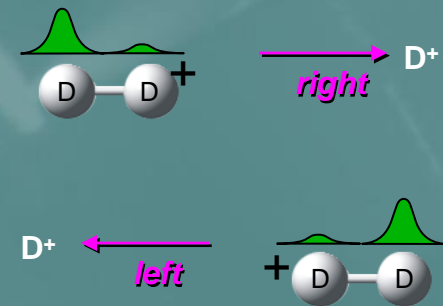
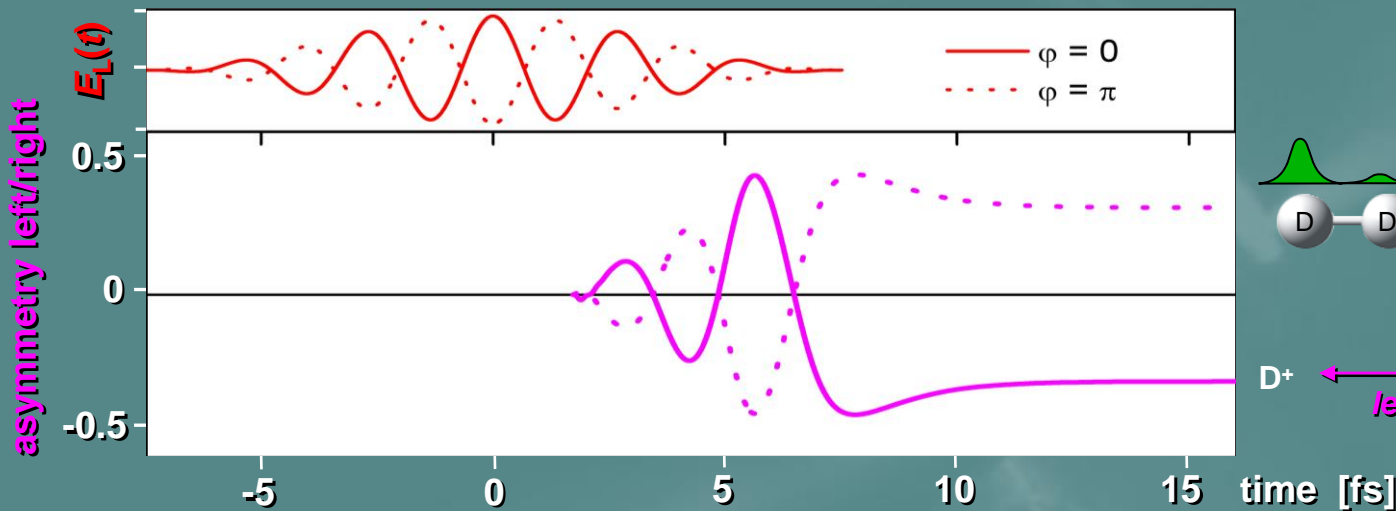


**End**

# may attosecond control of electronic motion in chemical bonds affect the outcome of molecular dynamics?



- 1** ionization of  $D_2$
- 2** recollisional excitation
- 3** formation of a coherent superposition ( $1s\sigma_g^+, 2p\sigma_u^+$ ) state in  $D_2^+$



⇒ YES: direction of emission of  $D^+$  is controlled by light waveform

