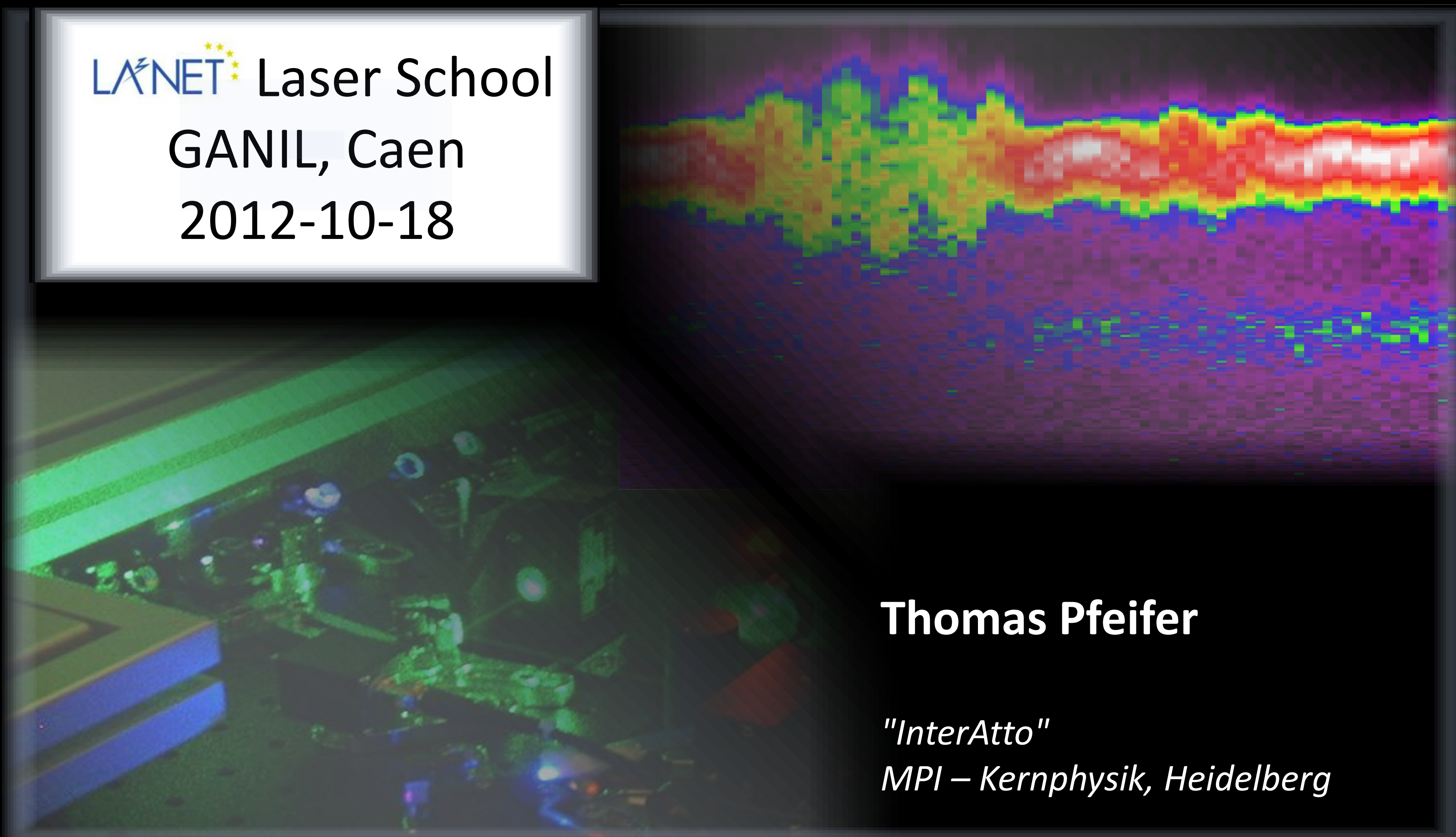
Ultrashort pulsed lasers

and science on femto- and attosecond time scales

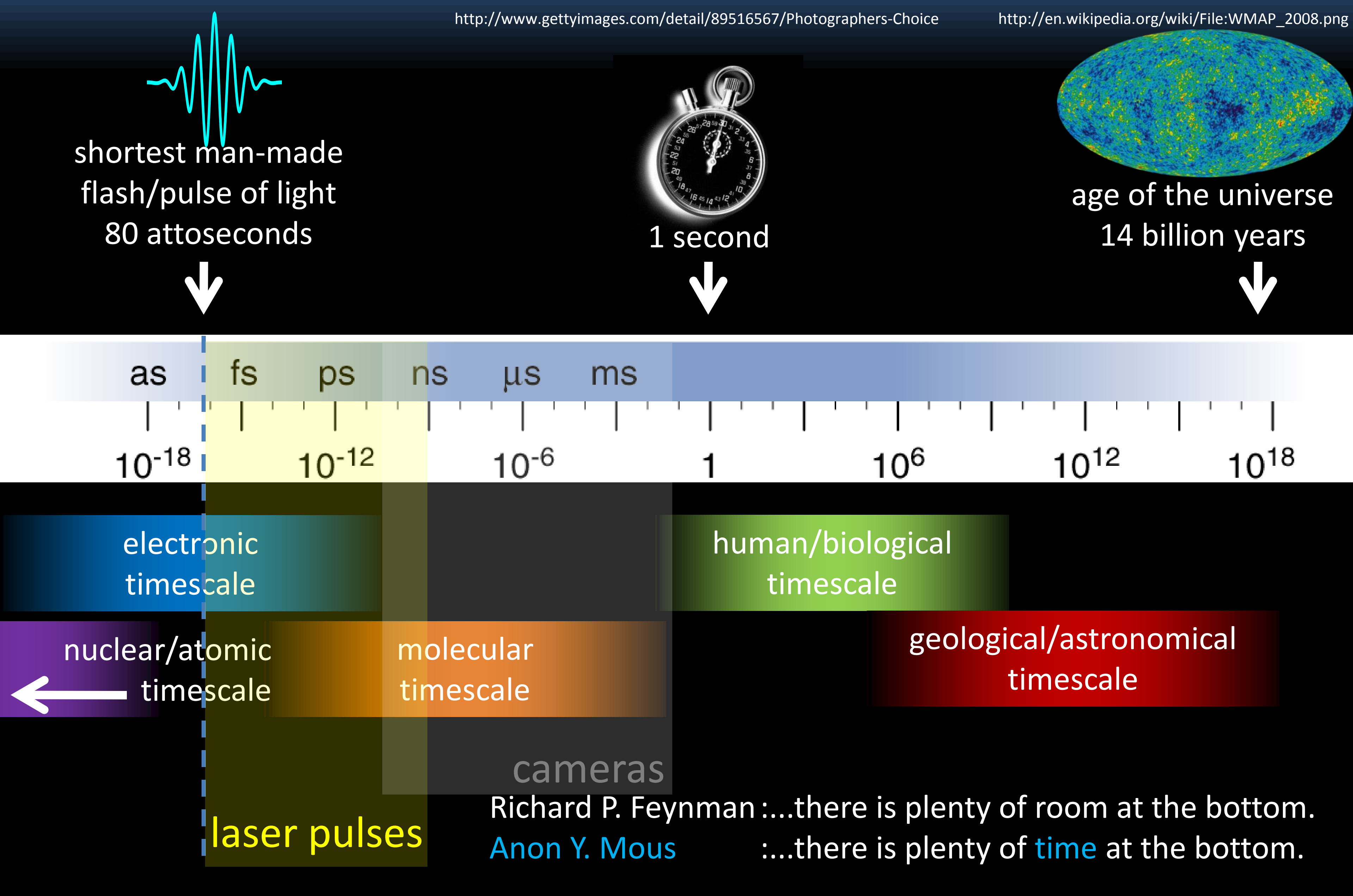


LA³NET Laser School
GANIL, Caen
2012-10-18

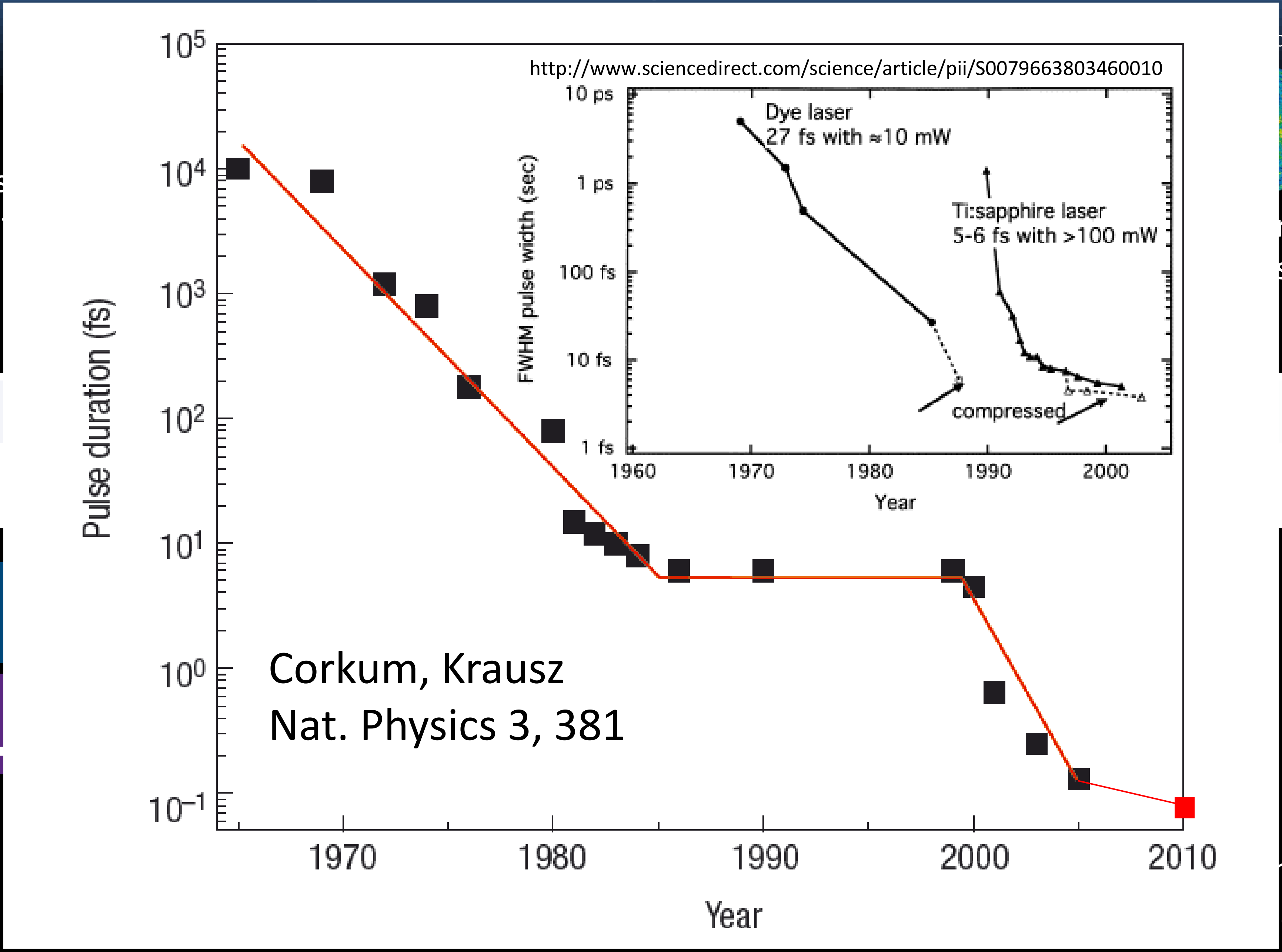
Thomas Pfeifer

"InterAtto"
MPI – Kernphysik, Heidelberg

time scales in science



history of laser pulse duration

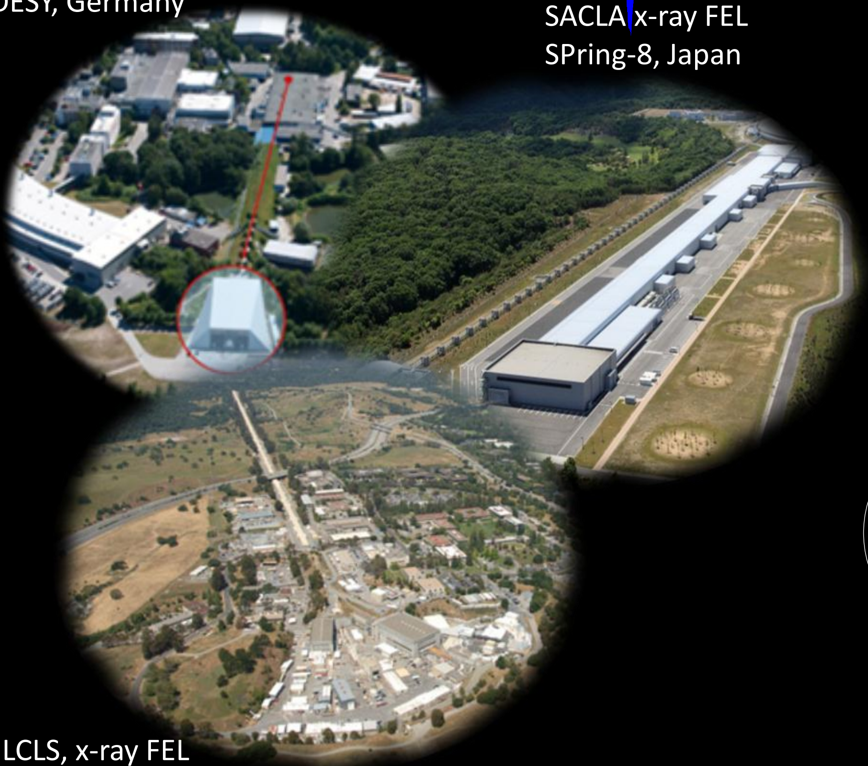


Two parallel revolutions in laser science

Free Electron Lasers

FLASH, XUV-FEL
DESY, Germany

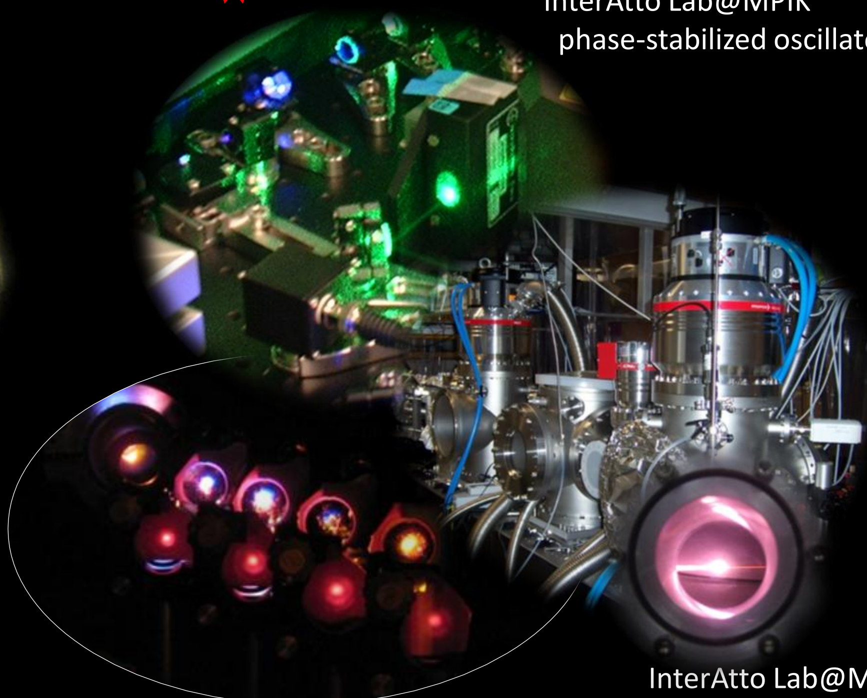
SACLA x-ray FEL
SPring-8, Japan



LCLS, x-ray FEL
SLAC, USA

High Harmonic Generation

InterAtto Lab@MPIK
phase-stabilized oscillator



InterAtto Lab@MPIK
vacuum system

InterAtto Lab@MPIK
chirped-mirror compressor

Frontiers of Ultrashort x-ray/XUV Pulses

Free **E**lectron **L**asers | **H**igh **H**armonic **G**eneration

~1 mm

lowest wavelength

~1 Å | ~1 nm

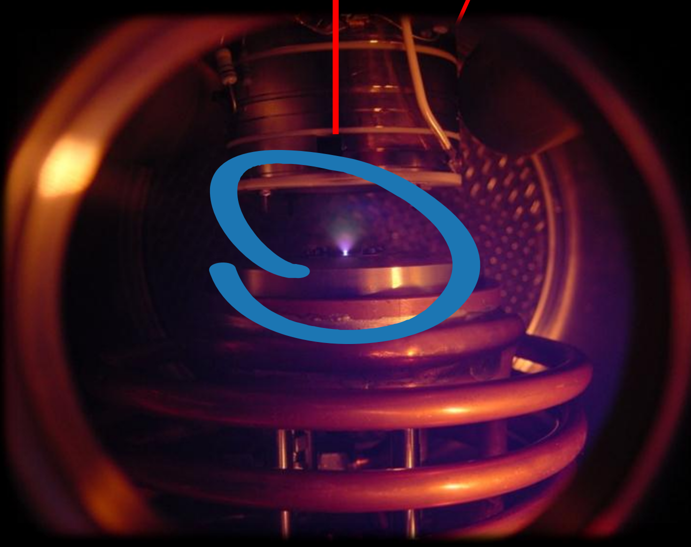
pulse energy

>1 mJ | <1 μJ

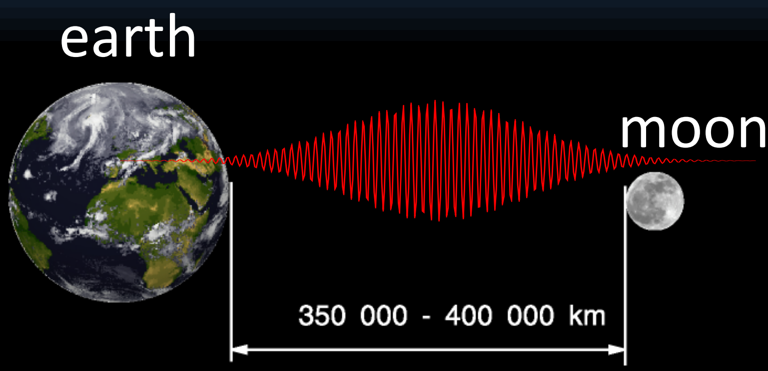
pulse duration

~5 fs | >50 as

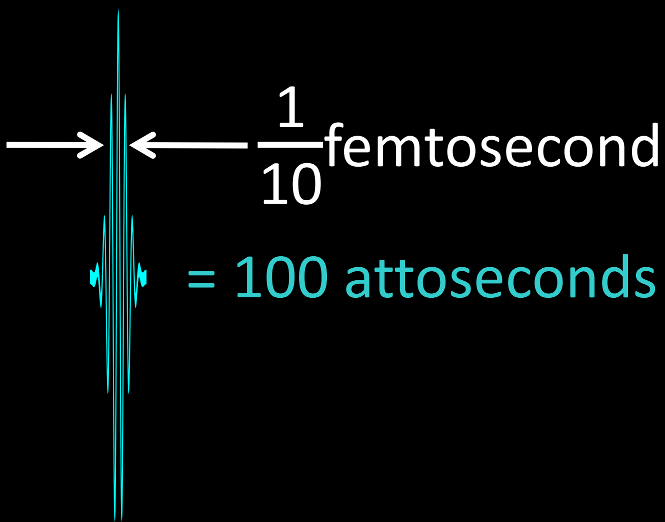
partially coherent | fully coherent



How “long” is one attosecond?



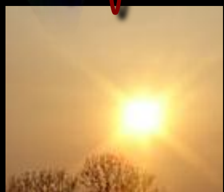
A light pulse of 1 second duration measures 300 000 km in length.

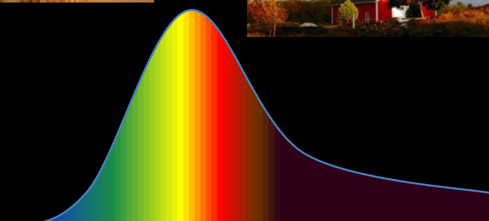


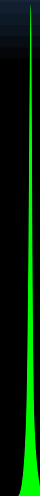
A light pulse of 1 attosecond duration measures 0.3 nm in length.

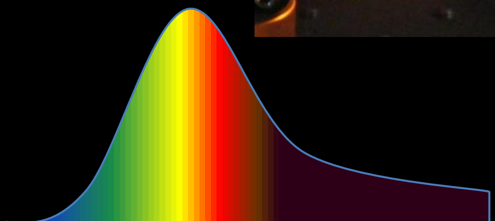
Ref.: Physikalisches Institut, Universität Würzburg

Properties of different light sources







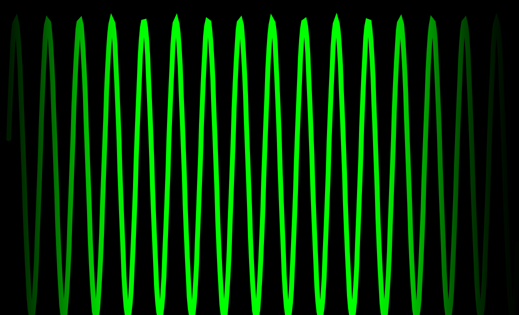


solar spectrum

cw laser spectrum

fs pulse laser spectrum

difference to sun: spectral coherence



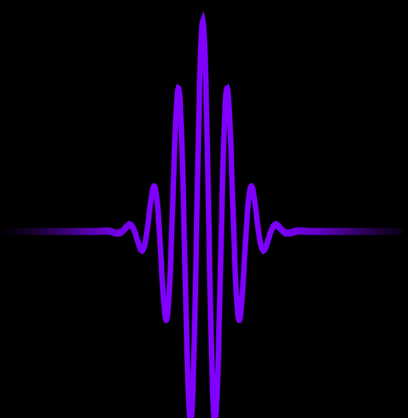


Bild-Quellen: http://www.bhakti-love.com/fotos/sonnenlicht.jpg,
http://climate.met.psu.edu/www_prod/data/frost/images/rainbow.jpg

Maxwell's Equations

$$\nabla \cdot \mathbf{E} = \frac{\rho}{\varepsilon_0} \qquad \nabla \cdot \mathbf{B} = 0$$

$$\nabla \times \mathbf{E} = -\frac{\partial \mathbf{B}}{\partial t} \qquad \nabla \times \mathbf{B} = \mu_0 \mathbf{J} + \mu_0 \varepsilon_0 \frac{\partial \mathbf{E}}{\partial t}$$

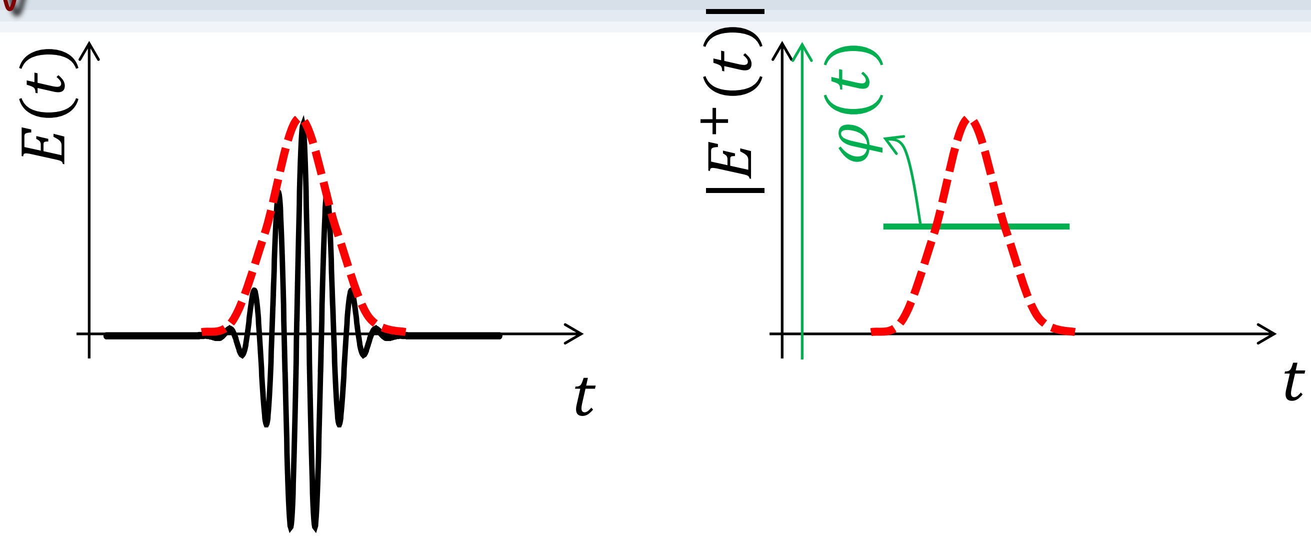
Resulting wave equations:

$$\left(\nabla^2 - \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right)\mathbf{E} = 0 \qquad \left(\nabla^2 - \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right)\mathbf{B} = 0$$

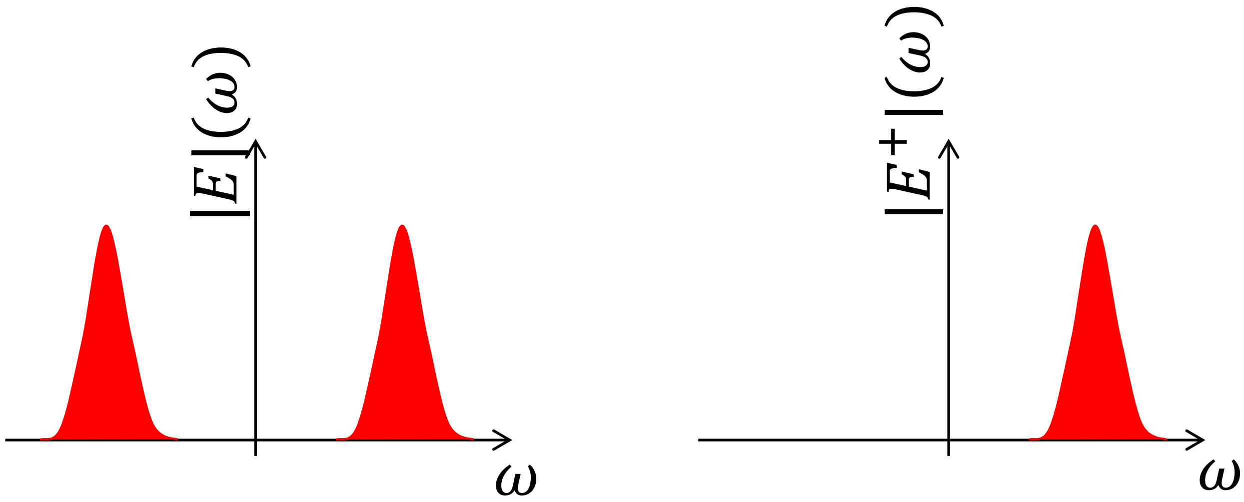
Solutions:

$$\mathbf{E}(\mathbf{r},t) = \mathbf{E}_0 e^{i(\omega t - \mathbf{k}\cdot\mathbf{r})} + c.c. \qquad \mathbf{B}(\mathbf{r},t) = \mathbf{B}_0 e^{i(\omega t - \mathbf{k}\cdot\mathbf{r})} + c.c.$$

E and E^+ representations



$$G(t)\cos(\omega t + \Phi(t)) \rightarrow G(t)\exp(i\omega t + i\Phi(t))$$



Mathematics of ultrashort pulses

$$E^{+}(t) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} \tilde{E}^{+}(\omega) e^{i\omega t} \, d\omega. \qquad (1.6)$$

The inverse transformation returns

$$\tilde{E}^{+}(\omega) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} E^{+}(t) e^{-i\omega t} \, dt. \qquad (1.7)$$

Spectral phase and dispersion

$$E^+(t) \; = \; A(t)e^{i\phi(t)} \qquad \qquad \tilde{E}^+(\omega) \; = \; \tilde{A}(\omega)e^{i\tilde{\phi}(\omega)}$$

Taylor expansion of the phase functions yields:

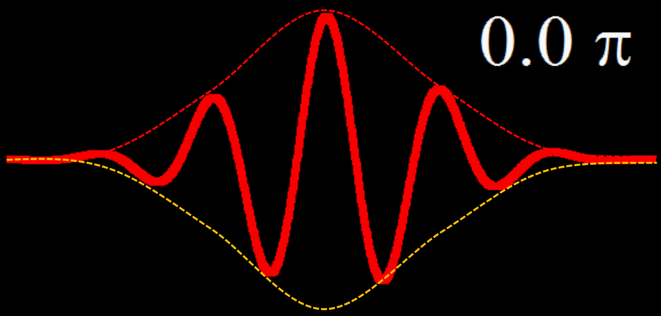
$$\phi(t) = \sum_{j=0}^{\infty} \frac{a_j}{j!} t^j, \qquad \qquad \tilde{\phi}(\omega) = \sum_{j=0}^{\infty} \frac{\tilde{a}_j}{j!} \omega^j.$$

a_0💻 constant phase (CEP)
a_1💻 spectral shift by a_1 (along ω)
a_2💻 linear chirp

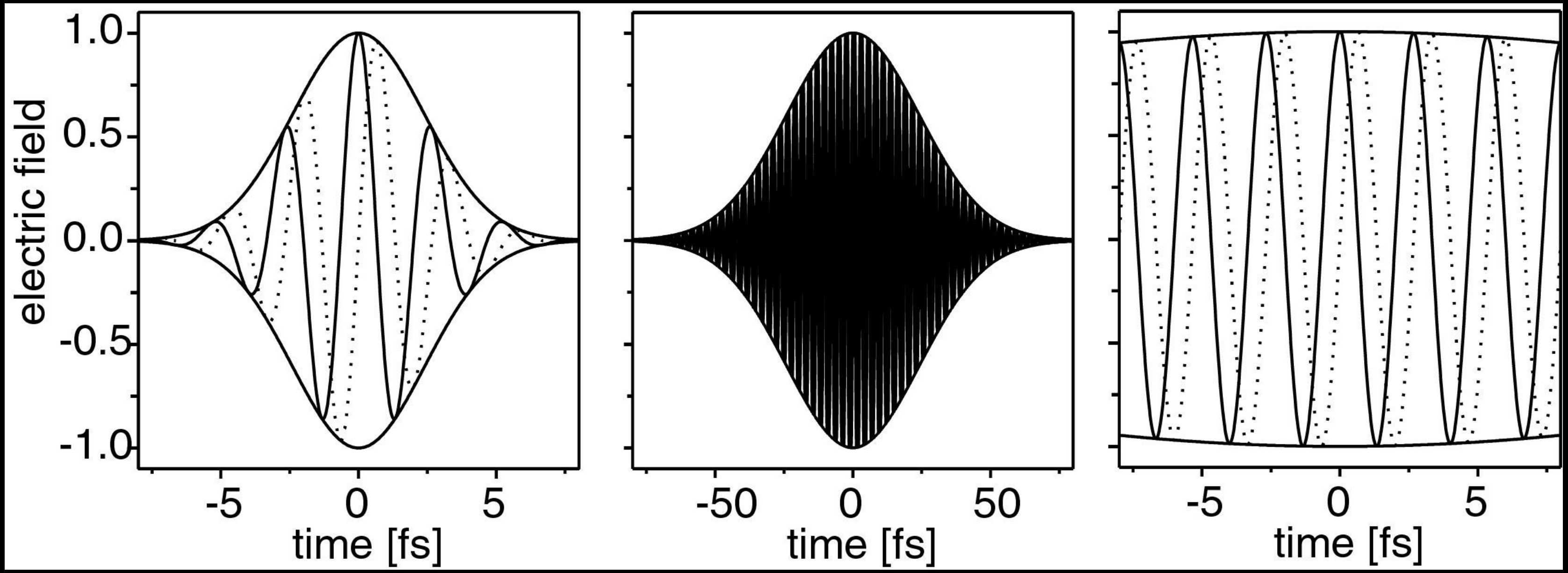
$\tilde{a}_0$💻 constant phase (CEP)
$\tilde{a}_1$💻 temporal shift by $\tilde{a}_1$ (along t)
$\tilde{a}_2$💻 linear chirp

T.P. doctoral thesis, Würzburg University (2004)

absolute (carrier-envelope) phase



CEP: Carrier-Envelope Phase

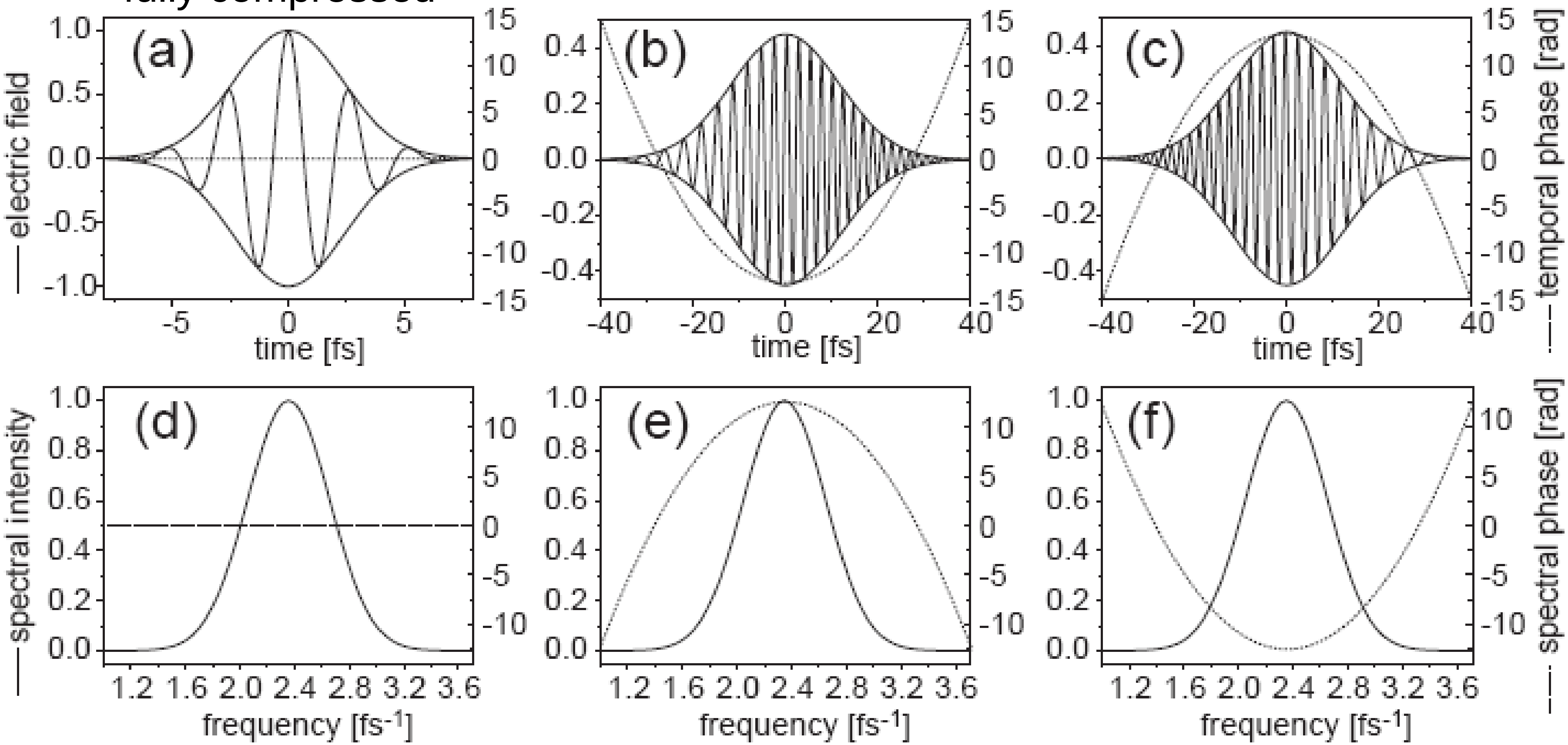


Chirped pulses

bandwidth-limited pulse
fully compressed

up-chirped pulse

down-chirped pulse



flat spectral phase

negatively-curved
spectral phase

positively-curved
spectral phase

Spectral phase and dispersion

$$E^+(t) = A(t)e^{i\phi(t)} \qquad \tilde{E}^+(\omega) = \tilde{A}(\omega)e^{i\tilde{\phi}(\omega)}$$

Taylor expansion of the phase functions yields:

$$\phi(t) = \sum_{j=0}^{\infty} \frac{a_j}{j!} t^j, \qquad \tilde{\phi}(\omega) = \sum_{j=0}^{\infty} \frac{\tilde{a}_j}{j!} \omega^j.$$

a_0 💻 constant phase (CEP)
a_1 💻 spectral shift by a_1 (along ω)
a_2 💻 linear chirp

$\tilde{a}_0$ 💻 constant phase (CEP)
$\tilde{a}_1$ 💻 temporal shift by $\tilde{a}_1$ (along t)
$\tilde{a}_2$ 💻 linear chirp

Propagation along direction z in refractive media results in a change of the electricd fields:

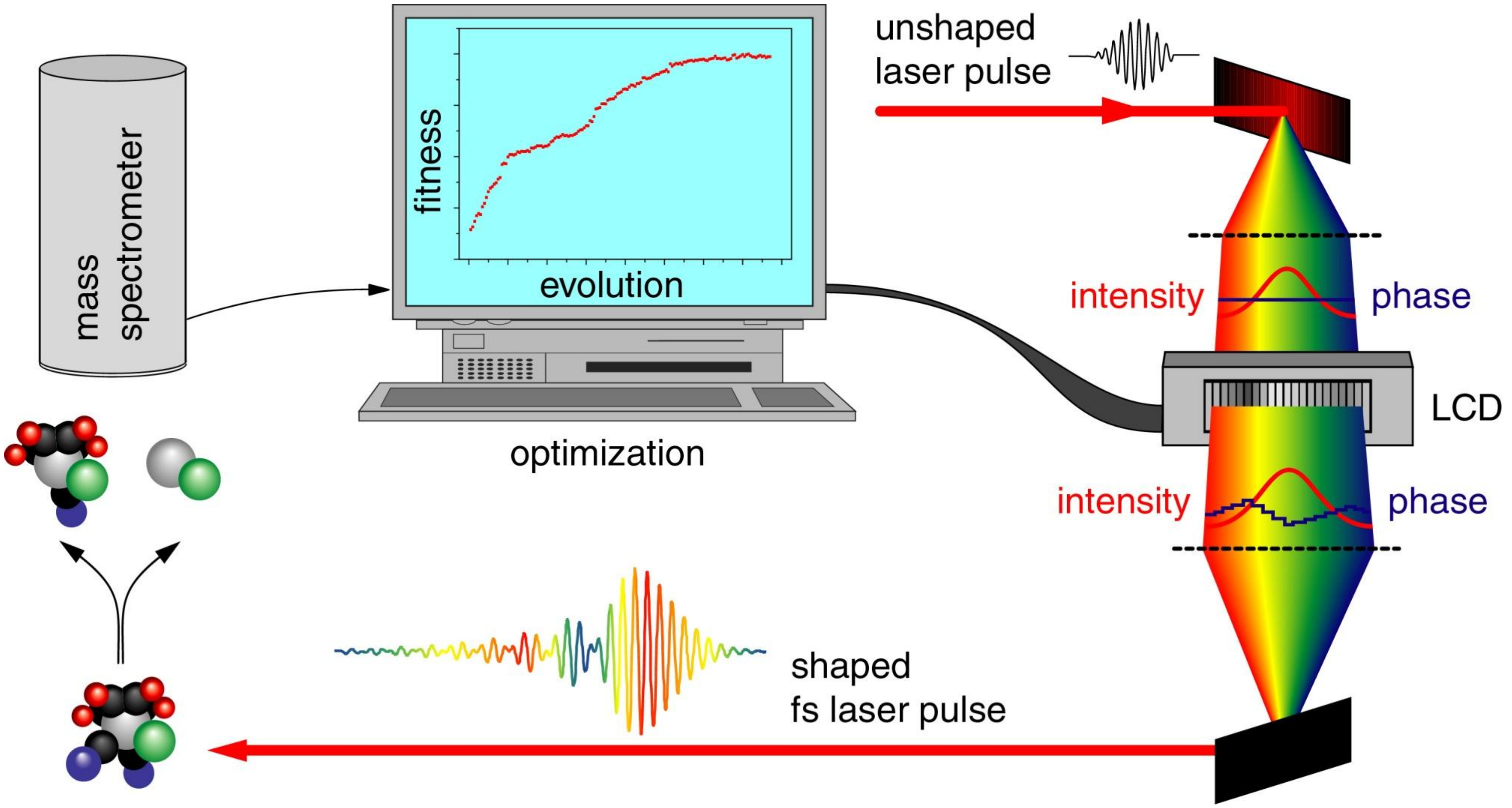
$$\tilde{E}^+(\omega, z) = \tilde{A}(\omega, z)e^{i\tilde{\phi}(\omega)}e^{-ik(\omega)z}$$

where

$$\begin{aligned} k(\omega) &= n(\omega)k_{\text{vac}} \\ &= n(\omega)\frac{\omega}{c}, \end{aligned}$$

T.P. doctoral thesis, Würzburg University (2004)

Femtosecond Laser Pulse Shaping

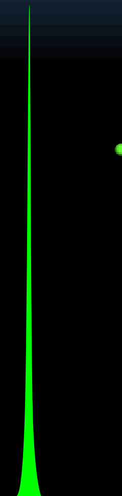


A. Assion et al.
Science **282**, 919 (1998)

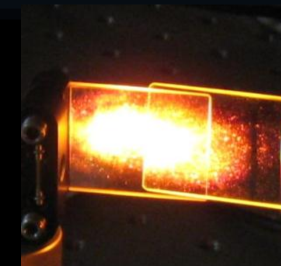
Properties of different light sources



solar spectrum



cw laser spectrum




fs pulse laser spectrum

difference to sun: spectral coherence

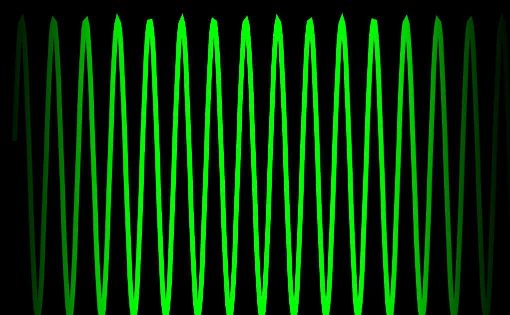
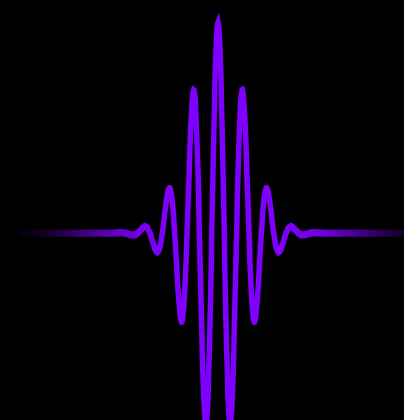
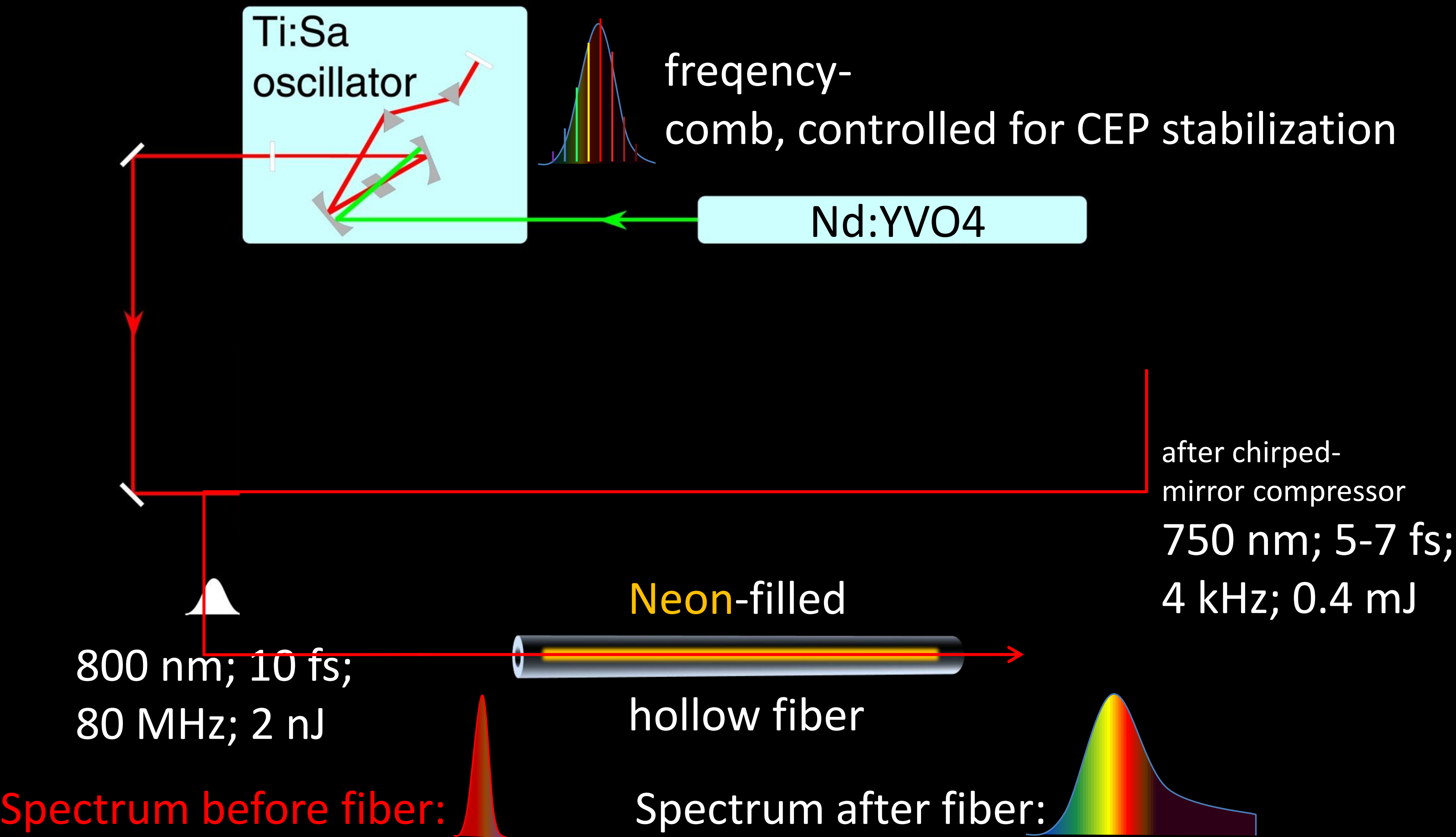




Bild-Quellen: http://www.bhakti-love.com/fotos/sonnenlicht.jpg,
http://climate.met.psu.edu/www_prod/data/frost/images/rainbow.jpg

Ultrafast Laser System

for generating few-cycle pulses



Short pulse measurement

“to measure a fast event, you need an at least equally fast probe”

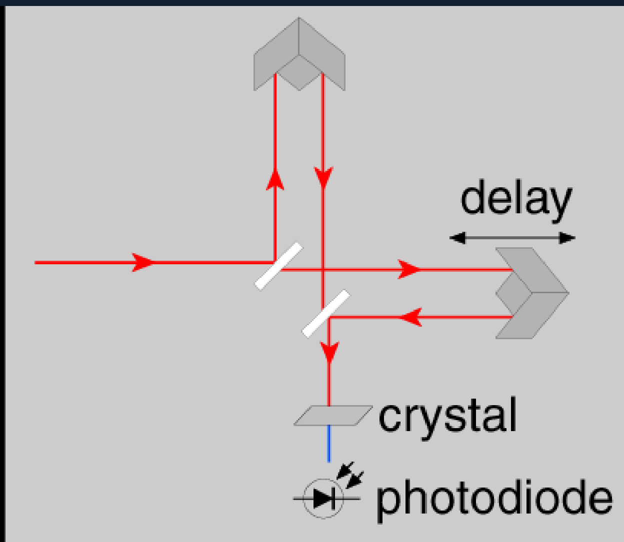
- Autocorrelation
  ‘Auto...’ -> 'self'...

- **F**requency-**R**esolved **O**ptical **G**ating
  FROG, building upon Autocorrelation

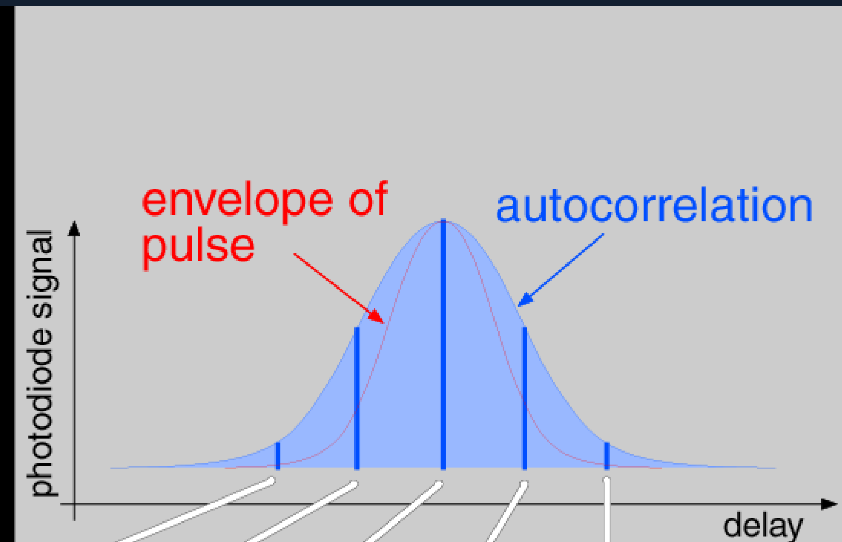
- **T**emporal **A**nalysis by **D**ispersing a **P**air Of **L**ight **E**lectric **F**ields
  TADPOLE (also known as "spectral interferometry")

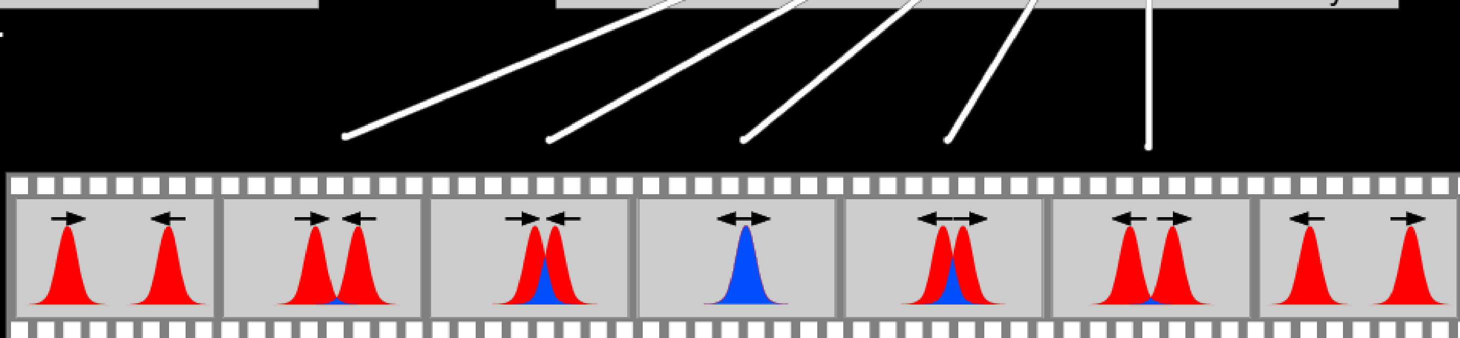
- **S**pectral **I**nterferometry for **D**irect **E**lectric **F**ield **R**econstruction
  SPIDER, building upon TADPOLE

Autocorrelation



autocorrelator



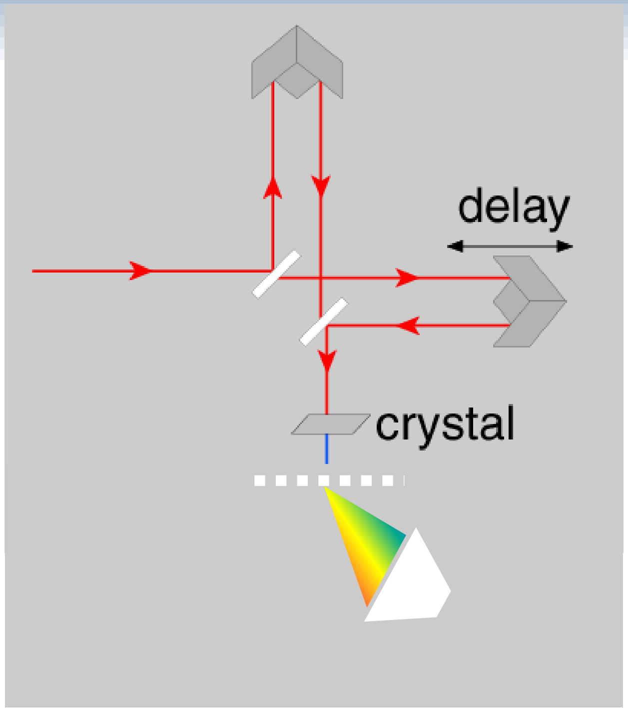


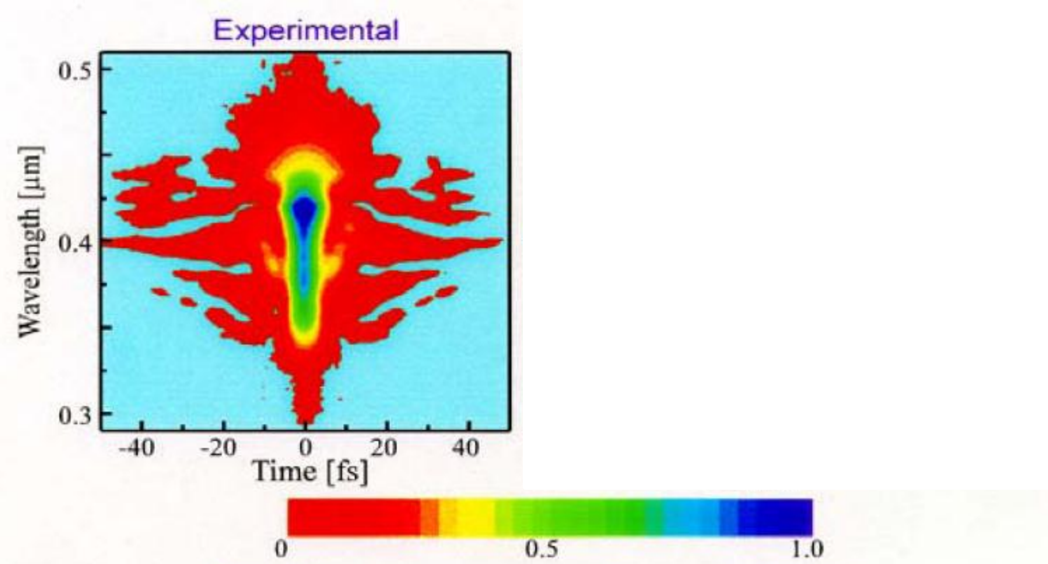
linear (no crystal) $$I_M(\tau) = \int_{-\infty}^{+\infty} |E(t) + E(t-\tau)|^2 dt$$

nonlinear (with crystal) $$I_M(\tau) = \int_{-\infty}^{+\infty} |(E(t) + E(t-\tau))^2|^2 dt$$

FROG idea

D. J. Kane and R. Trebino, Opt. Lett. 18, 823 (1993)





measure spectrum as
a function of time delay

2-dim. data sets: ‘FROG-trace’

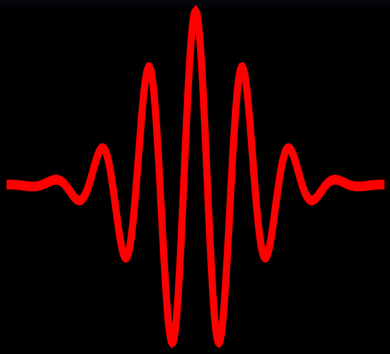
analysis by iterative algorithm

Ref: http://www.physics.gatech.edu/frog/

Applications of femtosecond pulses

ultrashort laser pulses

1 femtosecond = 10^{-15} seconds



1000000000000000

observation of ultrafast processes

Ref: Ulrich Weichmann, Department of Physics, Wuerzburg University

Snapshots of Fast Processes

exposure time too large:
blurred image

insufficient temporal resolution

exposure time short enough:
sharp image

sufficient temporal resolution

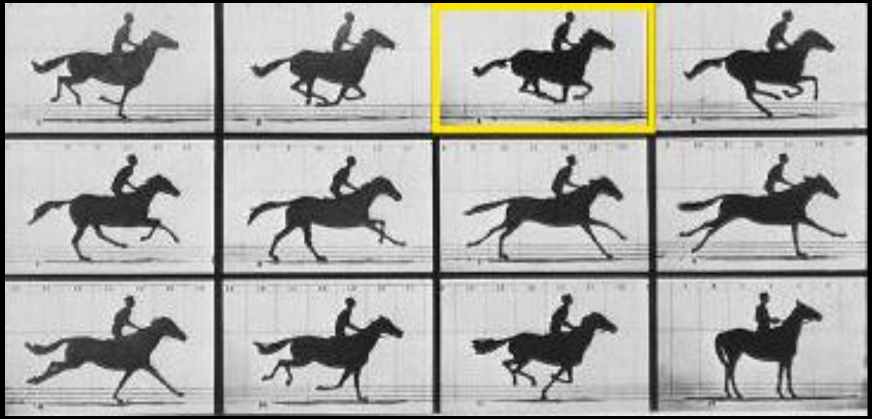
Ref: Physics Department, University of Wuerzburg

Why use ultrashort laser pulses?

Does a galloping horse, at any time, have all legs in the air?

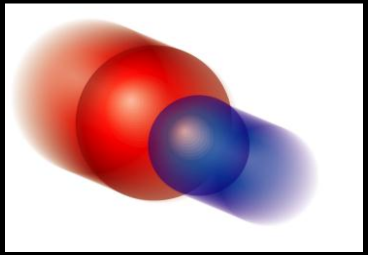
required time resolution: milliseconds

1877, Eadweard Muybridge, Leland Stanford



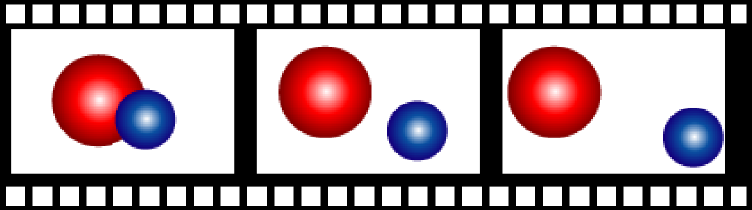
slow-motion with short exposure times helps to clear up fast events

How do atoms move within molecules?



required time resolution: femtoseconds

1 fs = 10^{-15} s



extreme slow-motion with fs laser pulses helps to illuminate ultrafast events

Ref: Physics Department, University of Wuerzburg

Estimation of Characteristic Time Scales

Molecular rotation frequency

T_r=300 fs

$$\omega = \frac{L}{I} \approx \frac{\hbar}{m_p a_0^2} \approx \frac{1}{2000} \text{ a.u.}$$

Molecular vibration frequency

T_v=7 fs

$$\omega \approx \sqrt{\frac{D}{m_p}} \approx \sqrt{\frac{1}{2000}} \approx \frac{1}{50} \text{ a.u.}$$

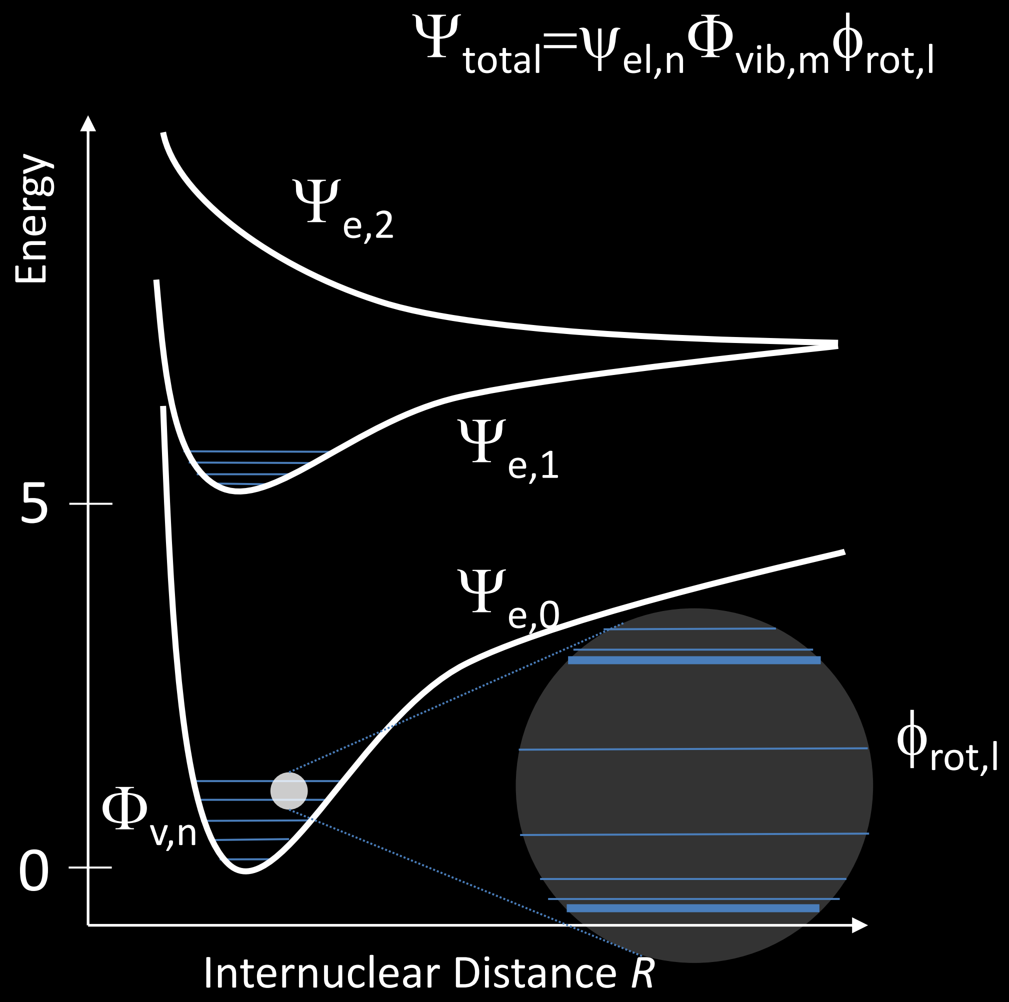
Electron (rotation) frequency

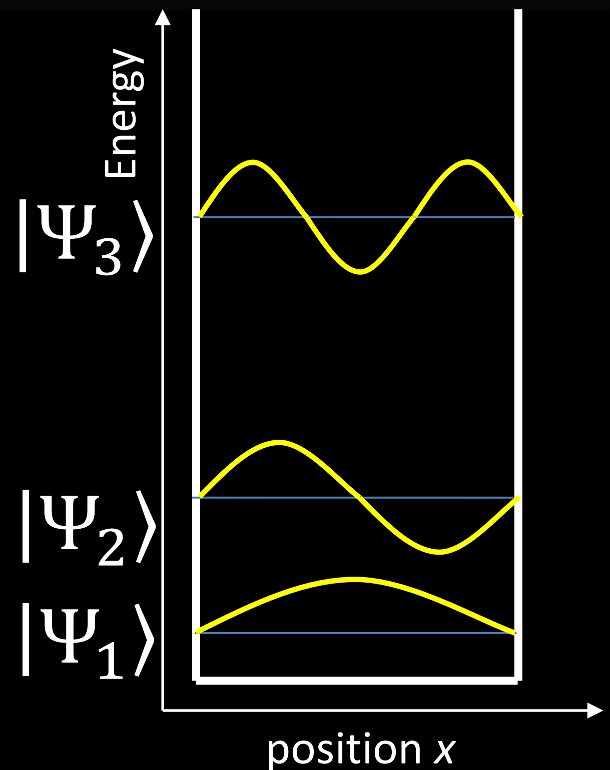
(quasi-classical Bohr model) T_e=150 as

$$\omega = \frac{L}{I} \approx \frac{\hbar}{m_e a_0^2} = \frac{1}{1} \text{ a.u.}$$

Quantum Level Spacings

Separation: Electronic, Vibrational, Rotational



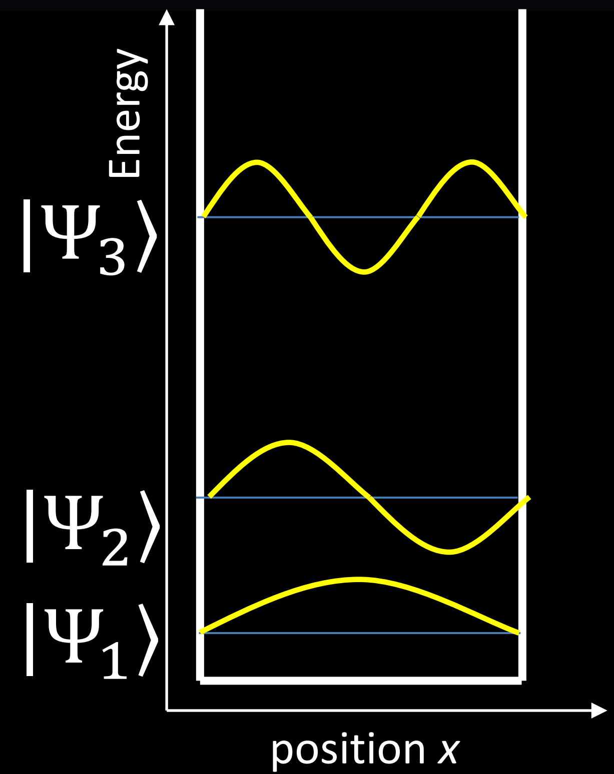


model system: potential well

Wavepacket dynamics and observation

Quantum beat period:

$$\Delta T = \frac{\hbar}{\Delta E} \approx 4.1 \text{ fs}/\ \Delta E\,[\text{eV}]$$



e.g. time-dependent position:

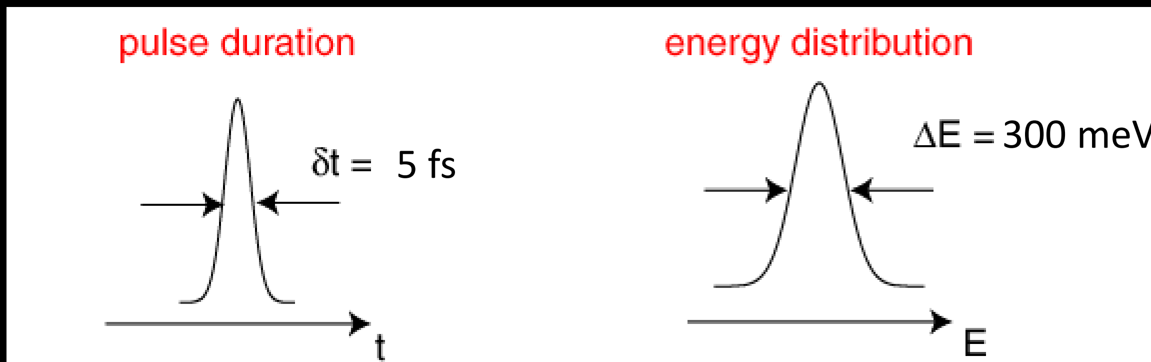
$$x(t) = \langle \Psi(t)|\hat{x}|\Psi(t)\rangle$$

$$= \tfrac{1}{2}\left\langle \Psi_1 e^{\frac{-i}{\hbar}E_1 t} + \Psi_2 e^{\frac{-i}{\hbar}E_2 t}\middle|\hat{x}\middle|\Psi_1 e^{\frac{-i}{\hbar}E_1 t} + \Psi_2 e^{\frac{-i}{\hbar}E_2 t}\right\rangle$$

$$= |\langle \Psi_1|\hat{x}|\Psi_2)\rangle|\ \cos\left[\frac{(E_1 - E_2)}{\hbar}t + \varphi\right]$$

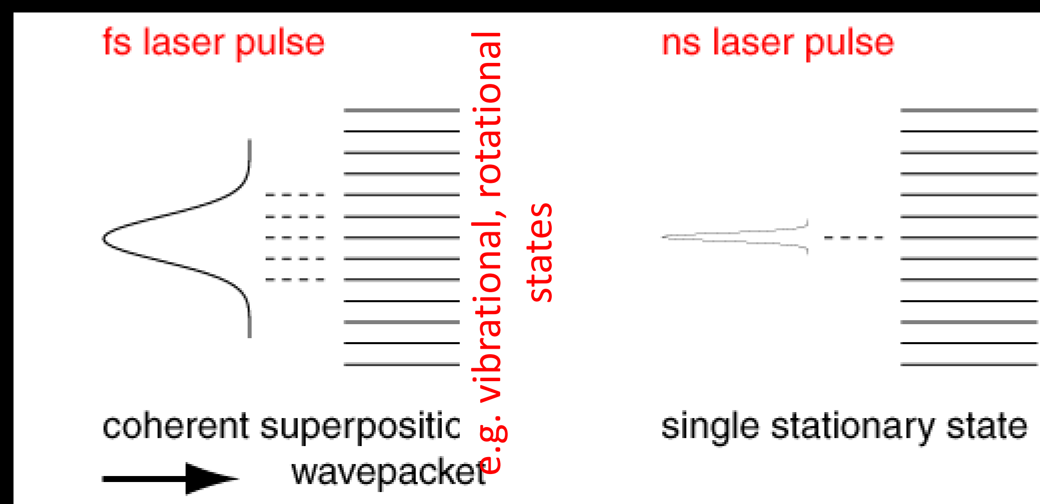
femtosecond laser pulses and processes

1 fs = 10^{-15} sec light travels: 0.3 μm



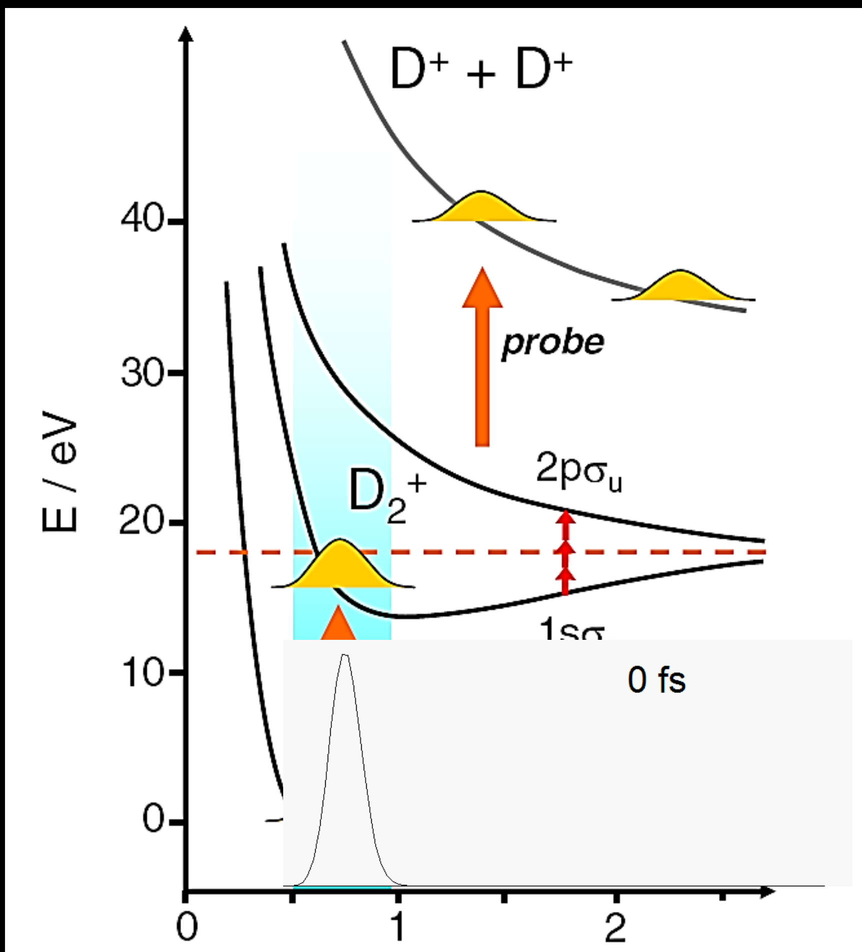
time-bandwidth product:
$\delta t \cdot \Delta E > 1.8$ fs·eV
(δ, Δ: FWHM)

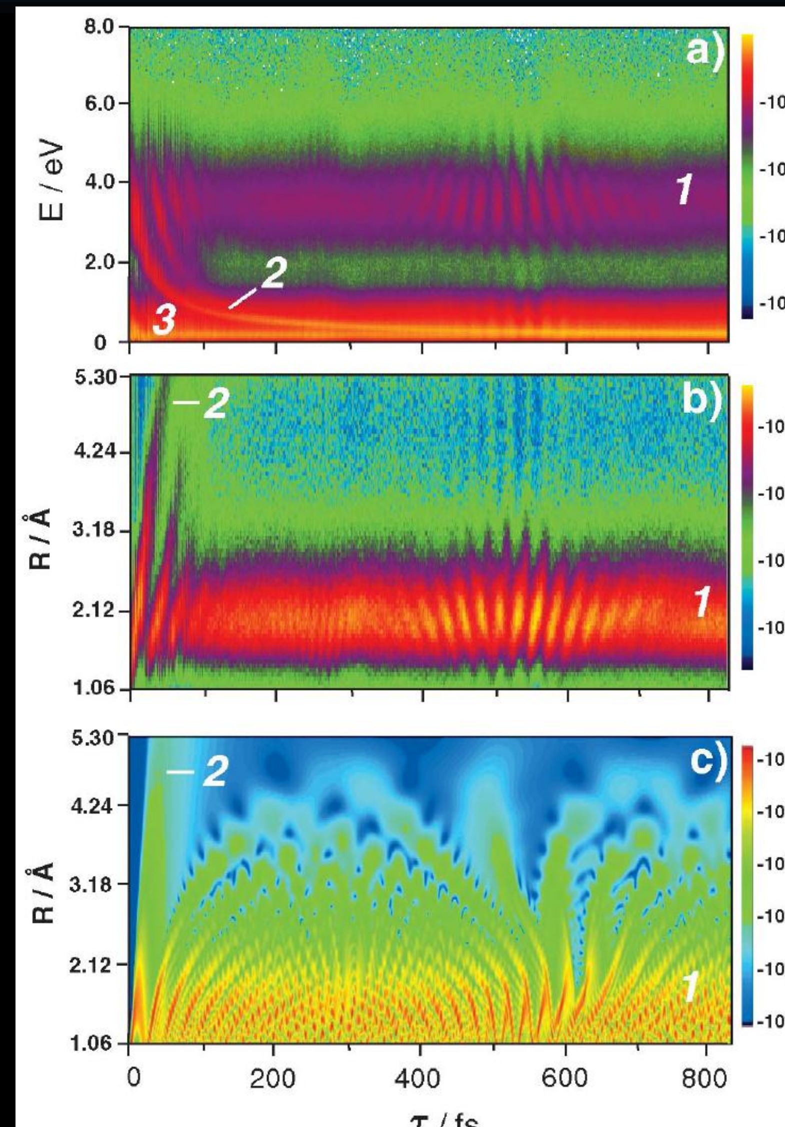
molecular vibrational periods (X_2):	**8 fs –**	**1 ps**
molecular rotational periods (X_2):	**0.2 ps –**	**1 ns**
conformational changes in proteins:	**20 fs – (>1) μs**	



pump–probe spectroscopy in D_2

T. Ergler *et al. (J. Ullrich, R. Moshammer),*
Phys. Rev. Lett. 97, 193001 (2006)

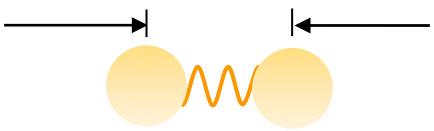




ultrafast quantum motion

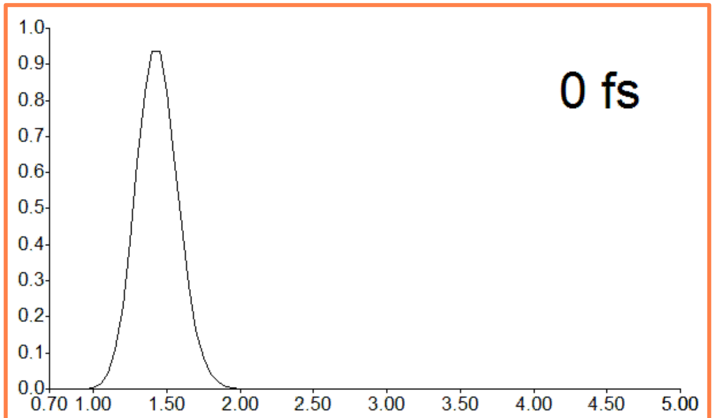
example:
diatomic molecule

internuclear
distance d ~ Å



vibrational period
T > 5 fs ($5 \cdot 10^{-15}$ s)

$|\Psi_{molecule}|^2$ D_2



d [a.u.]

femtosecond
pulsed lasers
(IR, Vis., UV)

femtosecond
spectroscopic techniques

pump–probe,
CARS
…

ultrafast quantum motion

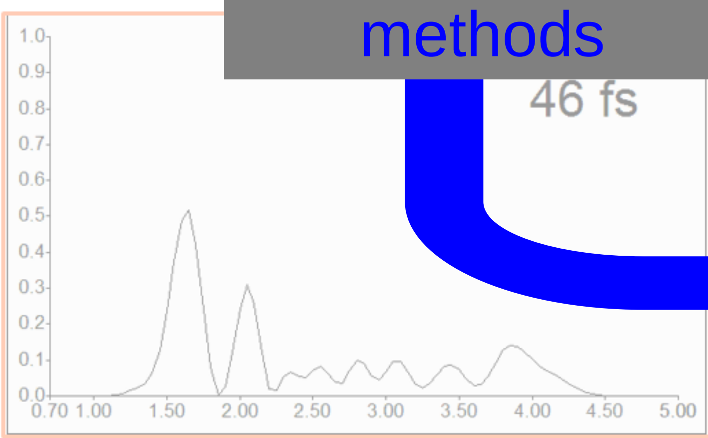
internuclear distance d ~ Å

attosecond pulsed source (soft x-ray)

vibrational period T ~ 5 fs

attosecond spectroscopy methods

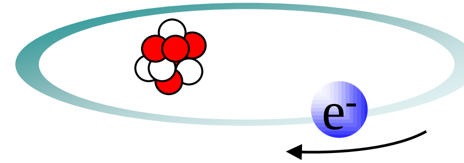
$|\Psi_{molecule}|^2$ D_2



46 fs

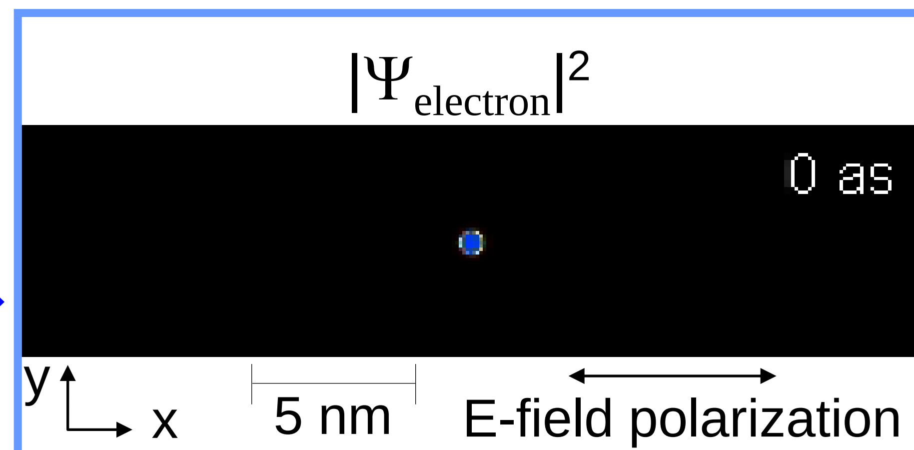
d [a.u.]

example:
electrons in atoms



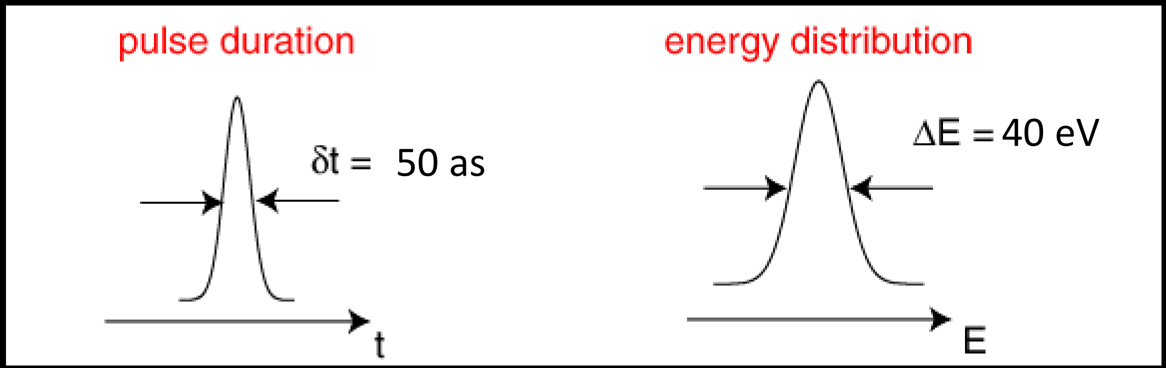
orbital size ~ Å

orbital period
T < (<<) 1 fs
attosecond = 10^{-18} s



attosecond pulses and processes

1 as = 10^{-18} s   light travels: 0.3 nm (3 Ångstrom)

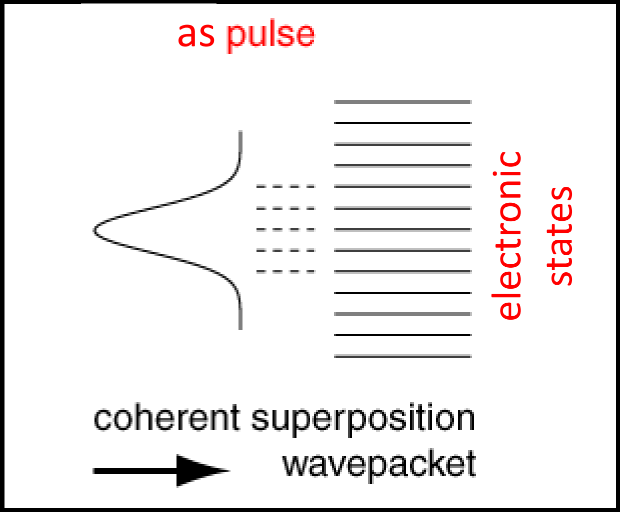


time-bandwidth product:
$\delta t \cdot \Delta E > 1.8$ fs·eV
(δ, Δ: FWHM)

Classical e^- orbit period, Hydrogen: **152 as**
1s-2s/p wavefunction period:
- Hydrogen: **~ 400 as**
- H-like Uranium **~ 0.05 as**

$2s2p\text{-}sp_{23+}$ 2-electron wavepacket period:
- Helium: **1.2 fs**

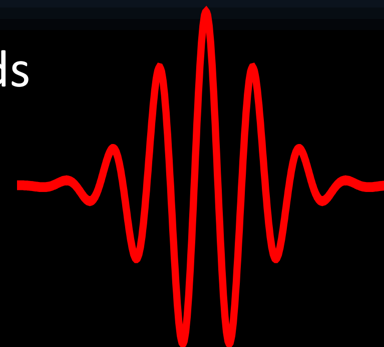
Auger (core-hole) lifetimes: **~100 as-~10 fs**



Generation of Attosecond Pulses

ultrashort laser pulses

1 femtosecond = 10^{-15} seconds



1000000000000000

observation of ultrafast processes

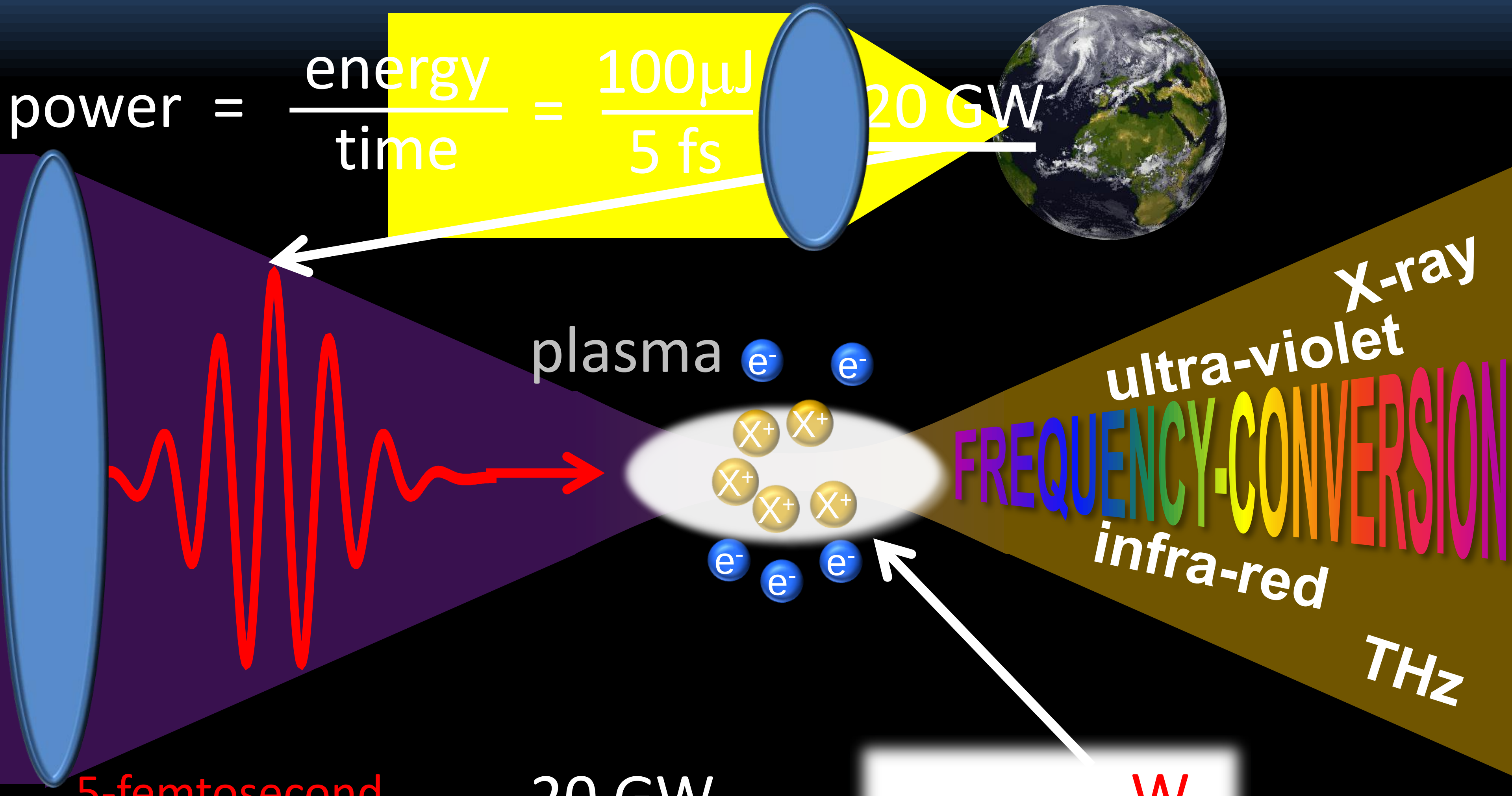
$$\text{power} = \frac{\text{pulse energy}}{\text{time}}$$

concentration of energy in time

Ref: Ulrich Weichmann, Department of Physics, Wuerzburg University

short pulses ⇒ high power

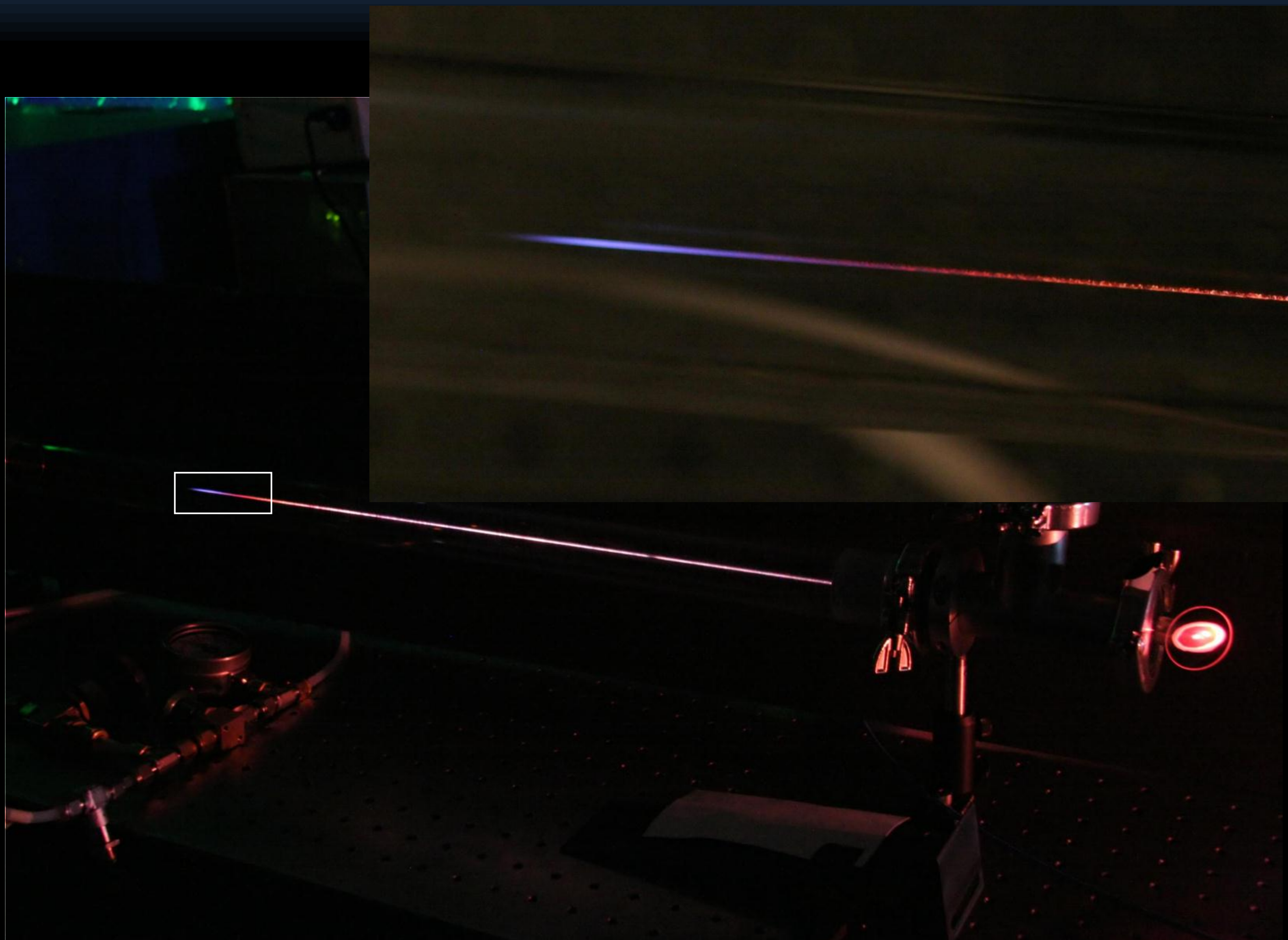
$$\text{power} = \frac{\text{energy}}{\text{time}} = \frac{100\,\mu\text{J}}{5\text{ fs}} = 20\text{ GW}$$



$$\frac{20\text{ GW}}{(10\,\mu\text{m})^2} = 2 \times 10^{16}\,\frac{\text{W}}{\text{cm}^2}$$

compare: solar intensity in earth orbit: $0.14\,\frac{\text{W}}{\text{cm}^2}$

light–matter interaction



High-(order) harmonic generation

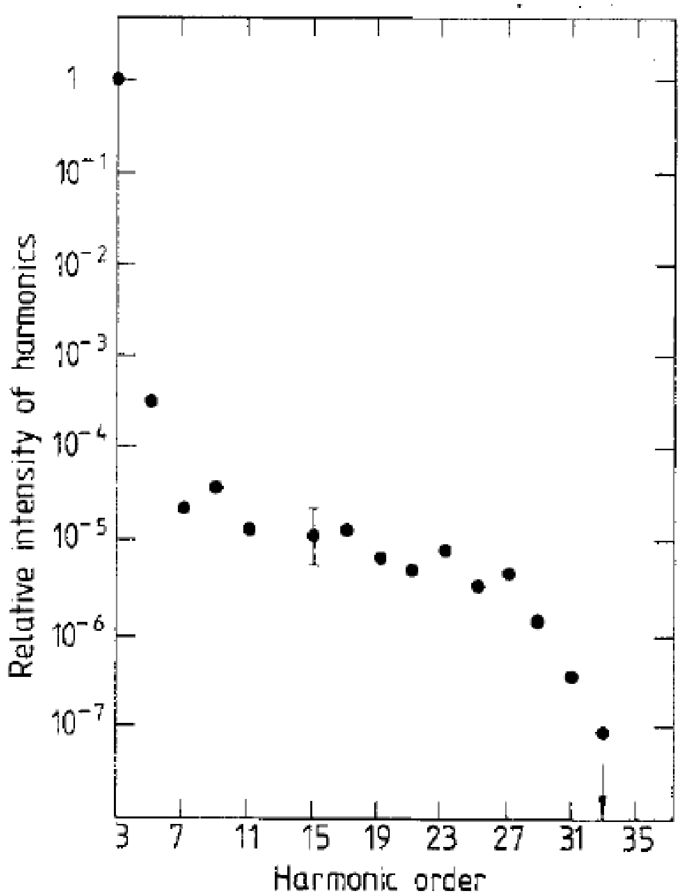


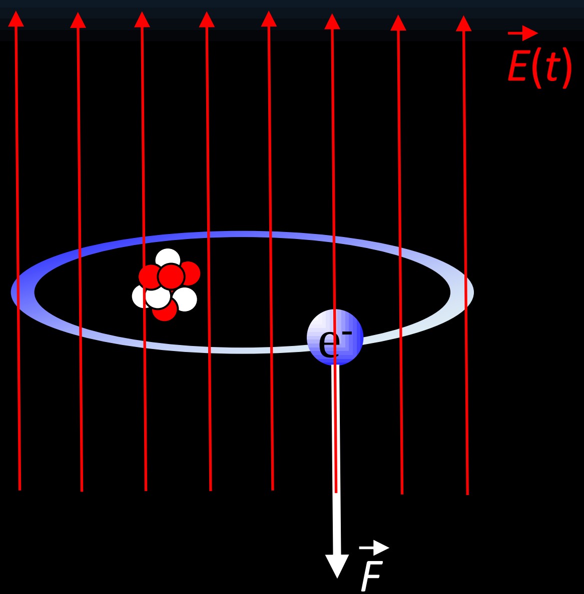
Figure 4. As figure 3 but for Ar. The 13th harmonic is missing due to a strong absorption of the 81.9 nm radiation in the photoexcitation of a 5d state.

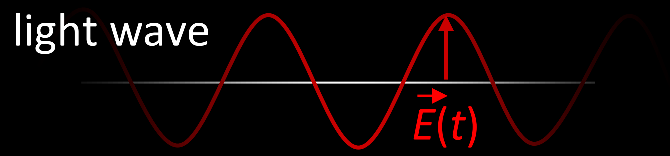
- McPherson *et al*.
  J. Opt. Soc. Am. B **21**, 595 (1987)
- M. Ferray, A. L'Huillier *et al*.
  J. Phys. B **21**, L31 (1988)

intensity:	~10^{13} W/cm^2
wavelength:	1064 nm
pulse duration:	1 ps

laser field acting on electrons

intensity	I =	$2 \cdot 10^{16}$	W/cm^2
electric field	E =	$4 \cdot 10^{11}$	V/m

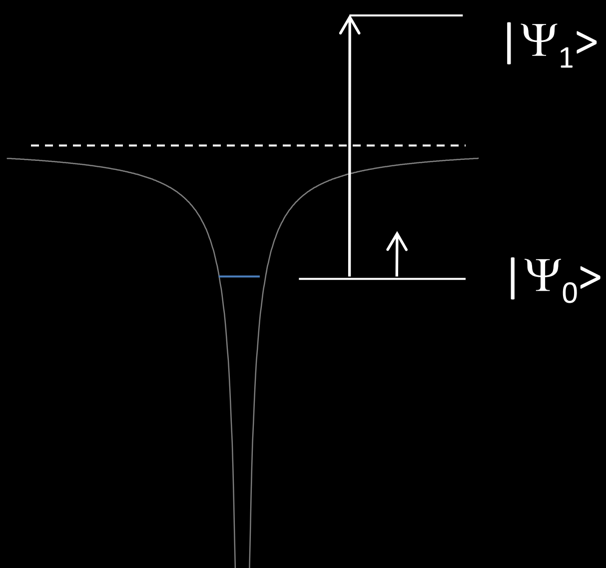




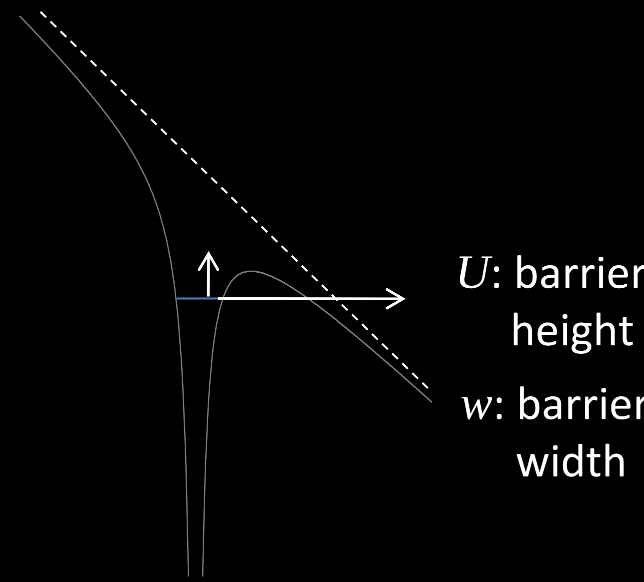
Ionization

Potential: $V = 1/r + e\,E$

Photoelectric effect (weak fields)
(direct transition)



Strong electric field
(Tunneling)



1st order perturbation theory

$$a_1(t) \sim \int_{-\infty}^{t} dt' E(t') \cdot e^{i\hbar(\omega_0 - \omega_1)t'}$$

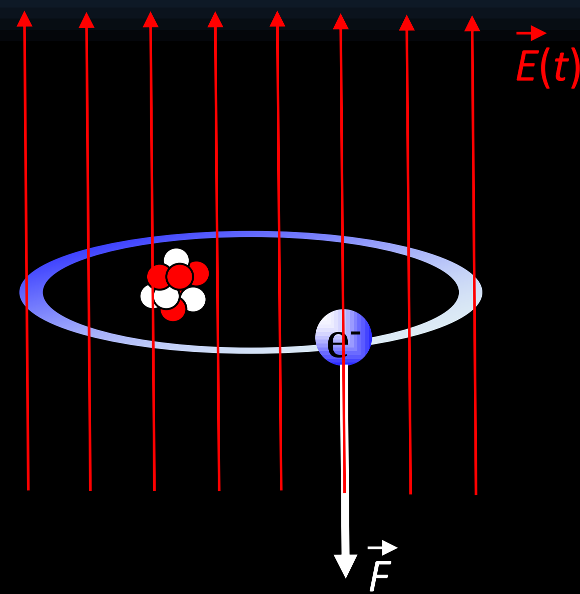
tunneling rate

$$a_c \sim \int_{-\infty}^{t} dt' e^{-\frac{w(E(t'))}{\hbar}\sqrt{2mU(E(t'))}}$$

laser field acting on electrons

intensity	I =	$2 \cdot 10^{16}$	W/cm^2
electric field	E =	$4 \cdot 10^{11}$	V/m
force	F =	64	nN
electron mass	m_e=	$9 \cdot 10^{-31}$	kg
acceleration	a =	$7 \cdot 10^{22}$	m/s^2

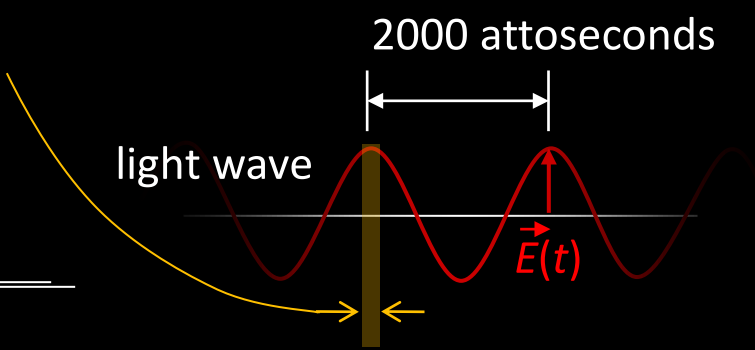
$F = m \cdot a$



constant acceleration for 200 attoseconds:

velocity $v = 1.4 \cdot 10^7$ m/s

= 5% c (c: speed of light)



Electron in Laser Field

$$E(t)=E_0\cos(\omega t)$$ linearly polarized along x axis

acceleration $$a(t)= -\frac{eE_0}{m}\cos(\omega t)$$

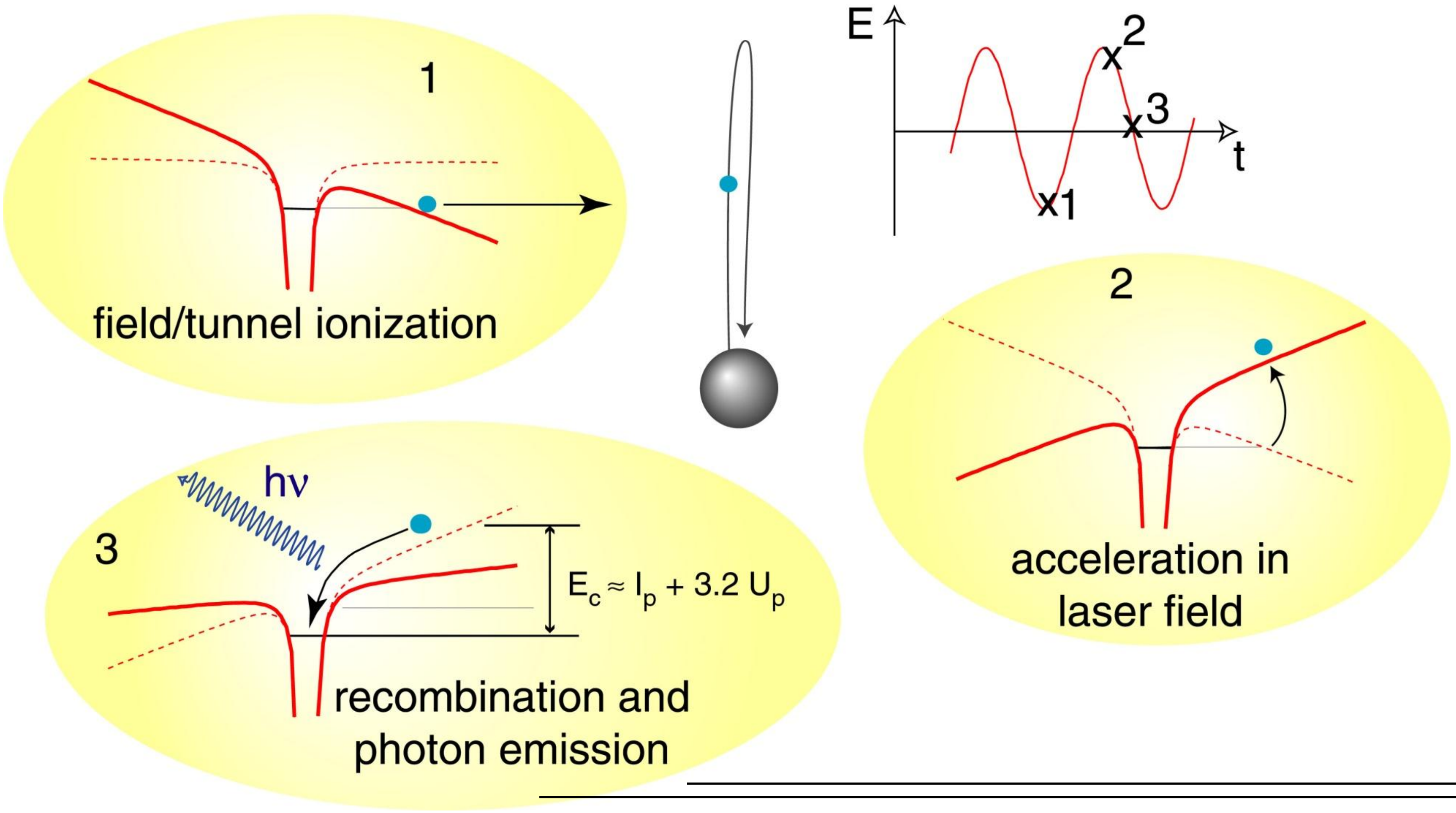
velocity (∫dt a) $$v(t)= -\frac{eE_0}{m\omega}\sin(\omega t)$$

position (∫dt v) $$x(t)= \frac{eE_0}{m\omega^2}\cos(\omega t)$$

ponderomotive potential $$U_p=E_{\mathrm{kin,av}}= \frac{e^2E_0^2}{4m\omega^2} = I\lambda^2\times 9.33\ \frac{\mathrm{eV}}{\mu\mathrm{m}^2 10^{14}\ \mathrm{W/cm}^2}$$

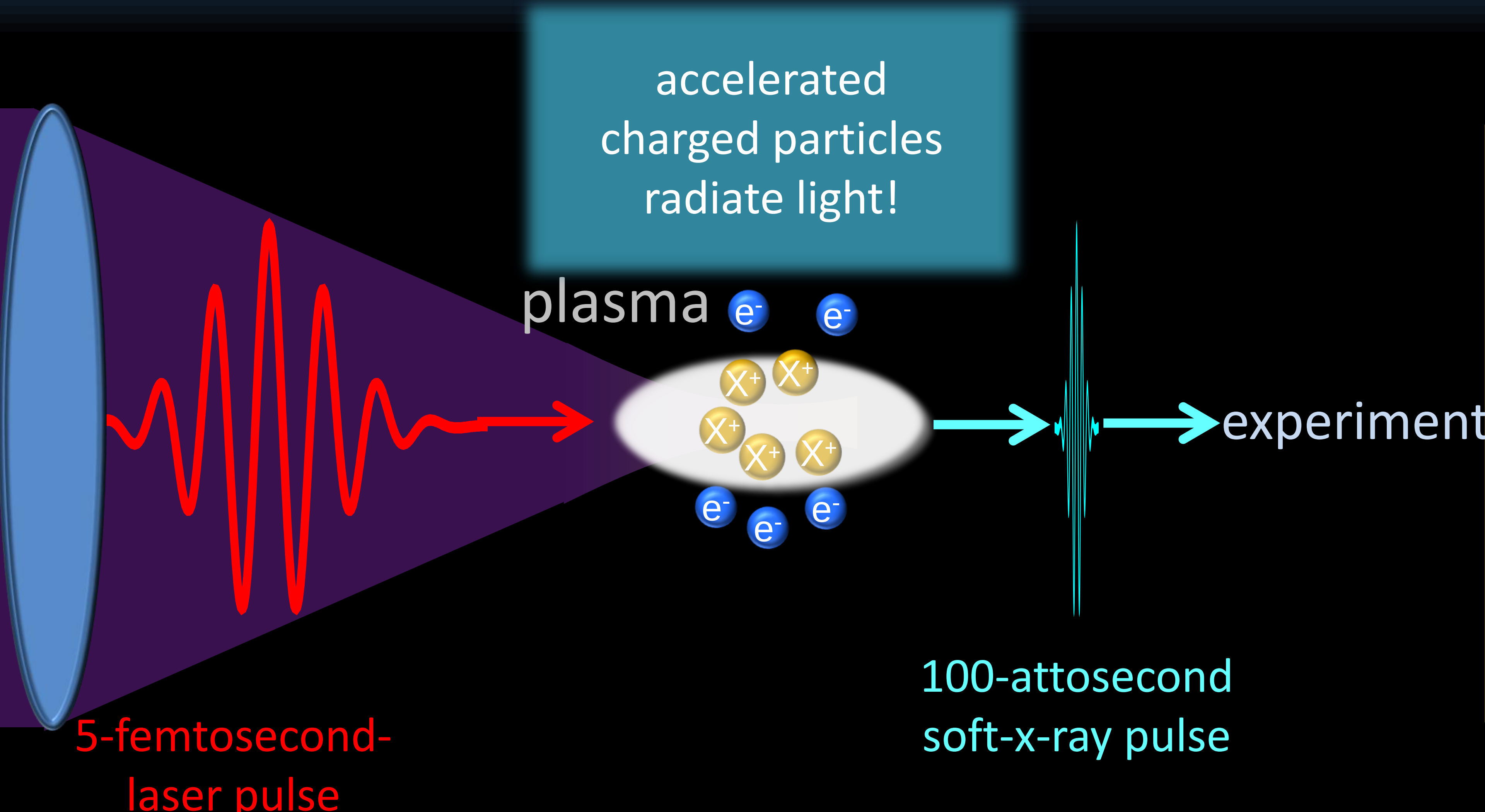
ponderomotive radius $$a_p= x_0 =\frac{eE_0}{m\omega^2}$$

Three-step model



P. Corkum, Phys. Rev. Lett. 71, 1994 (1993)
Kulander *et al.* Proc. SILAP, 95 (1993)

attosecond pulse generation



Röntgen-"X"-Rays

Wilhelm C. Röntgen

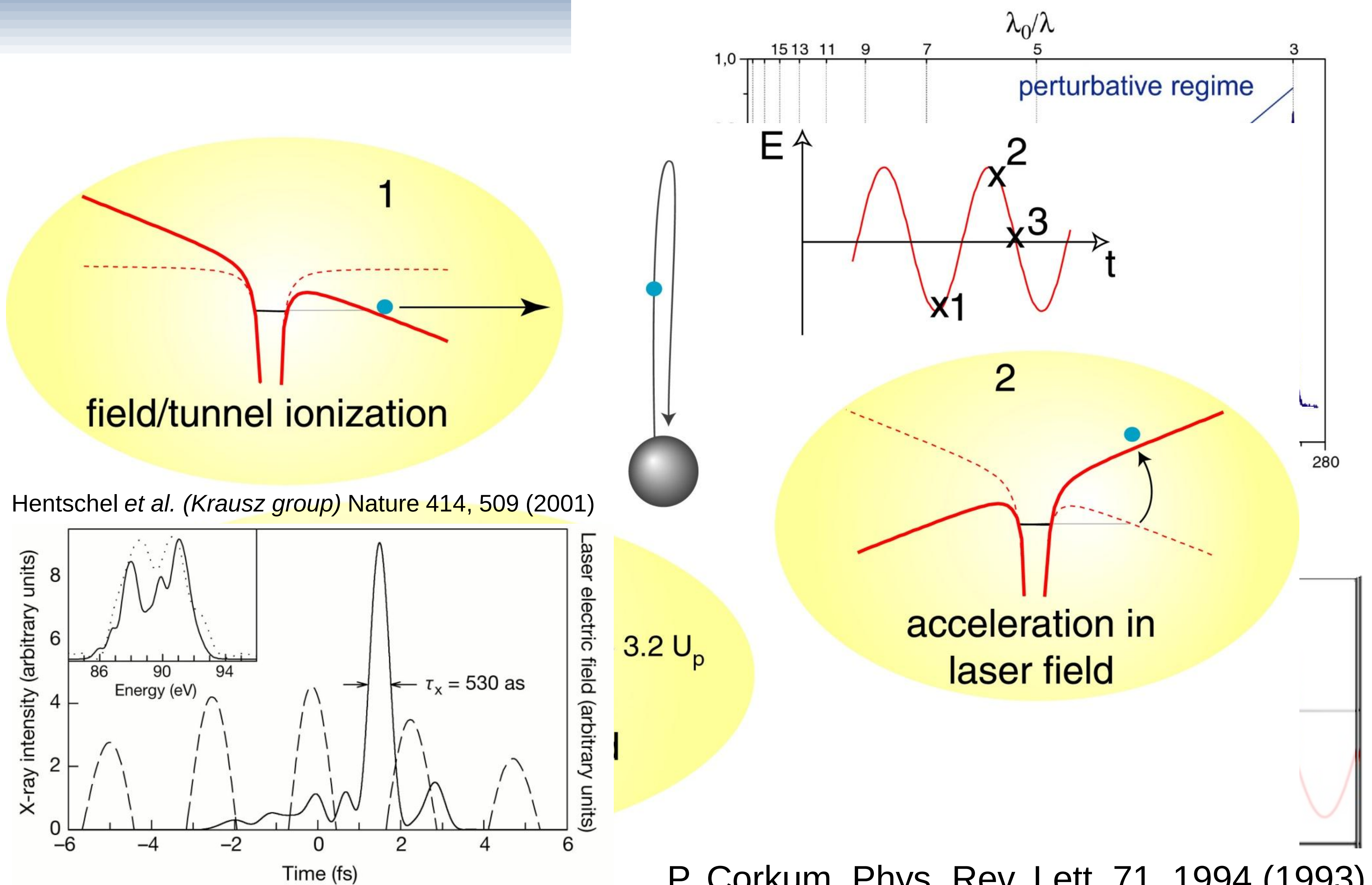
<u>1901, Physics</u>

"... in recognition of the extraordinary services
he has rendered by the discovery of
the remarkable rays subsequently named after him."

$$\text{speed of light } c = \frac{\lambda \text{ (wavelength)}}{T \text{ (optical cycle)}}$$

high spatial resolution comes with high temporal resolution

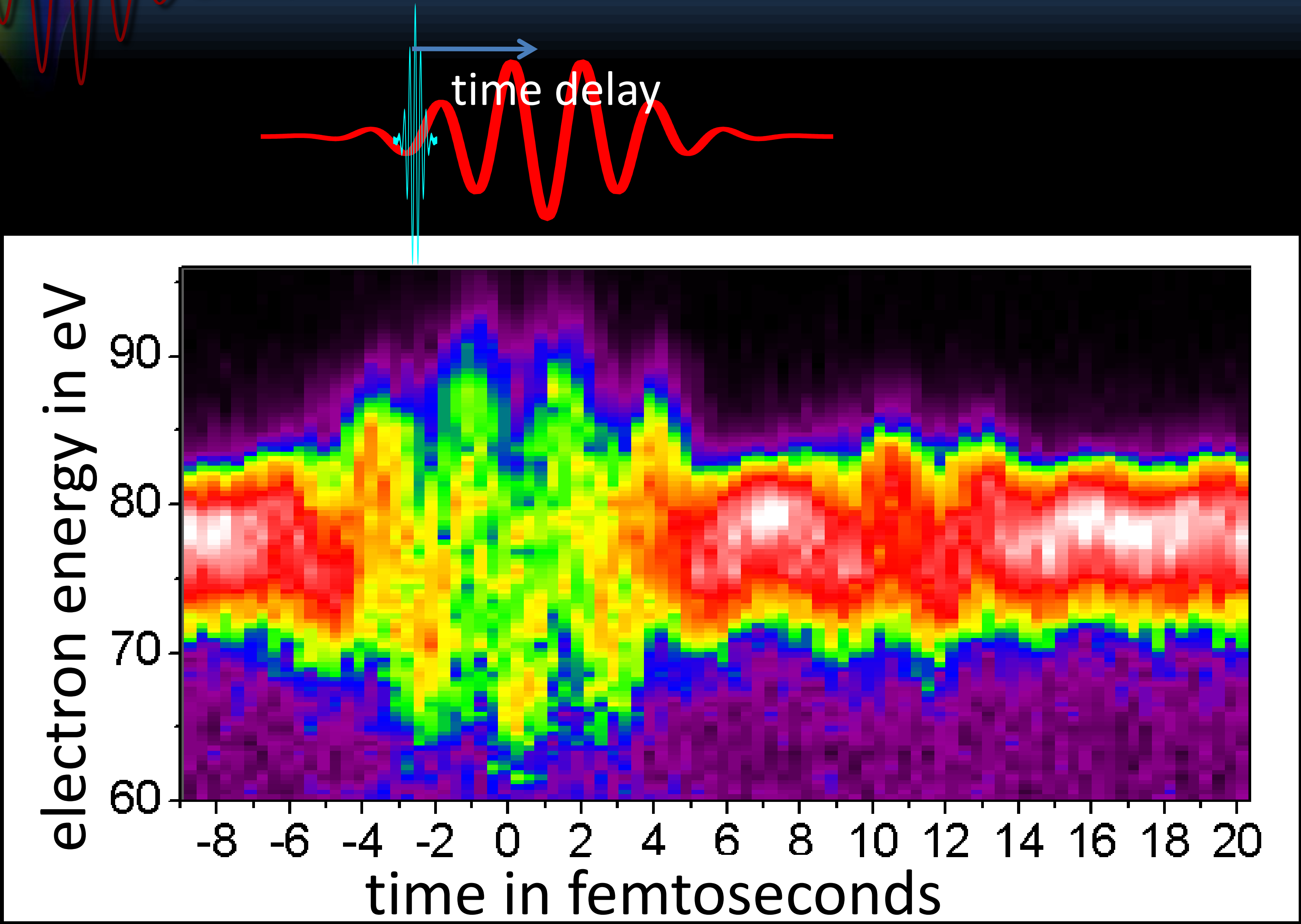
High-harmonic generation



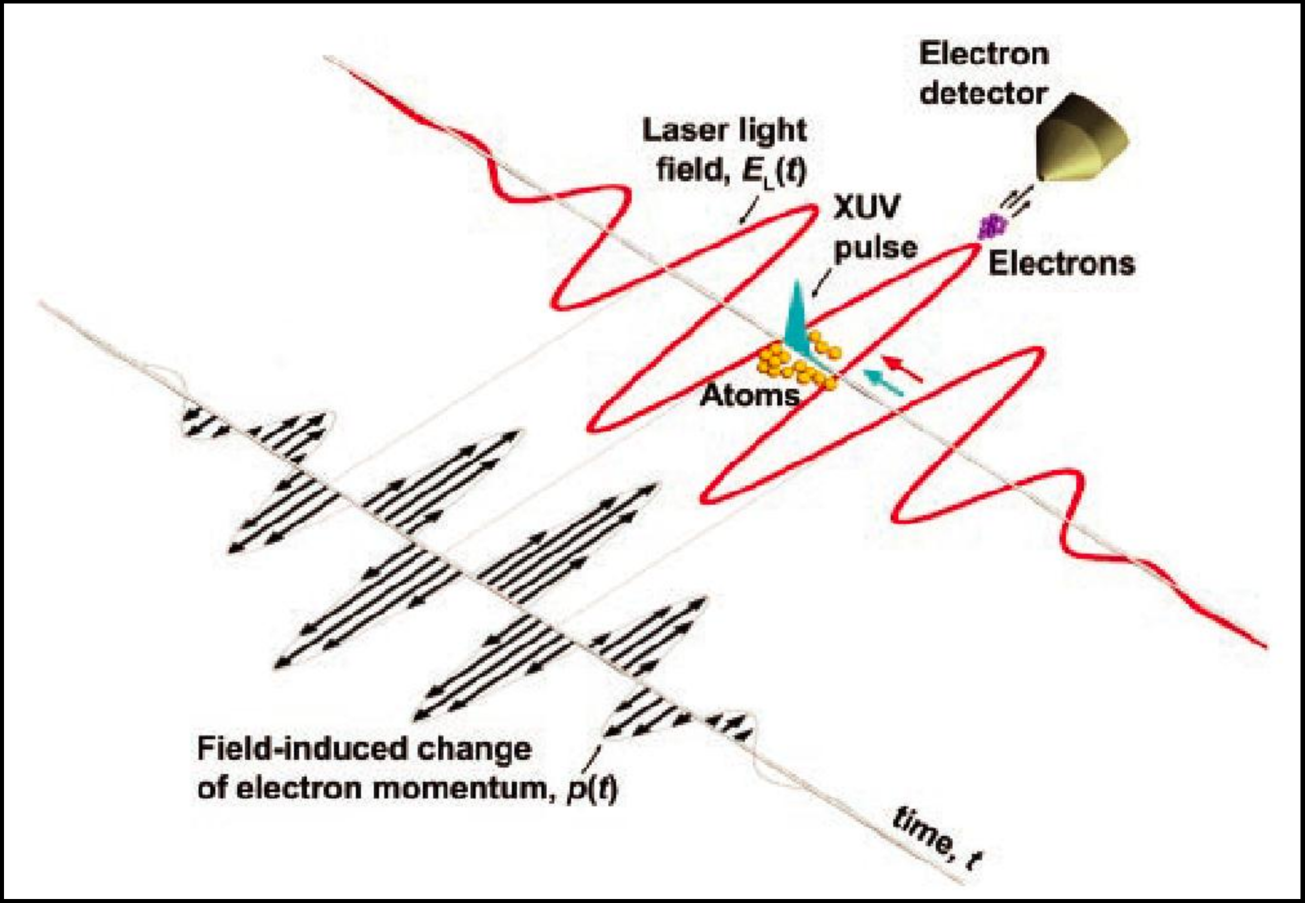
P. Corkum, Phys. Rev. Lett. 71, 1994 (1993)

Science with Attosecond Pulses

measurement of light waves
(a very fast oscilloscope)



Streaking

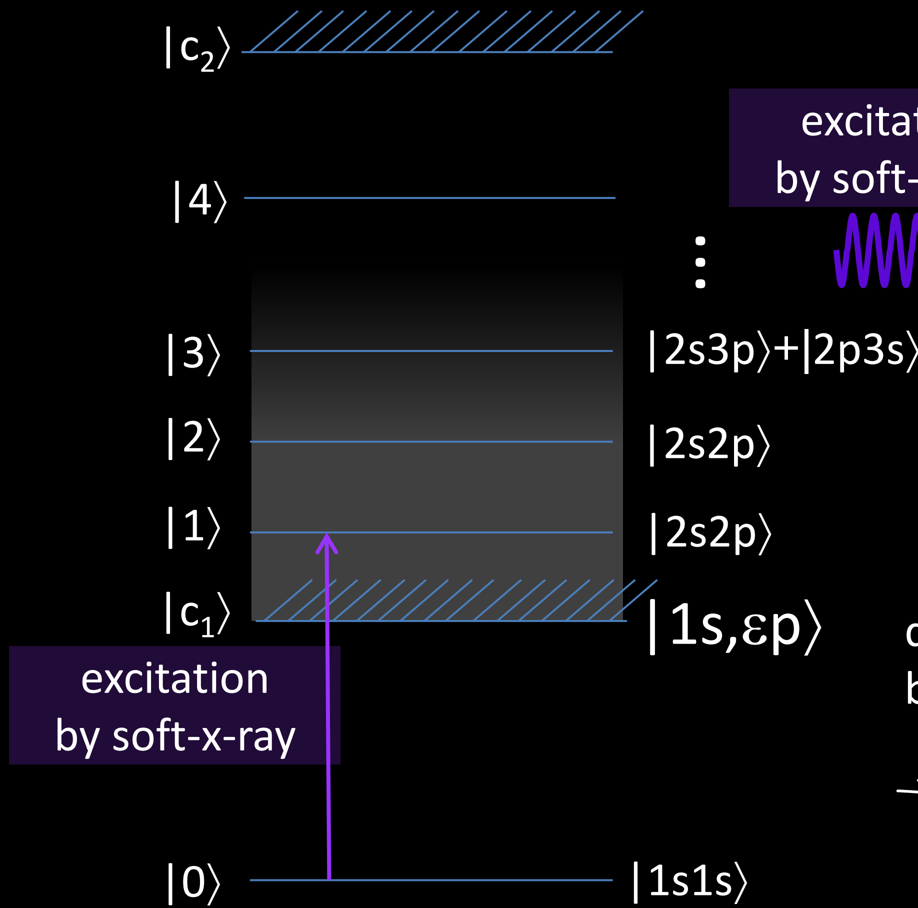


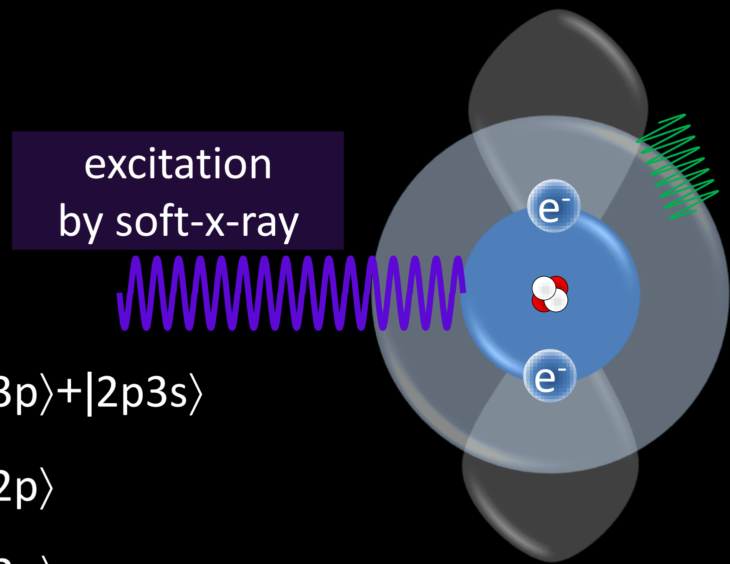
Goulielmakis *et al. (Krausz group)*, Science **305**, 1267 (2004)

fundamental e⁻–e⁻ interaction

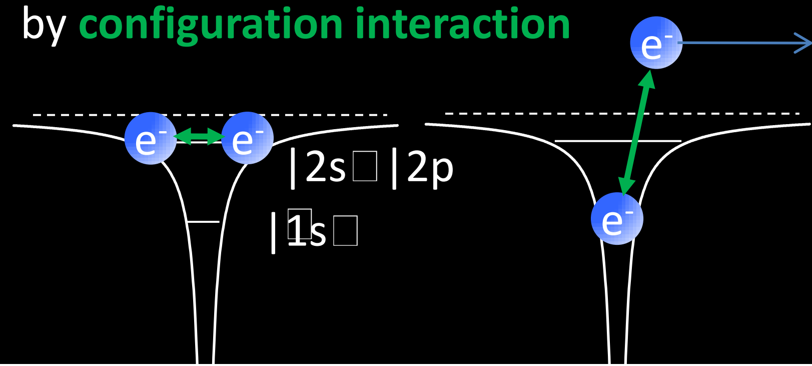
prototypical example:

the Helium atom





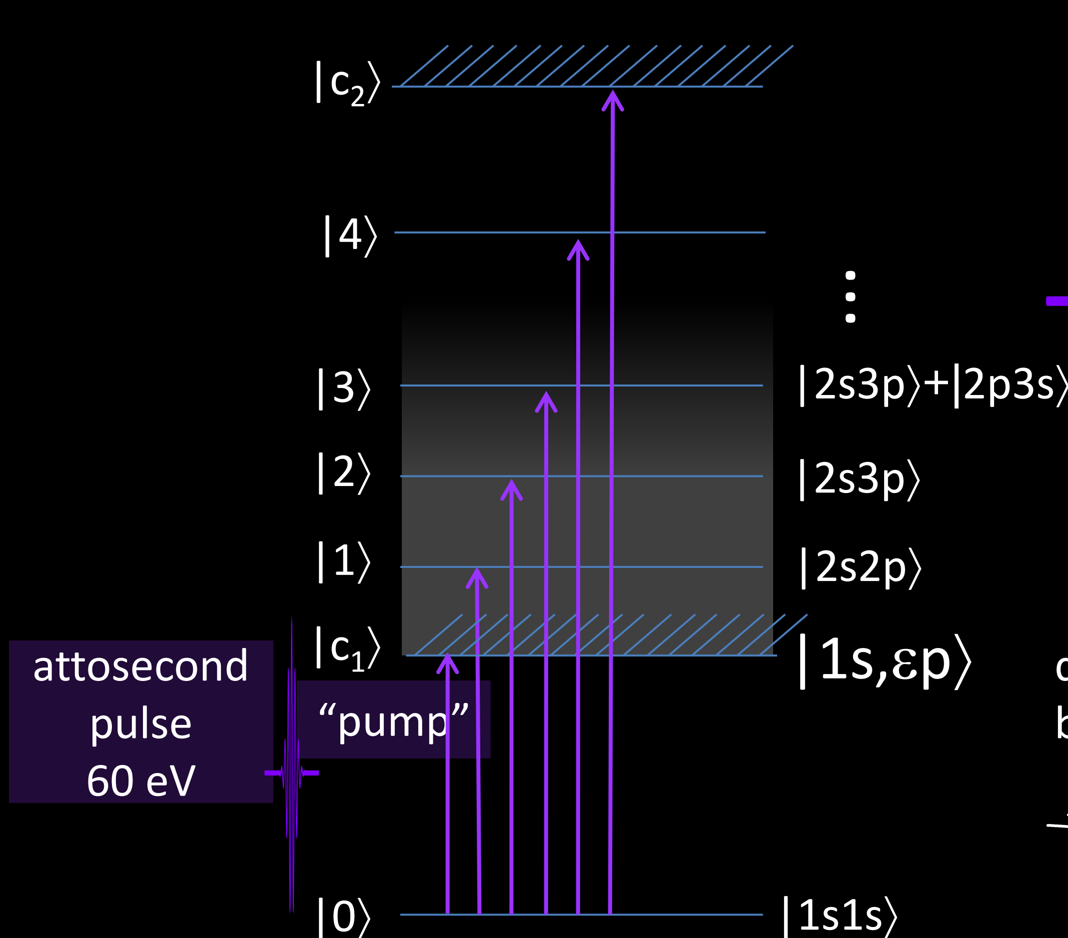
discrete states coupled to continuum by **configuration interaction**

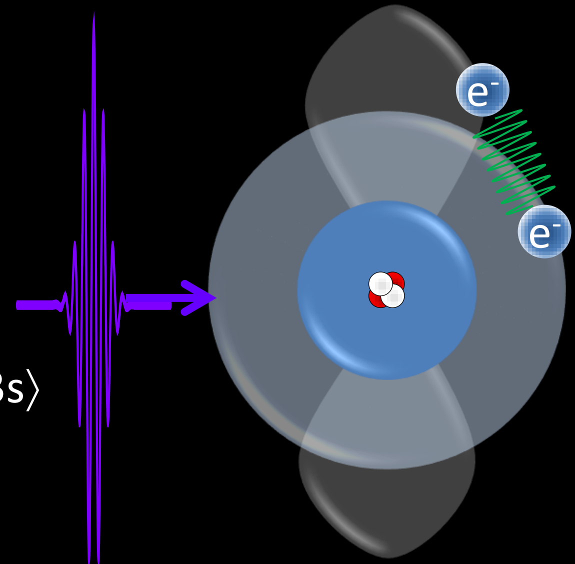


fundamental e^-–e^- interaction

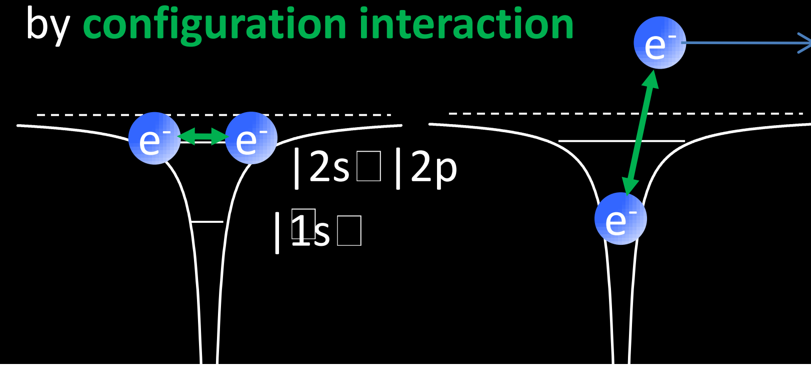
prototypical example:

the Helium atom





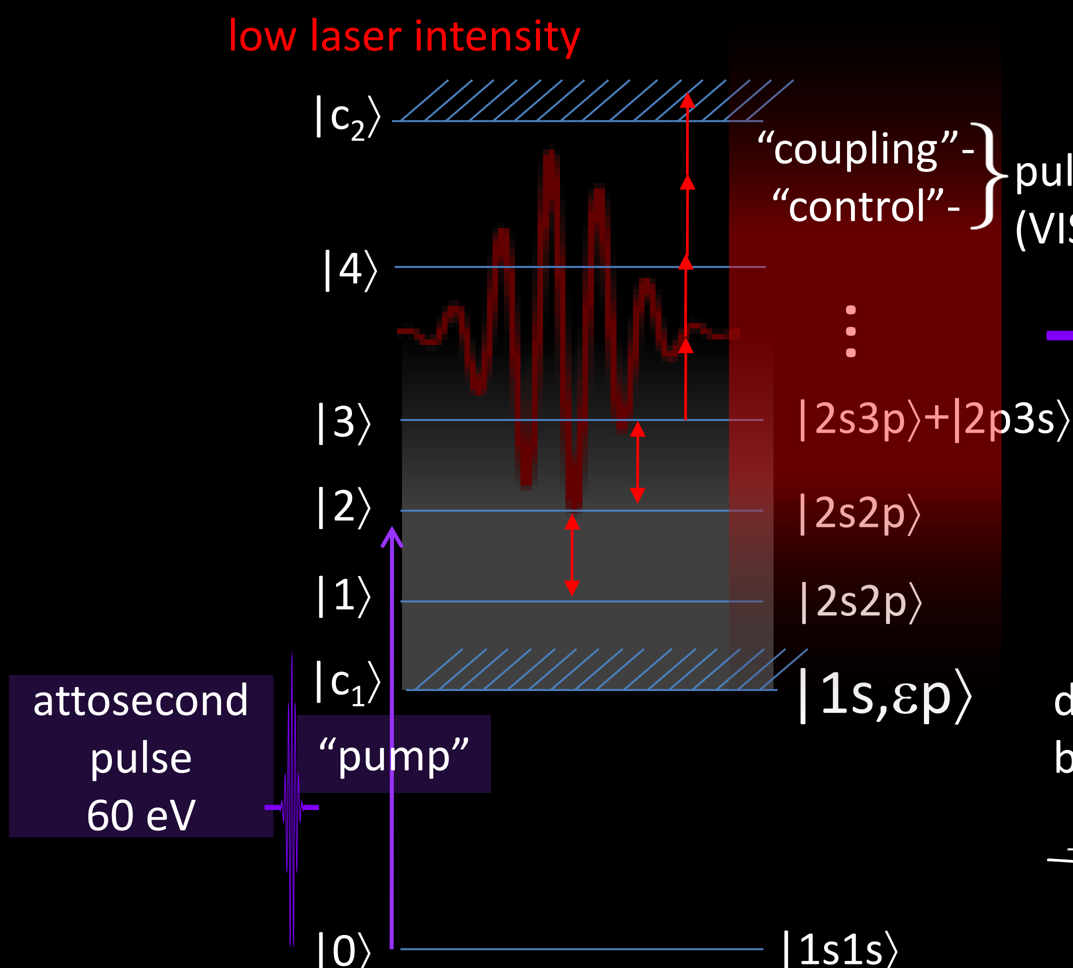
discrete states coupled to continuum by **configuration interaction**

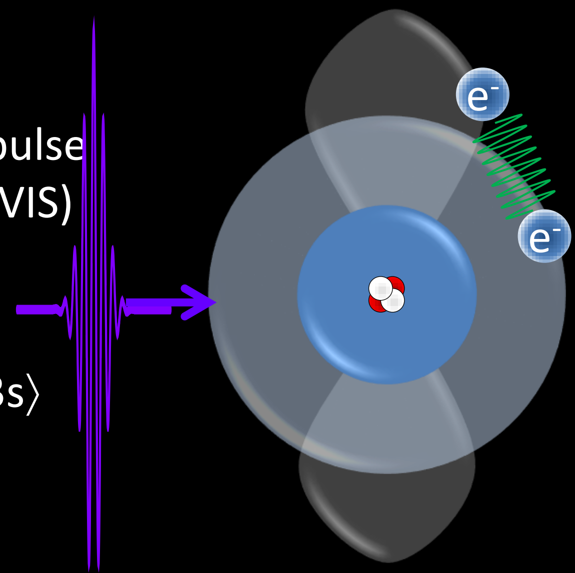


$2e^-$ measurement scheme

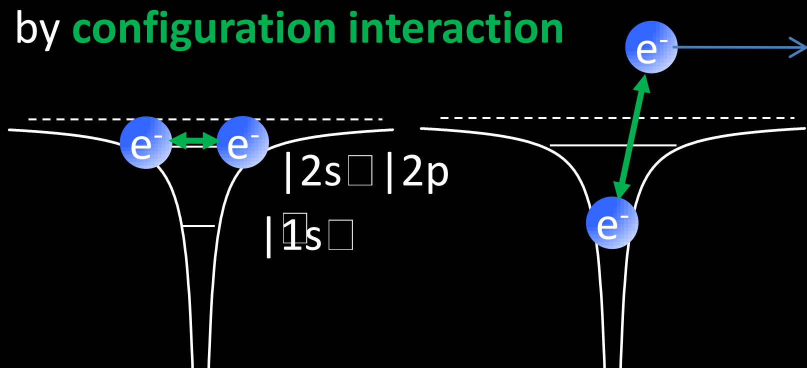
prototypical example:

the Helium atom



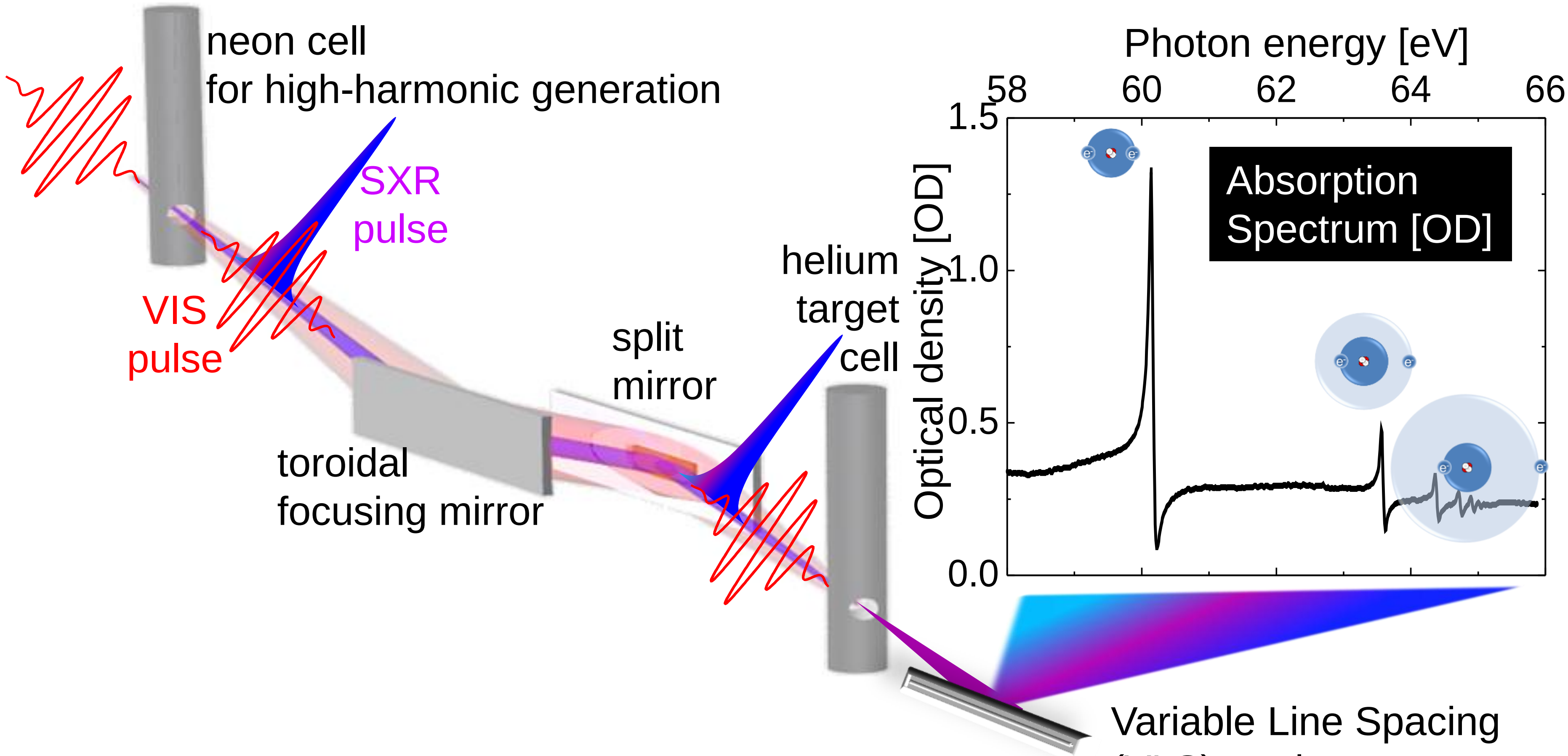


discrete states coupled to continuum by **configuration interaction**



C. Ott *et al.*, submitted (arXiv:1205.0519v1)

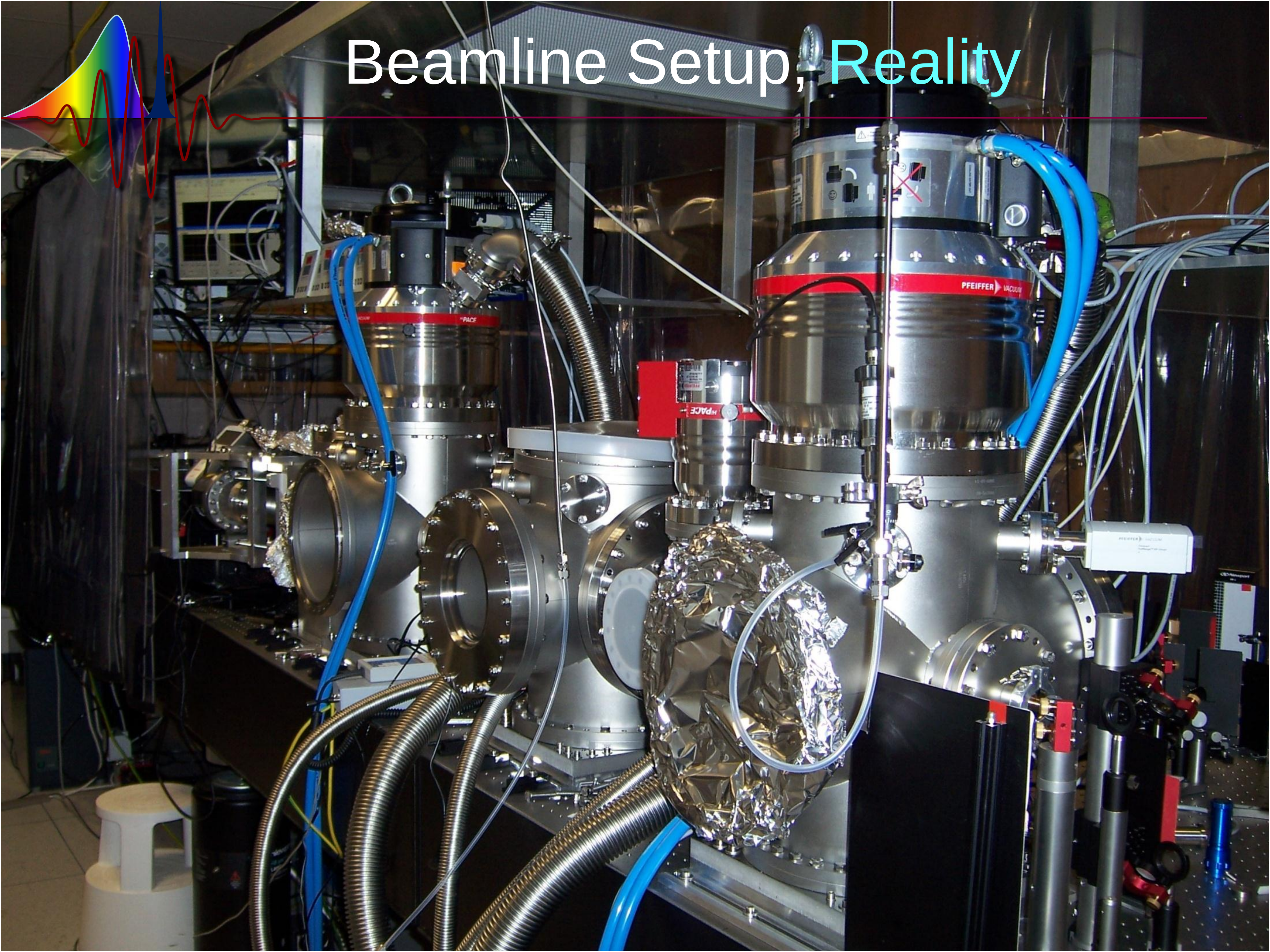
Schematic Setup



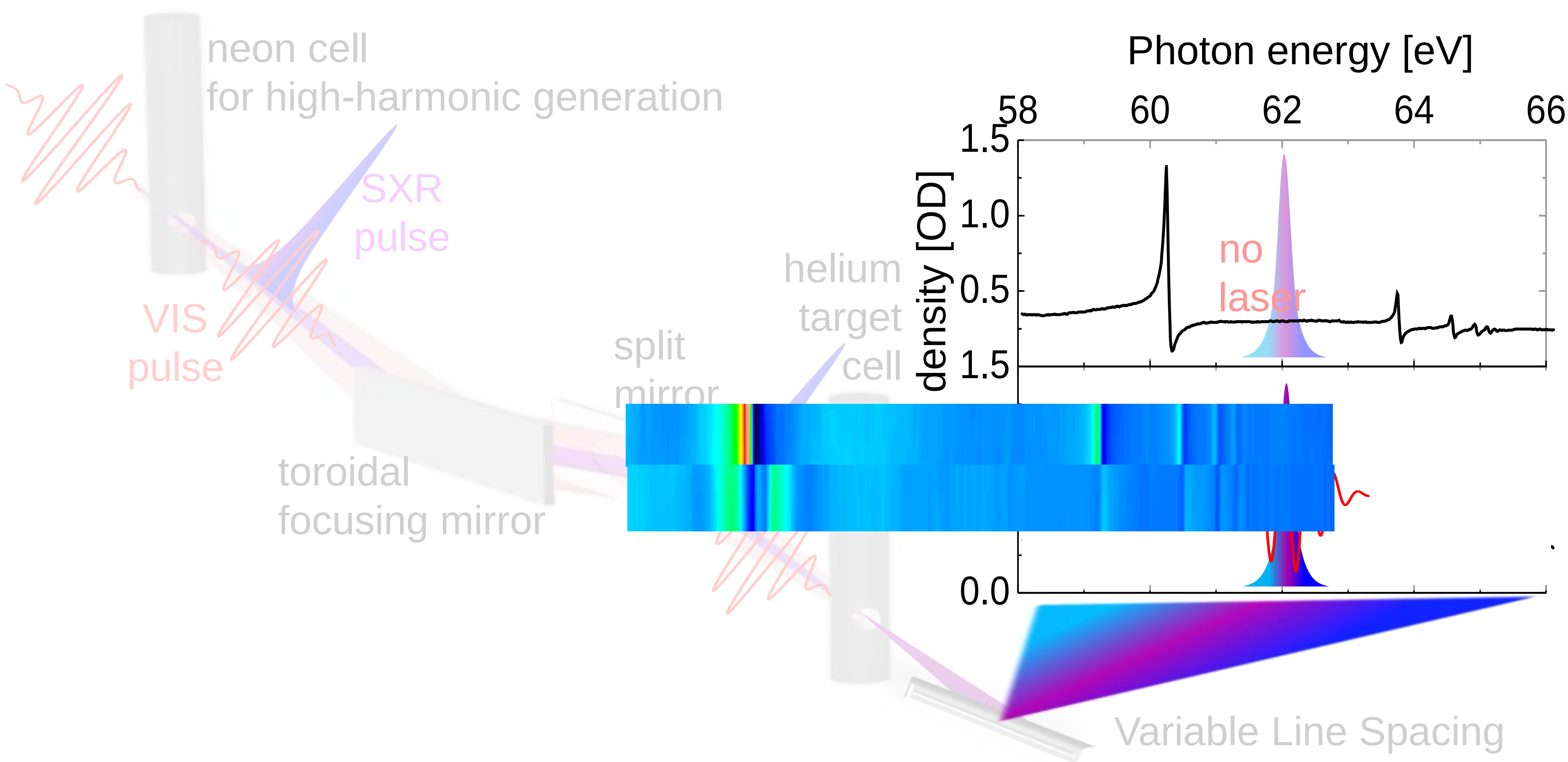
Resolution: $\Delta\tau$ = 10 as
ΔE = 20 meV (s.d. @60 eV)

C. Ott *et al.*, submitted (arXiv:1205.0519v1)

Beamline Setup, Reality

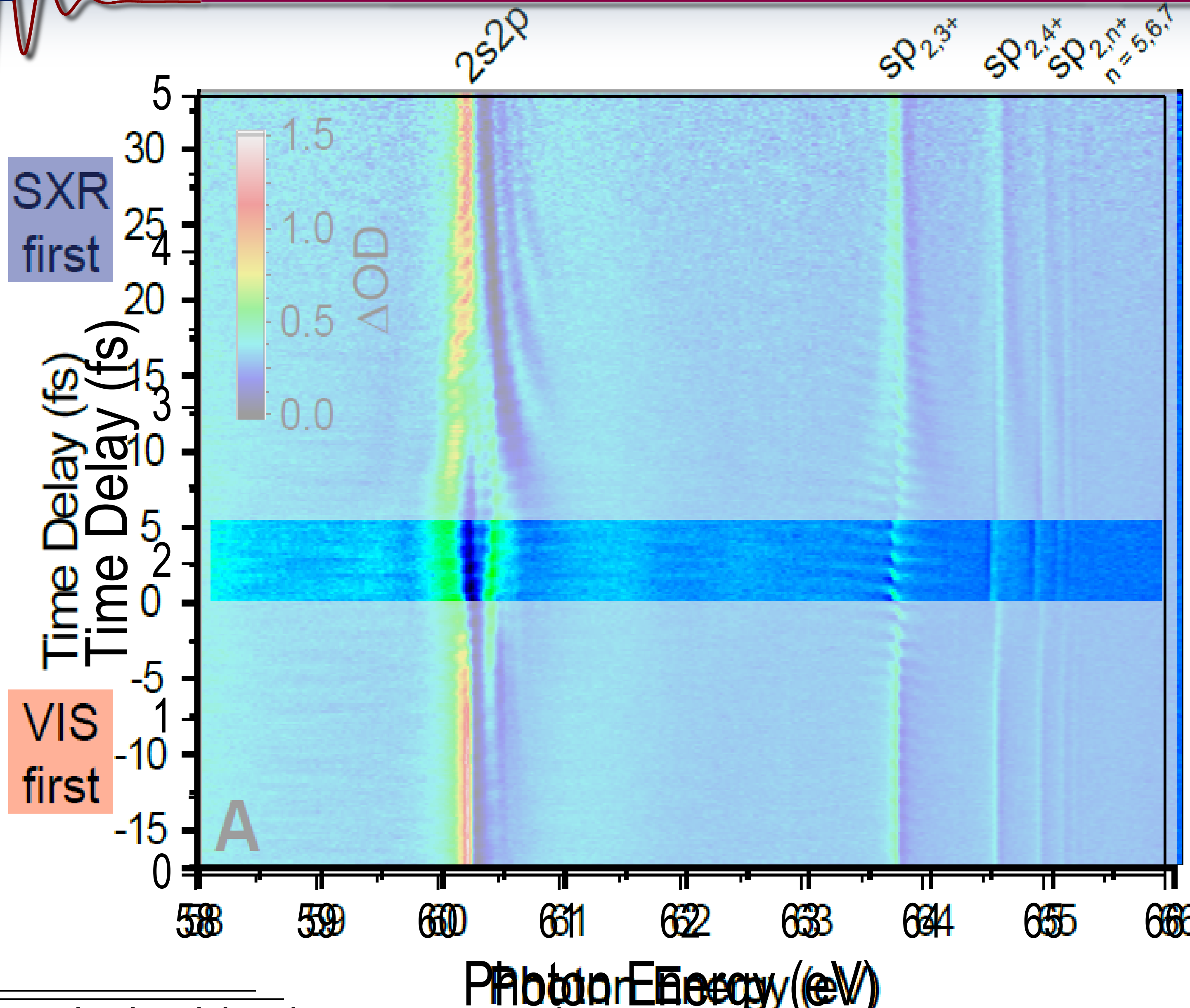


Schematic Setup



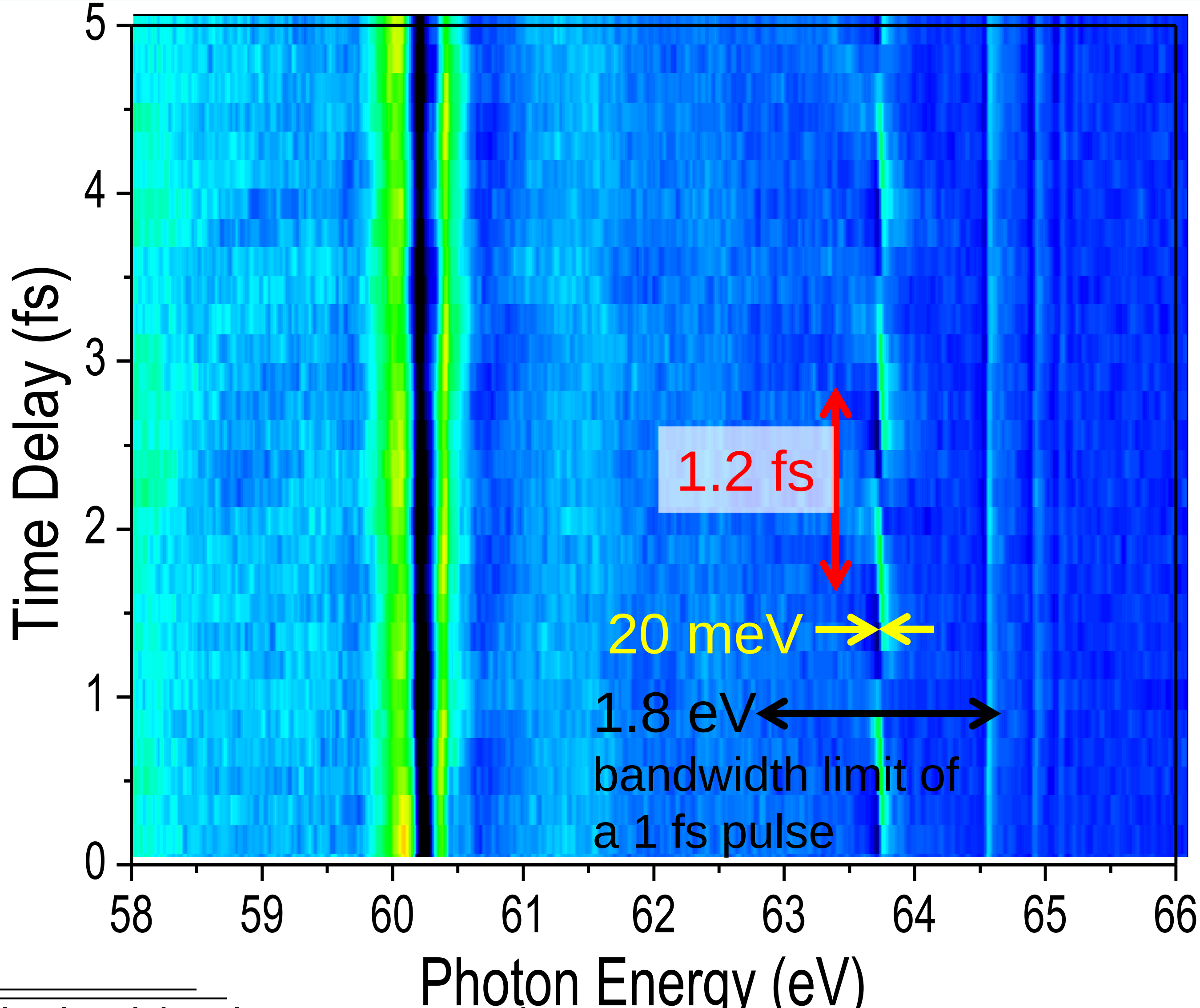
Resolution: Δτ = 10 as
ΔE = 20 meV (s.d. @60 eV)

Experimental Results



C. Ott *et al.*, submitted (arXiv:1205.0519v1)

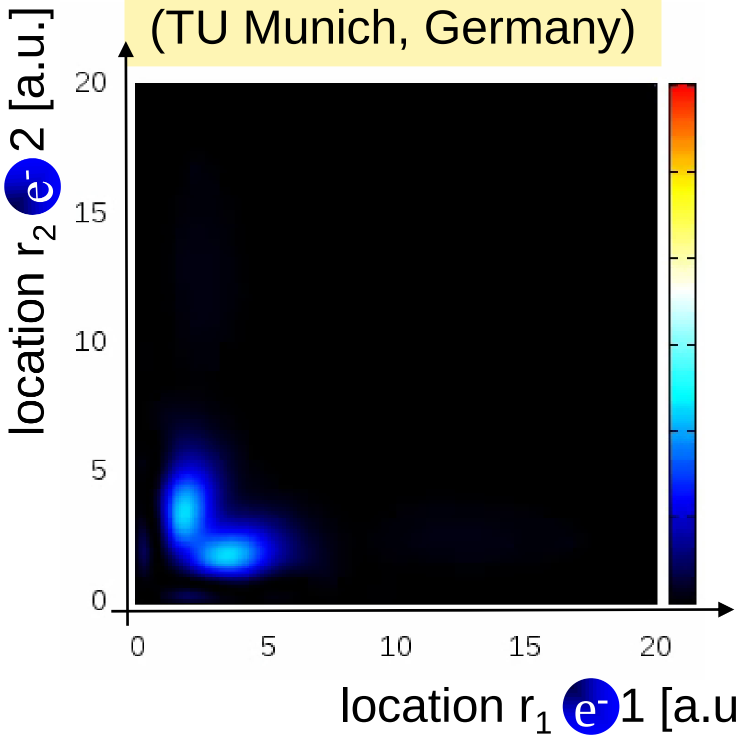
Experimental Results



C. Ott *et al.*, submitted (arXiv:1205.0519v1)

$2e^-$ wavepacket... and application

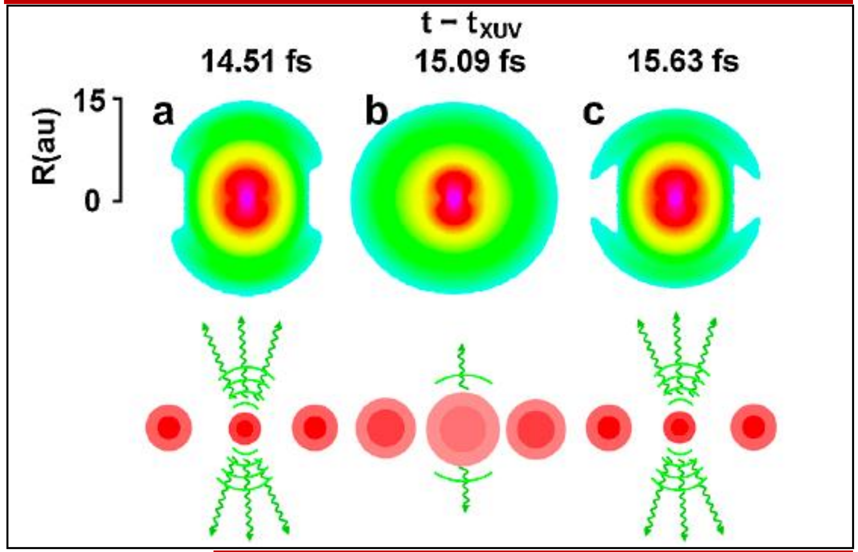
Reconstruction
with Theory support by
Javier Madroñero
(TU Munich, Germany)



reached goal of observing **natural** two-electron quantum dynamics with only **weak** laser fields

Experimentally realized:
A controlled **1-nm** source of pulsed electrons with a clock speed of **1 PHz(10^{15} Hz, 1 Mio.GHz)**

In such coherent superpositions:
Autoionization proceeds in 1-fs bursts



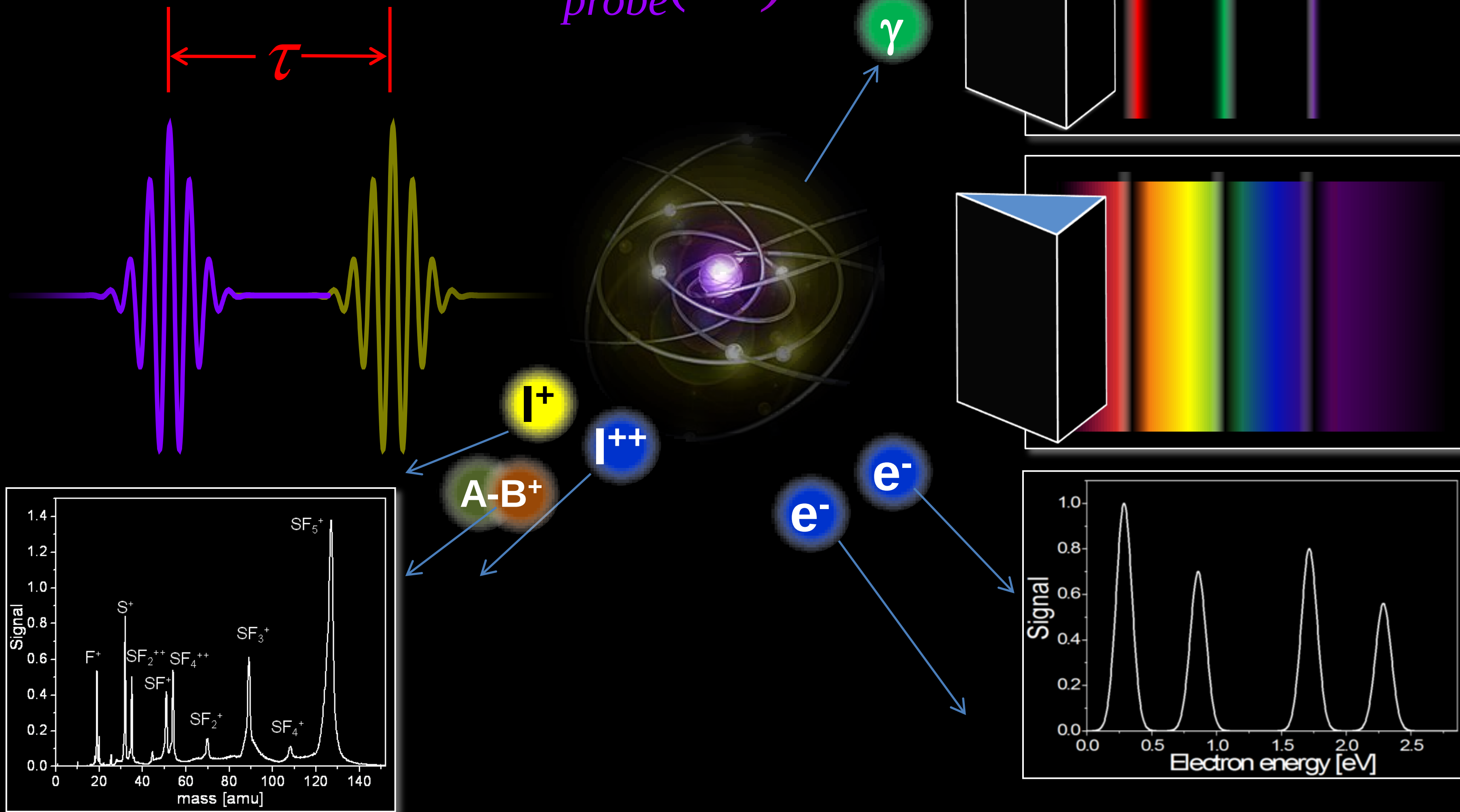
Argenti and Lindroth (Theory),
Phys. Rev. Lett. **105**, 053002 (2010)

What's more: an **application** may be on the horizon:

time-resolved spectroscopy

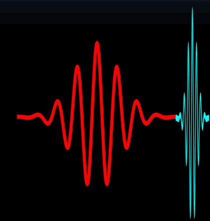
$$H(t)=H_0+rE_{pump}(t)$$
$$+rE_{probe}(t-\tau)$$

time delay



Summary

Short, controlled, coherent flashes of light



- Mathematical concepts/representation of pulses
- Generation of pulses
- Measurement of pulses
- Scientific Applications (of femto- and attosecond pulses):
  - time-resolved measurement and control
    of fast intra-atomic/-molecular quantum processes
    (e.g. electron dynamics, molecular wavepackets)

fundamental/basic research, but understanding important for:

- molecular electronics [PHz vs. GHz, 5-6 orders of magnitude]
  for extremely small and fast computers, devices
- understanding the primary steps in biological processes
  e.g. photosynthesis, vision, radiation damage