Reproducible Generation and Measurement of Isolated Sub-fs XUV Pulses with Phase Controlled Few-cycle Light

Reinhard Kienberger[1,2,(4)]

E. Goulielmakis[1], M. Uiberacker[1], A. Baltu_ka[1], M. Drescher[1,3]
and Ferenc Krausz[1,4]

1 Institut für Photonik, Technische Universität Wien, Austria
2 SLAC, Stanford Linear Accelerator Center, Menlo Park, CA, USA
3 Fakultät für Physik, Universität Bielefeld, Germany
4 Max-Planck Institut für Quantenoptik, Garching/München, Germany

ULTRAFAST X-RAYS 2004
April 30, 2004
San Diego, CA, USA

Coworkers & Cooperations

Theory:

A. Scrinzi, V. Yakovlev, TU Vienna

XUV optics & spectroscopy (1999-present):

M. Drescher, U. Kleineberg, Y. Lim, U. Heinzmann, Univ. Bielefeld, Germany
M. Wieland, T. Wilhein, FH Koblenz, RAC Remagen, Germany

Strong-field physics (1999-present):

P. B. Corkum, M. Yu Ivanov, NRC Canada, Ottawa

Light phase control (2000-present):

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

& measurement (2001-present):

G. Paulus, F. Lindner, H. Walther, Univ. Munich - MPQ Garching, Germany

Outline

1.) The tools:

- Phase-stabilized few-opt.-cycle laser pulses
- Single as pulses: High-order Harmonic Generation

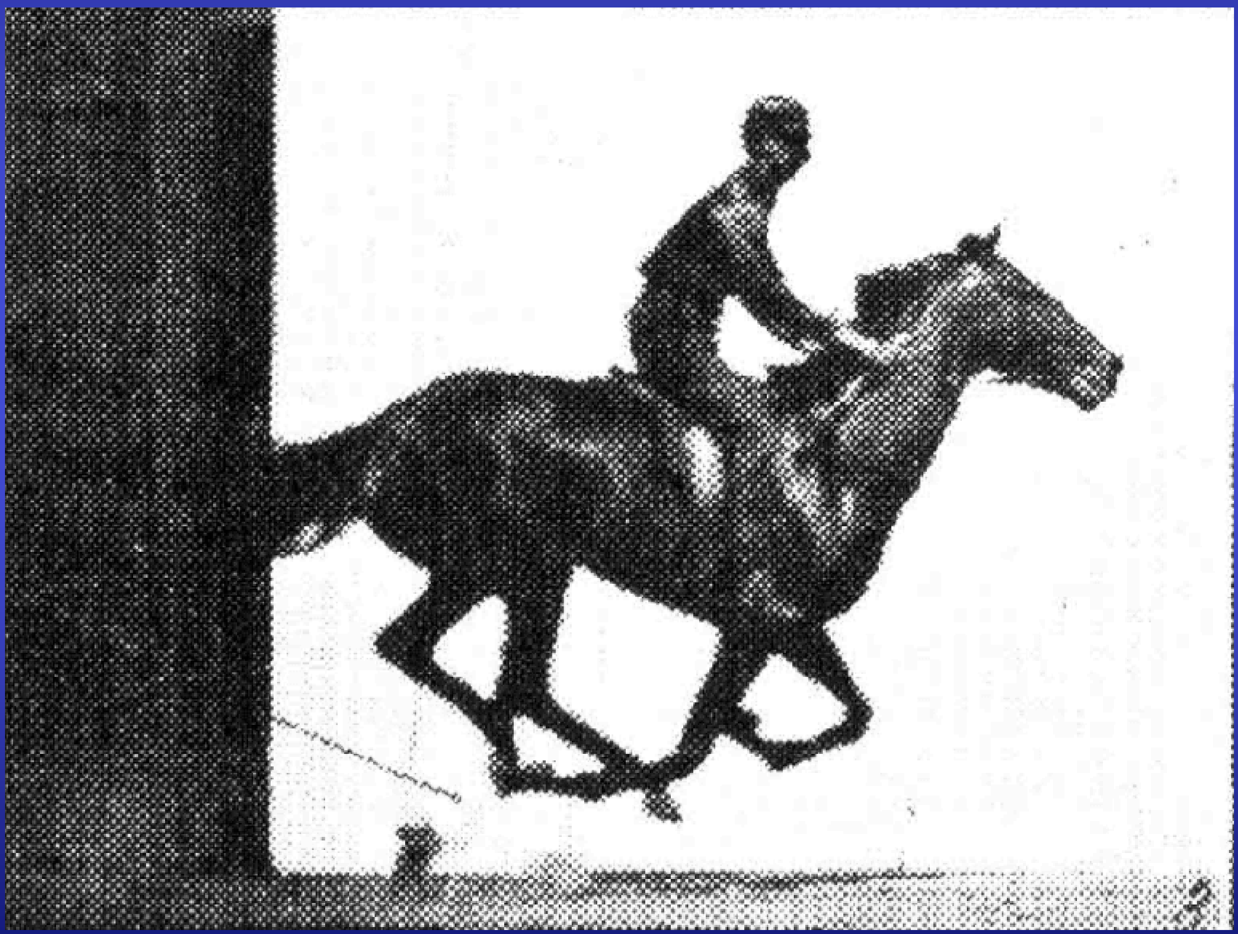
2.) Attosecond pulse measurement

- Photoelectron spectra
- Attosecond streak camera

3.) Application: Spectroscopy

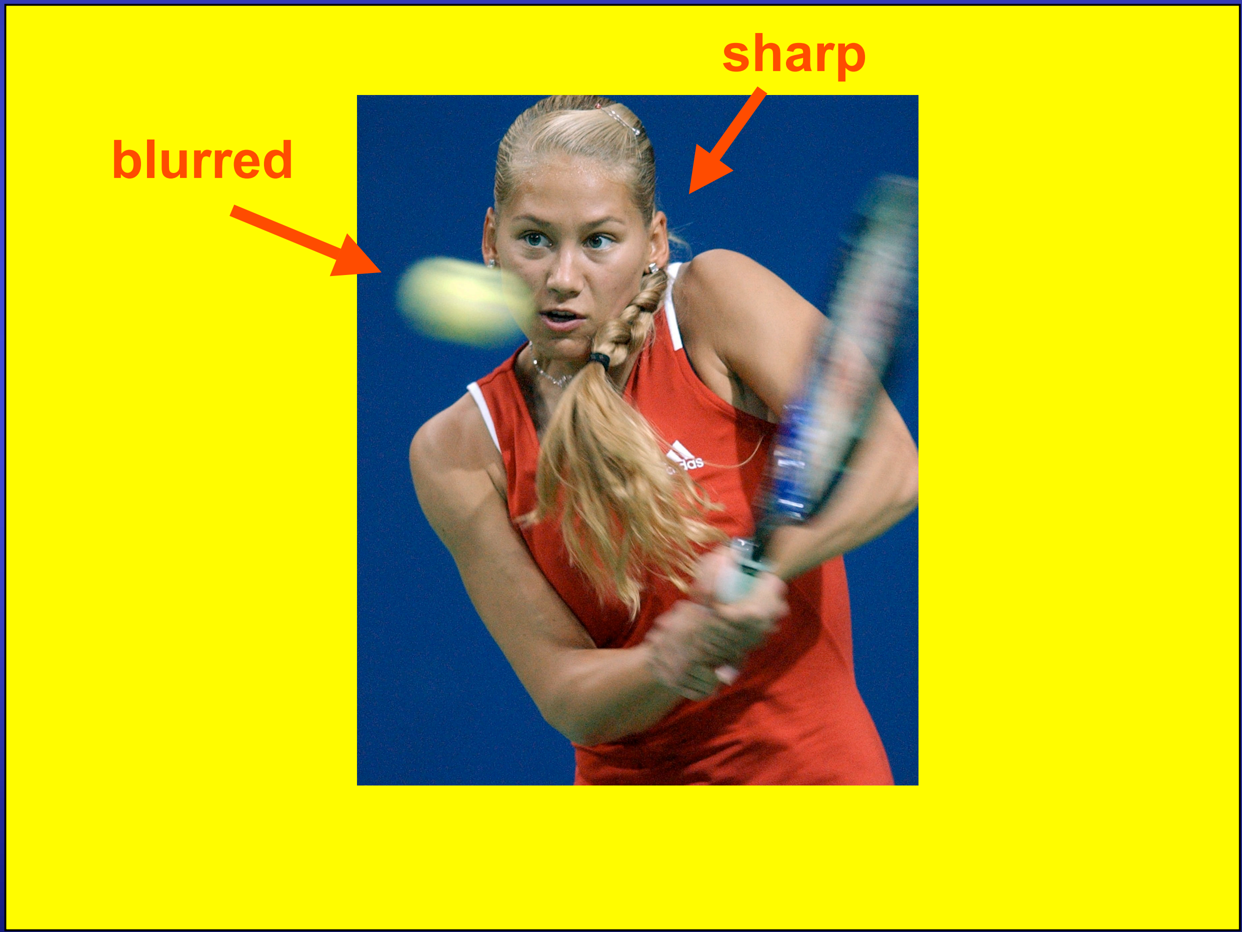
1878: E. Muybridge, Stanford

Tracing motion of animals
by spark photography



E. Muybridge, *Animals in Motion*, ed. by L. S. Brown (Dover Publ. Co., New York 1957)

Tracing motion of humans

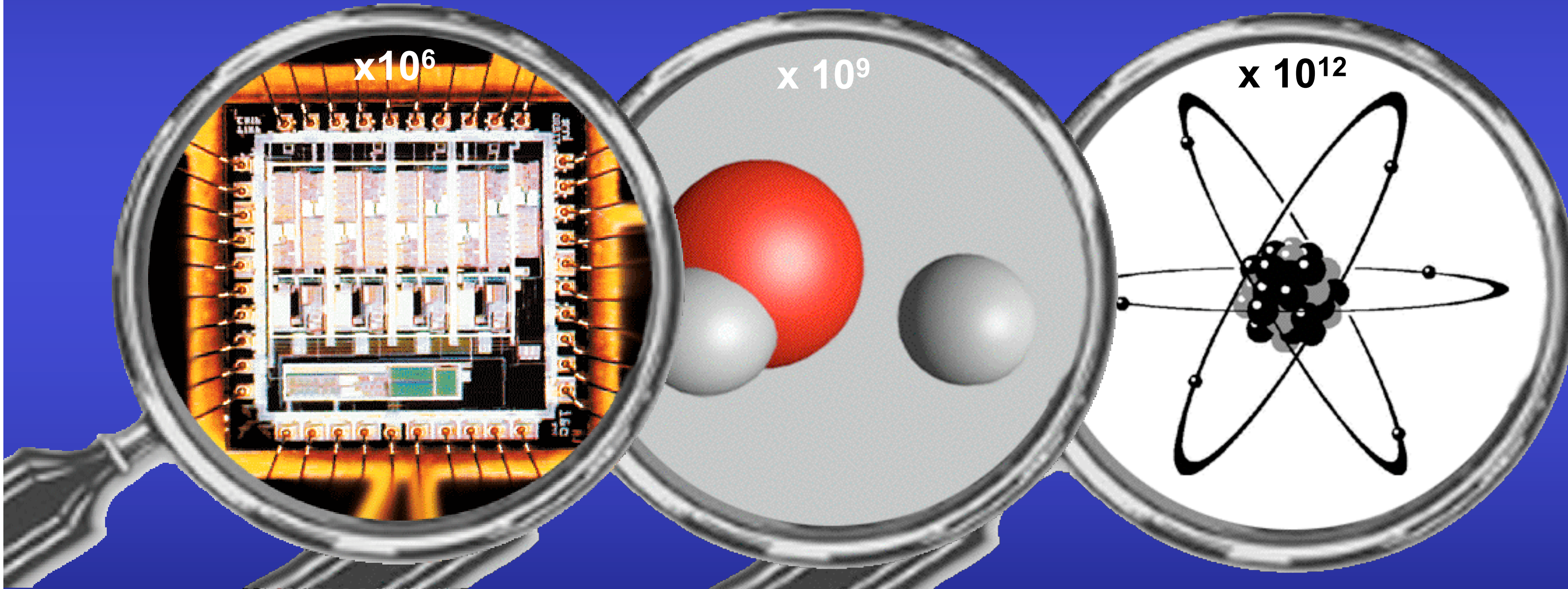


Motion in the Microcosm

micrometer — nanometer — picometer

10^{-6} m — 10^{-9} m — 10^{-12} m

Mesoscopic scale | **Molecular/atomic scale** | **Sub-atomic scale**



nanosec — picosec — femtosec — attosec

10^{-9} — 10^{-12} — 10^{-15} — 10^{-18} seconds

In solids: Carrying electric current

In molecules: Triggering chemical reactions

In atoms: Emitting X-rays

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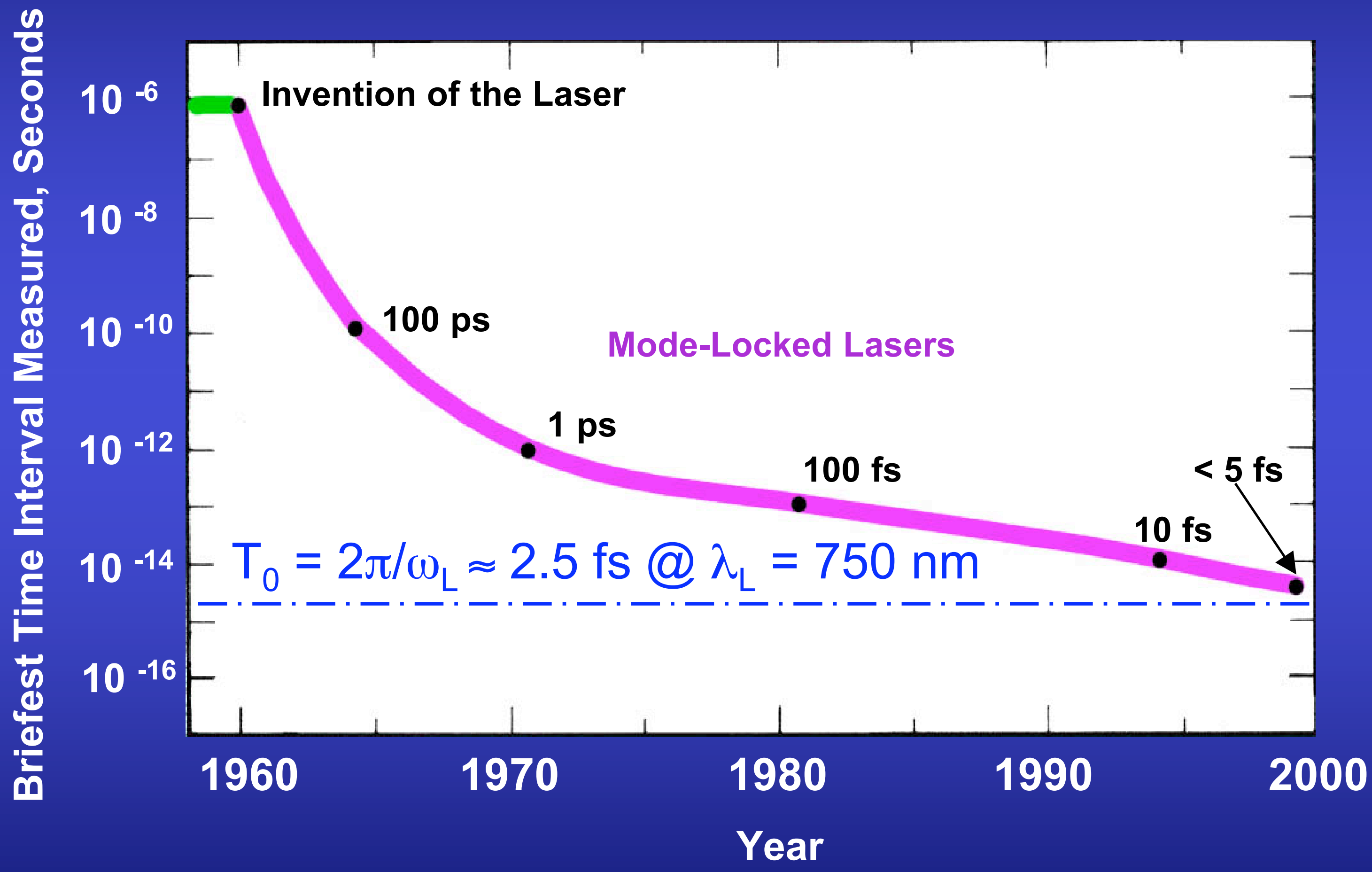
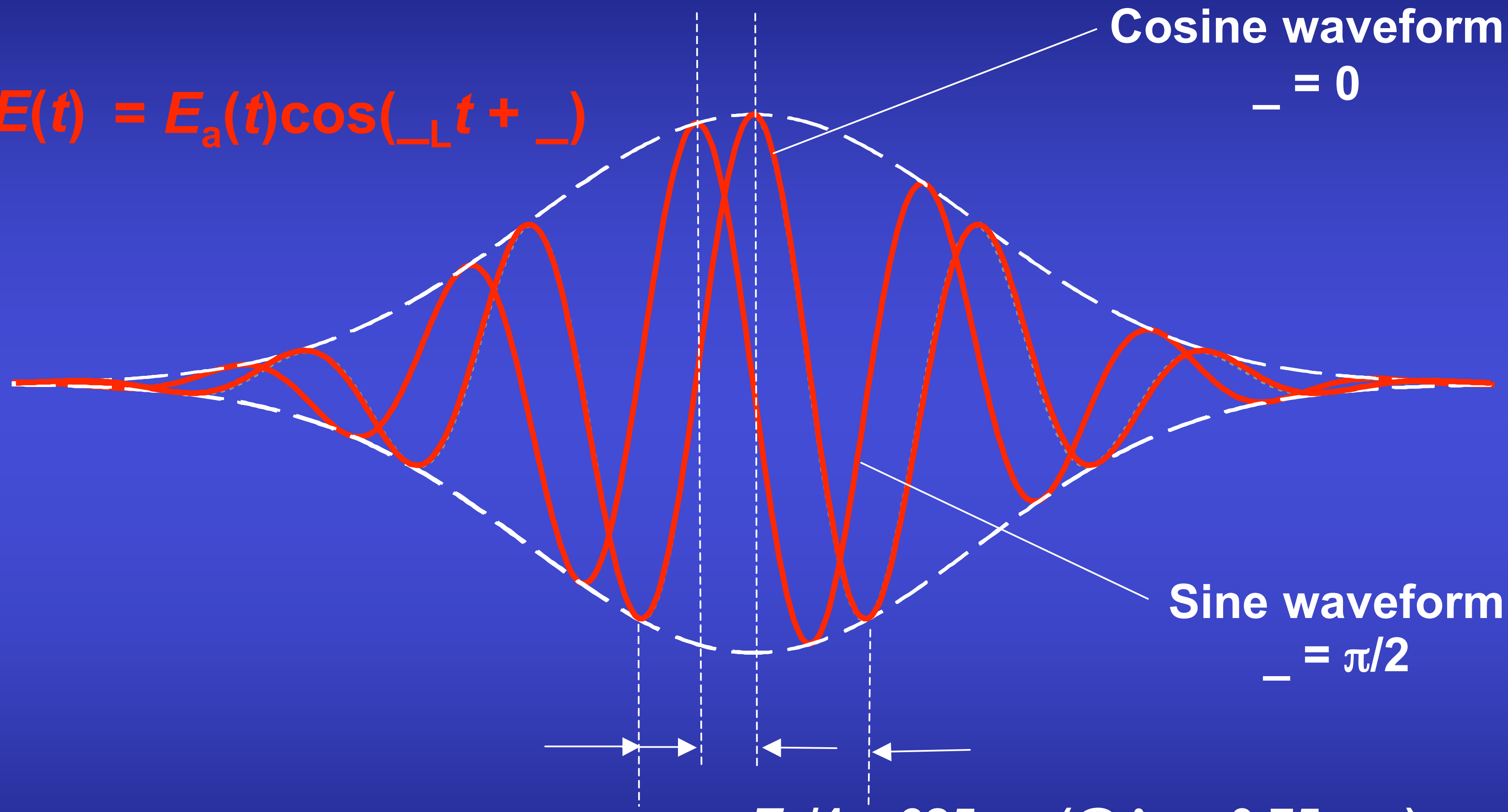
The Tools: Ultrashort Laser Pulses



Few-Cycle Laser Pulses
Control of the Waveform

$E(t) = E_a(t)\cos(\_{}_L t + \_)$

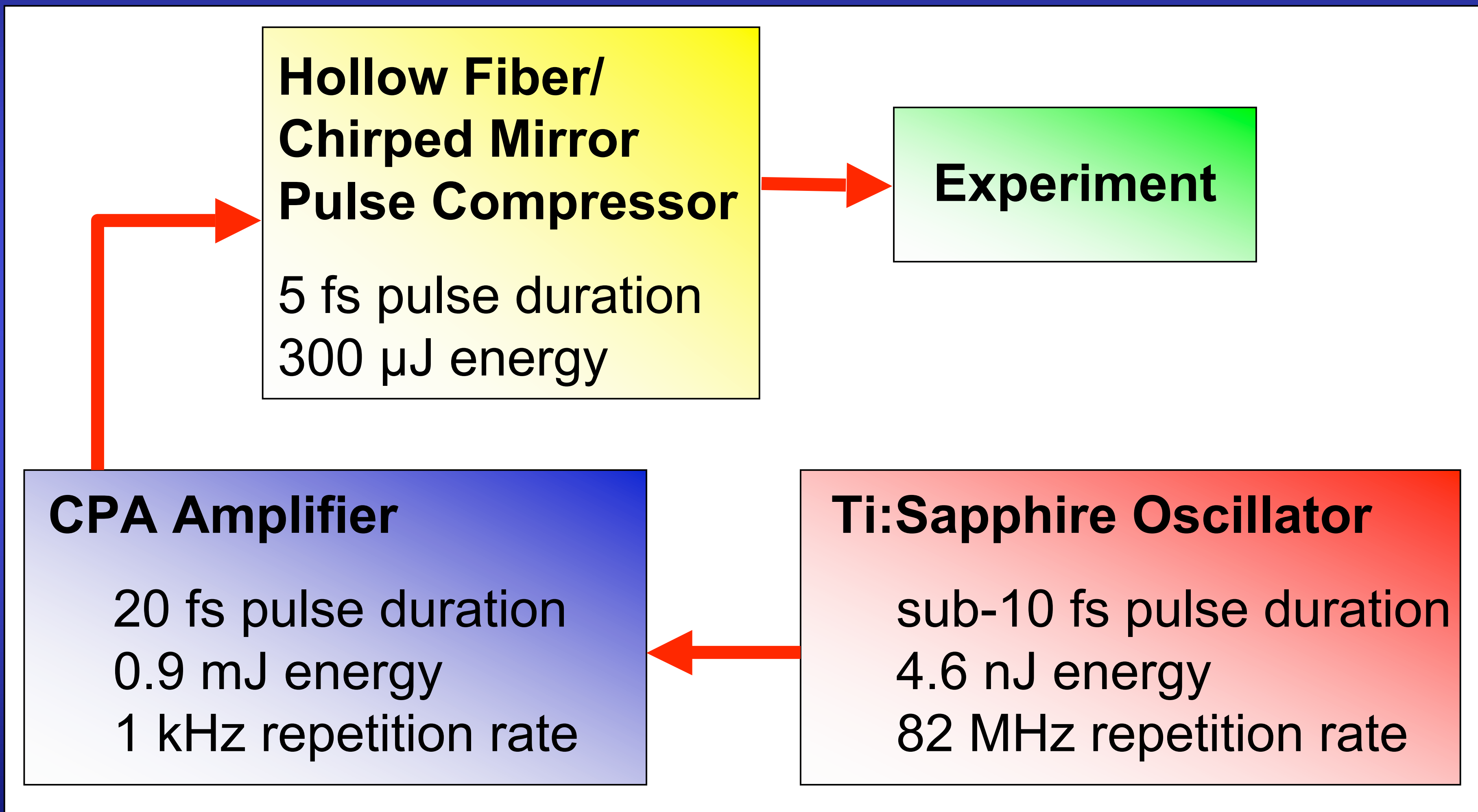


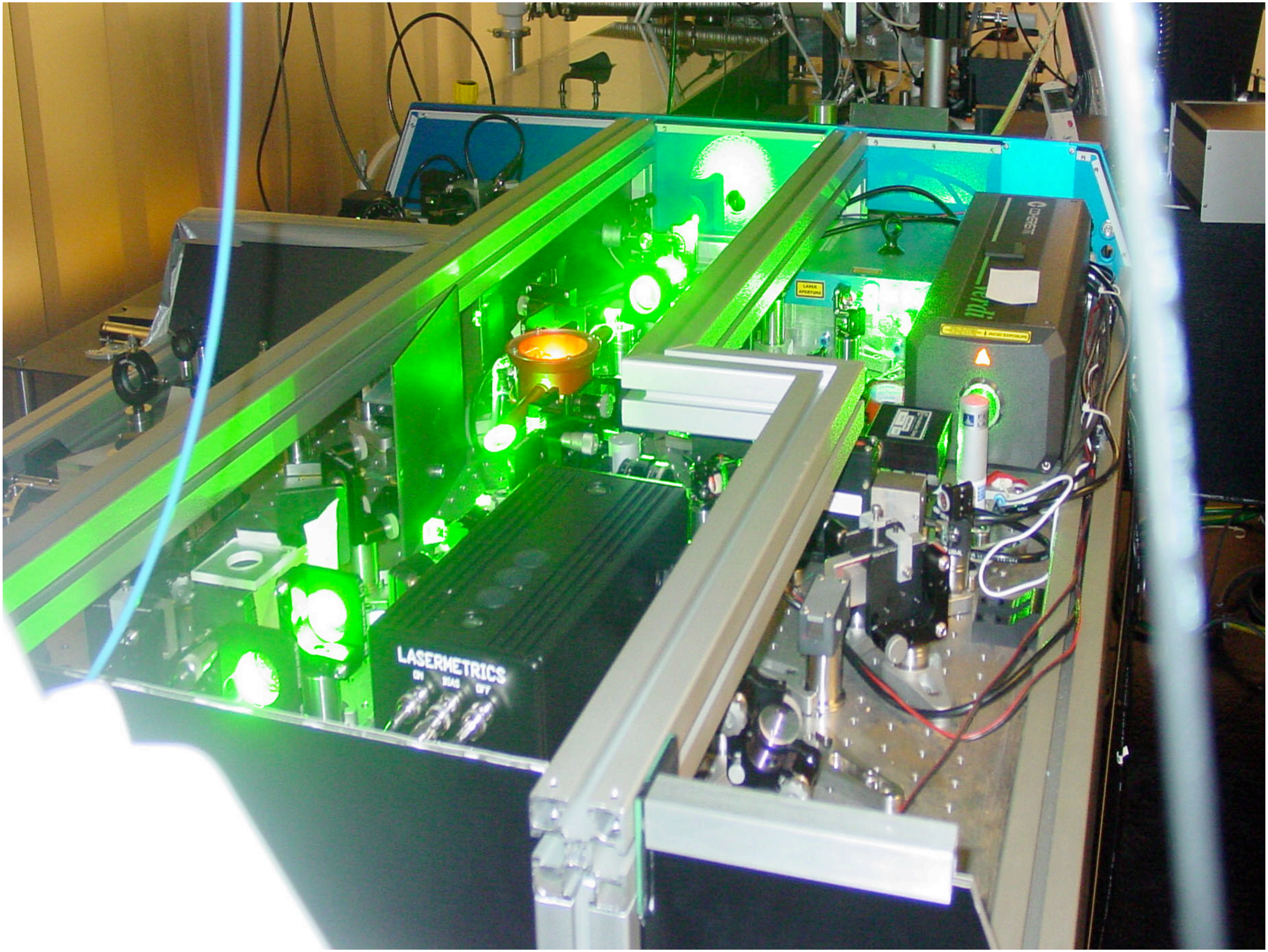
$T_0 = \lambda_0/c \approx 2.5$ fs

$T/4 \approx 625$ as (@ $\lambda_0 \approx 0.75$ μm)

Requires measurement & control of _

The Laser System



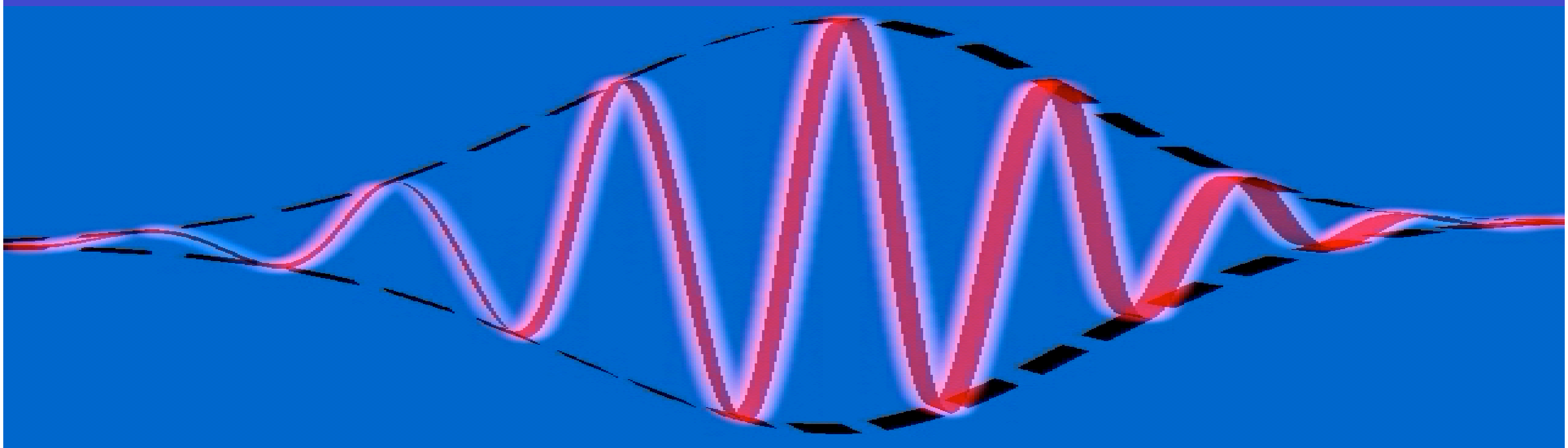

LASERMETRICS

Intense Few-Cycle Laser Pulses

Mode-locked lasers produce pulses with varying _

_n+1 = _n + __

First measurement of __, Vienna, 1996: Xu *et al, Opt. Lett.* **21**, 2008 (1996)



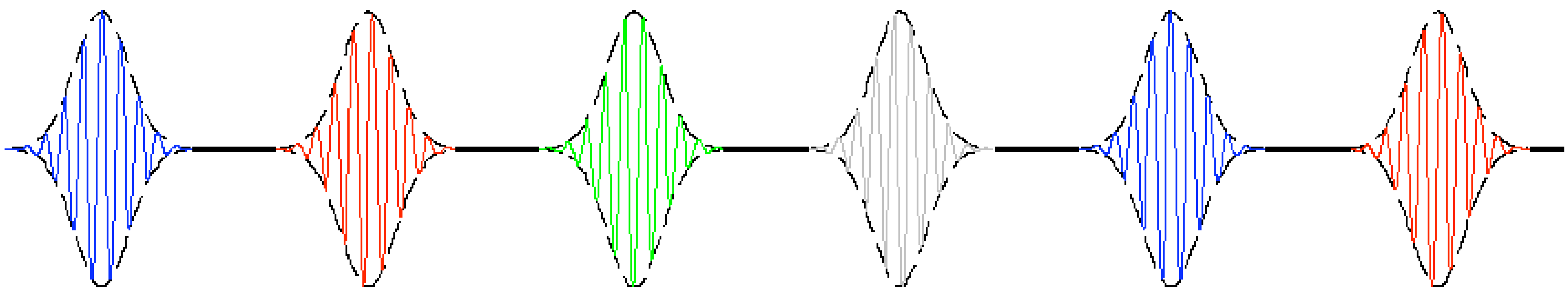
Intense few-cycle laser pulses with stabilized _

Vienna-Munich, 2003: Baltuska *et al*, *Nature* **421**, 611 (2003)

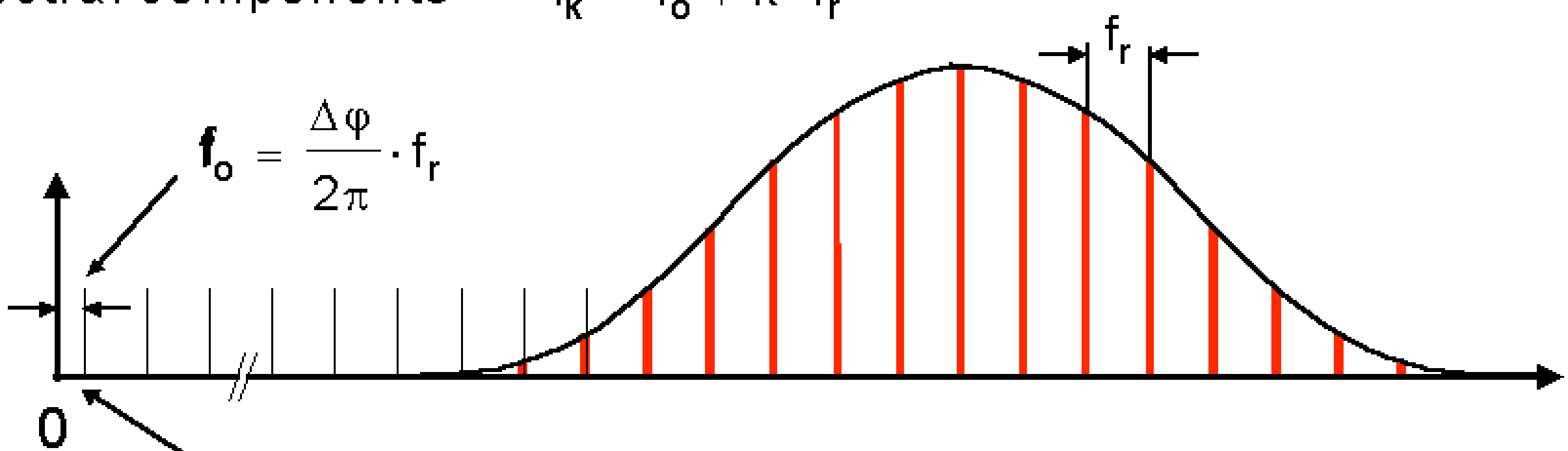
Pulse train from a mode-locked laser

$E_n(t) = A(t) \cdot e^{-i\omega_0 t + \varphi_n} + c.c.$

$\varphi_{n+1} = \varphi_n + \Delta\varphi$



Spectral components $f_k = f_o + k \cdot f_r$

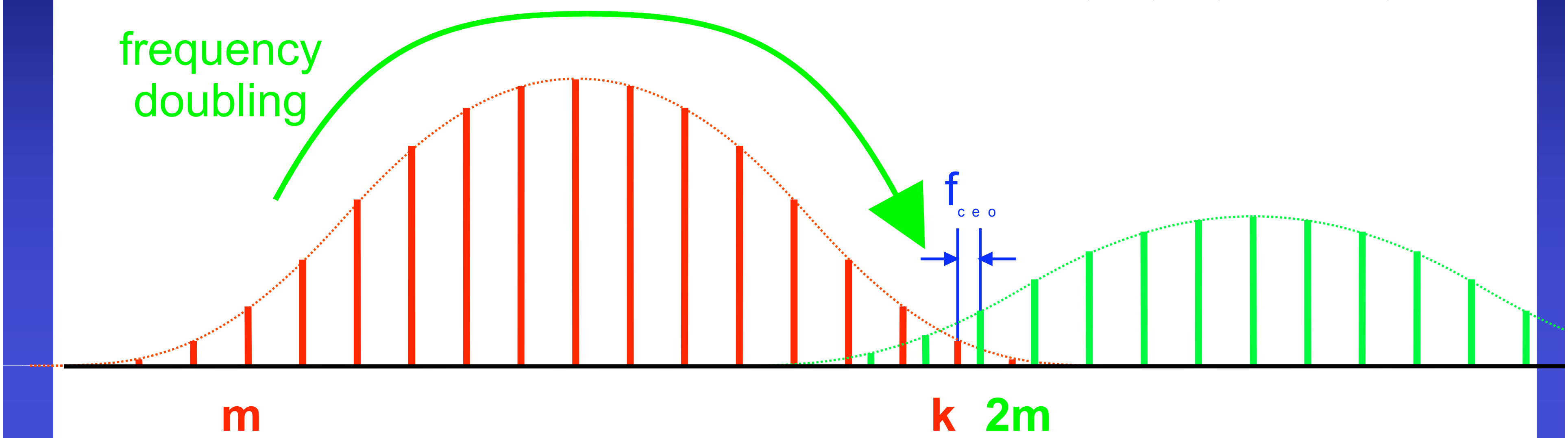


Carrier-envelope phase slip signal

$f_o \equiv f_{ceo}$: carrier-envelope-offset frequency

Frequency-Domain Control of $\Delta\varphi$

T. W. Hänsch *et al.*, 1997, 1999; U. Keller *et al.*, 1999



Beating of the fundamental $f_k = f_o + k \cdot f_r$

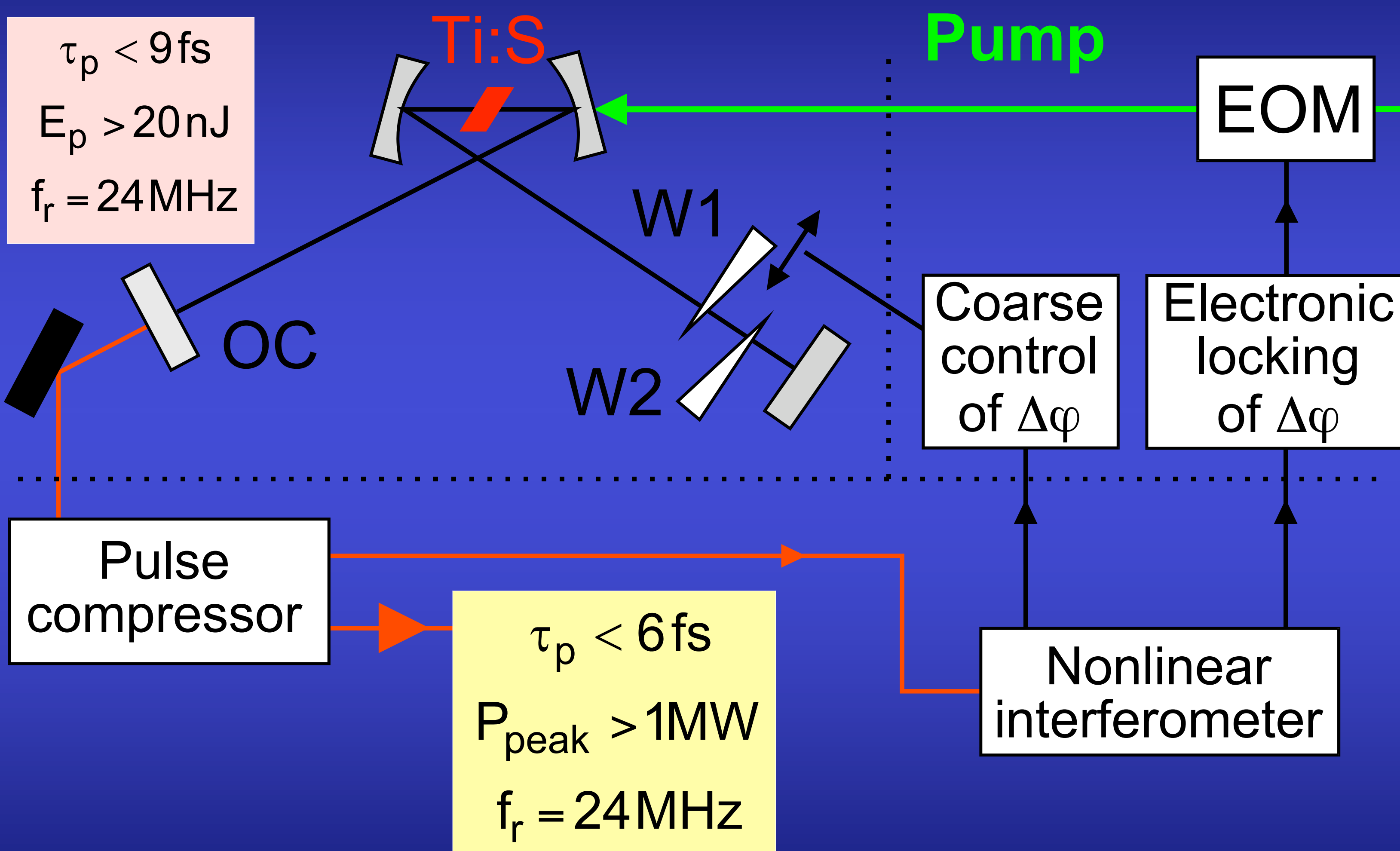
and SH $2 \cdot f_m = 2 \cdot f_o + 2 \cdot m \cdot f_r$

for k=2m: $2 \cdot f_m - f_k = 2 \cdot f_o - f_o + (2m - k) \cdot f_r = f_o \equiv f_{ceo}$

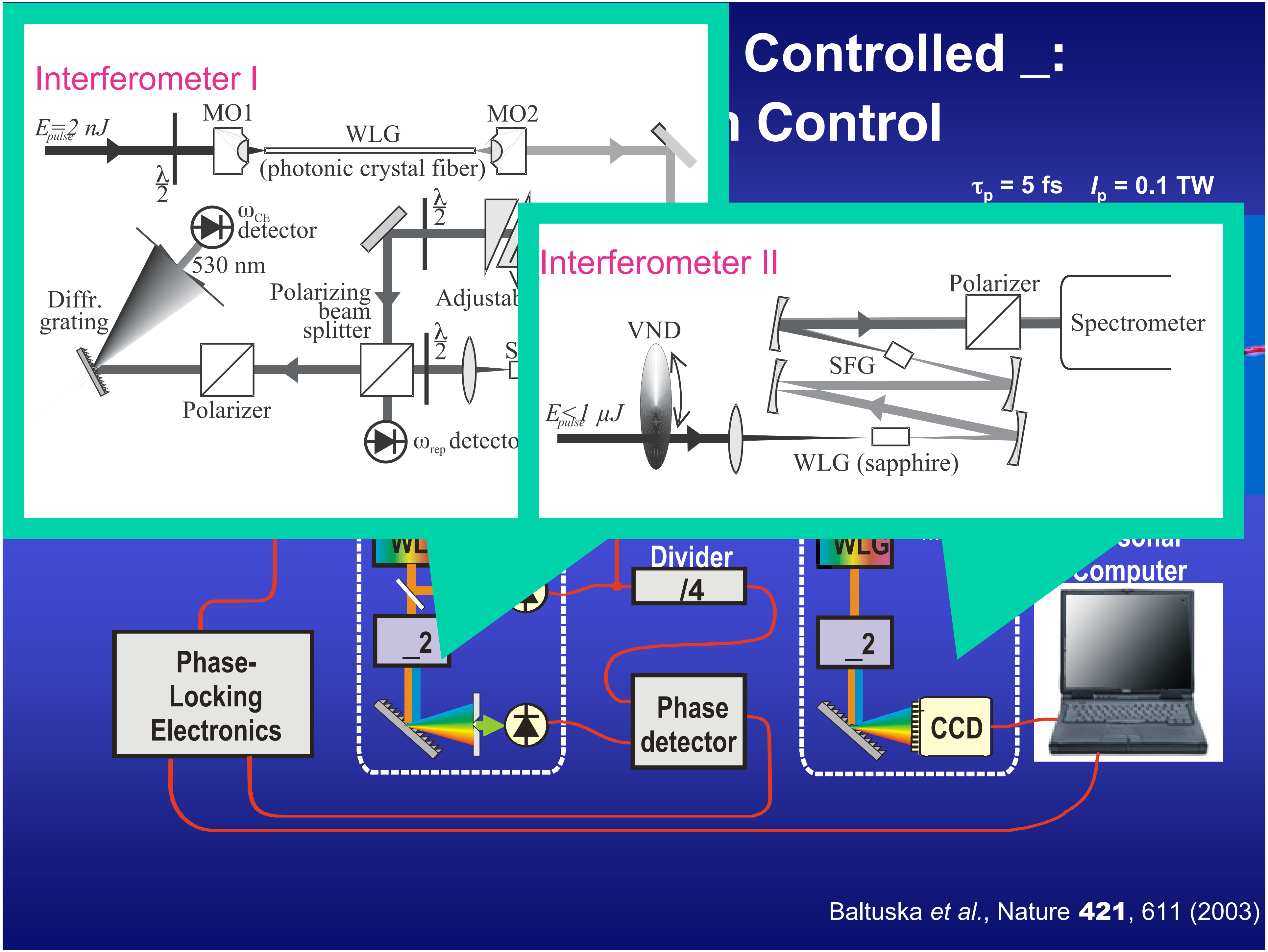
Beat signal yields temporal evolution of φ_n

First implementation: D. Jones *et al.*, Science **288**, 635 (2000); A. Apolonski *et al.*, PRL **85**, 740 (2000)

Phase-Controlled Few-Cycle Pulses

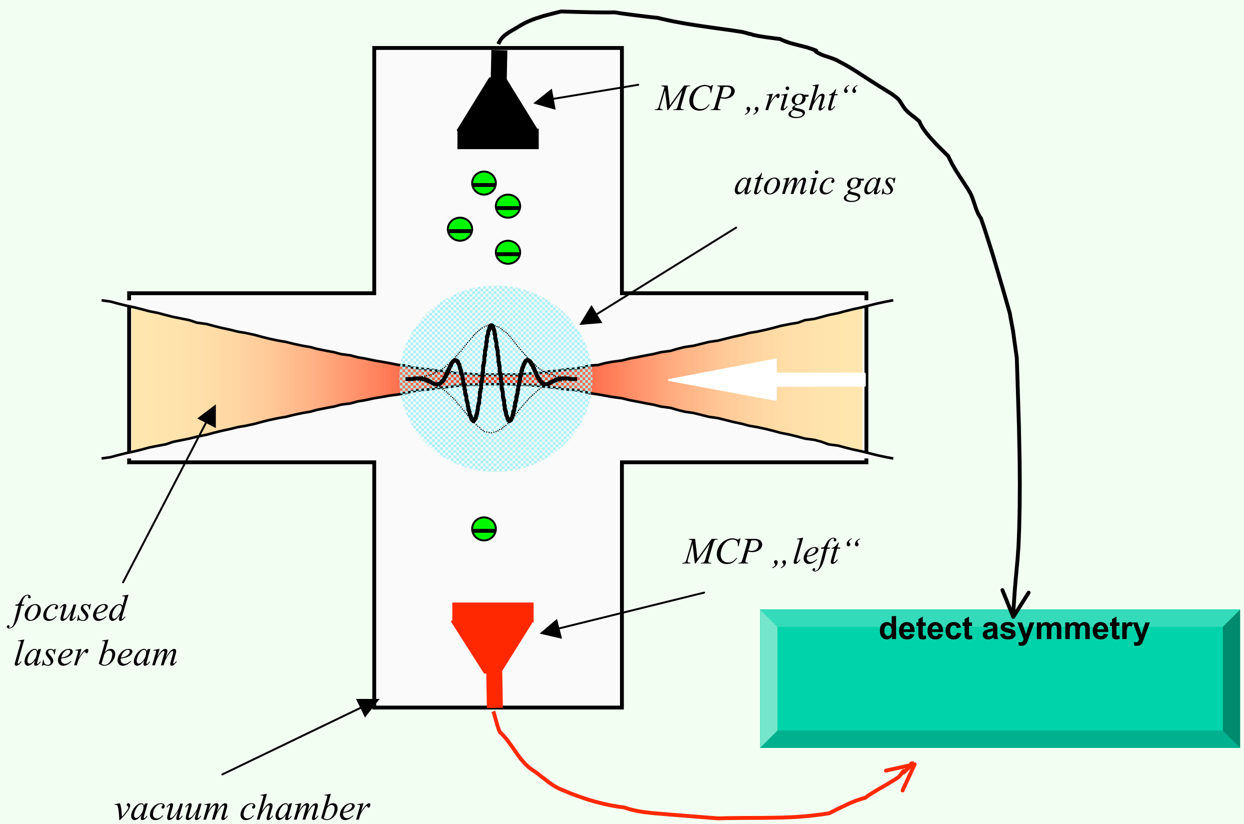


A. Apolonski *et al.*, PRL **85**, 740 (2000); A. Poppe *et al.*, Appl. Phys. B **72**, 373 (2001)


Controlled _:
n Control
τ_p = 5 fs I_p = 0.1 TW
Interferometer I
E_{pulse}=2 nJ
MO1
WLG
MO2
(photonic crystal fiber)
λ/2
ω_{CE} detector
530 nm
Diffr. grating
Polarizing beam splitter
Adjustab
Polarizer
ω_{rep} detecto
Interferometer II
Polarizer
Spectrometer
VND
SFG
E_{pulse}≤1 μJ
WLG (sapphire)
Divider
/4
Phase-Locking Electronics
Phase detector
CCD
Computer
_2
Baltuska *et al.*, Nature **421**, 611 (2003)

Phase Calibration Using ATI

ATI (Above-Threshold Ionization) Photoelectron Spectroscopy

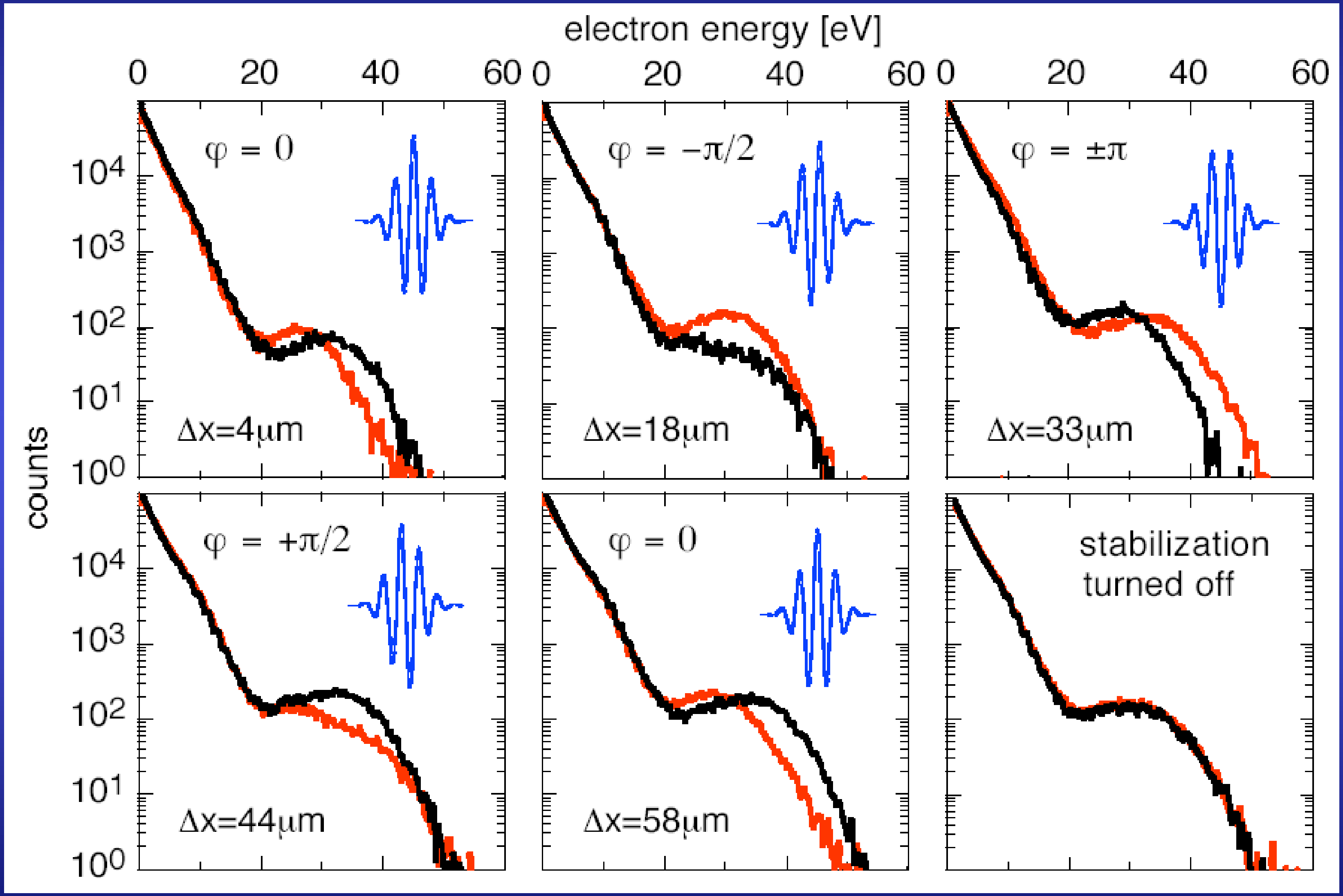


Gas: Xe, Laser peak intensity: 8.10^{13} W/cm^2

Apparatus
“Stereo ATI spectrometer”

Paulus *et al*. Nature, **414** 182 (2001)

Phase Calibration Using ATI



Photoelectron spectra

Paulus *et al.*, PRL (2003)

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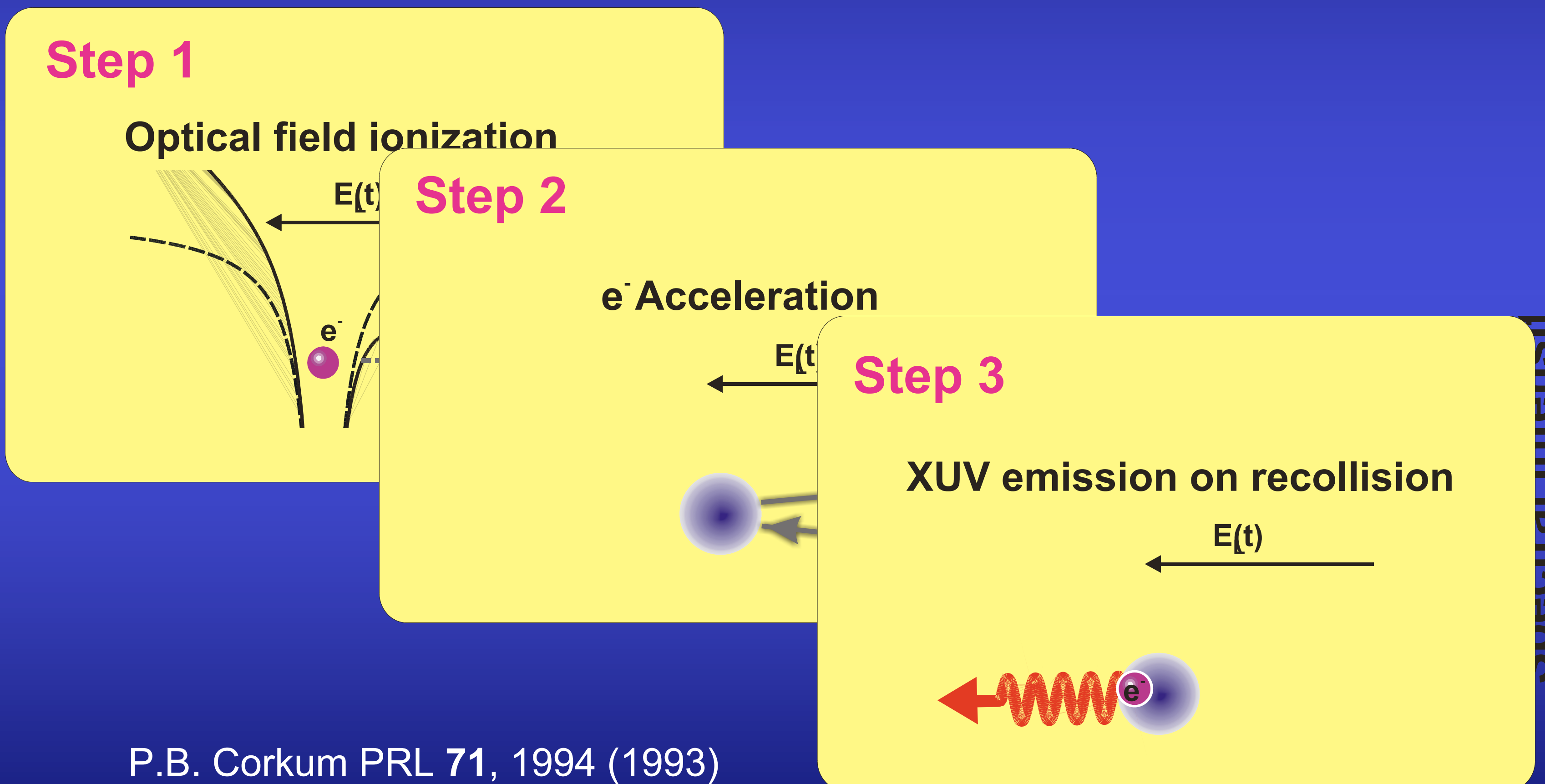
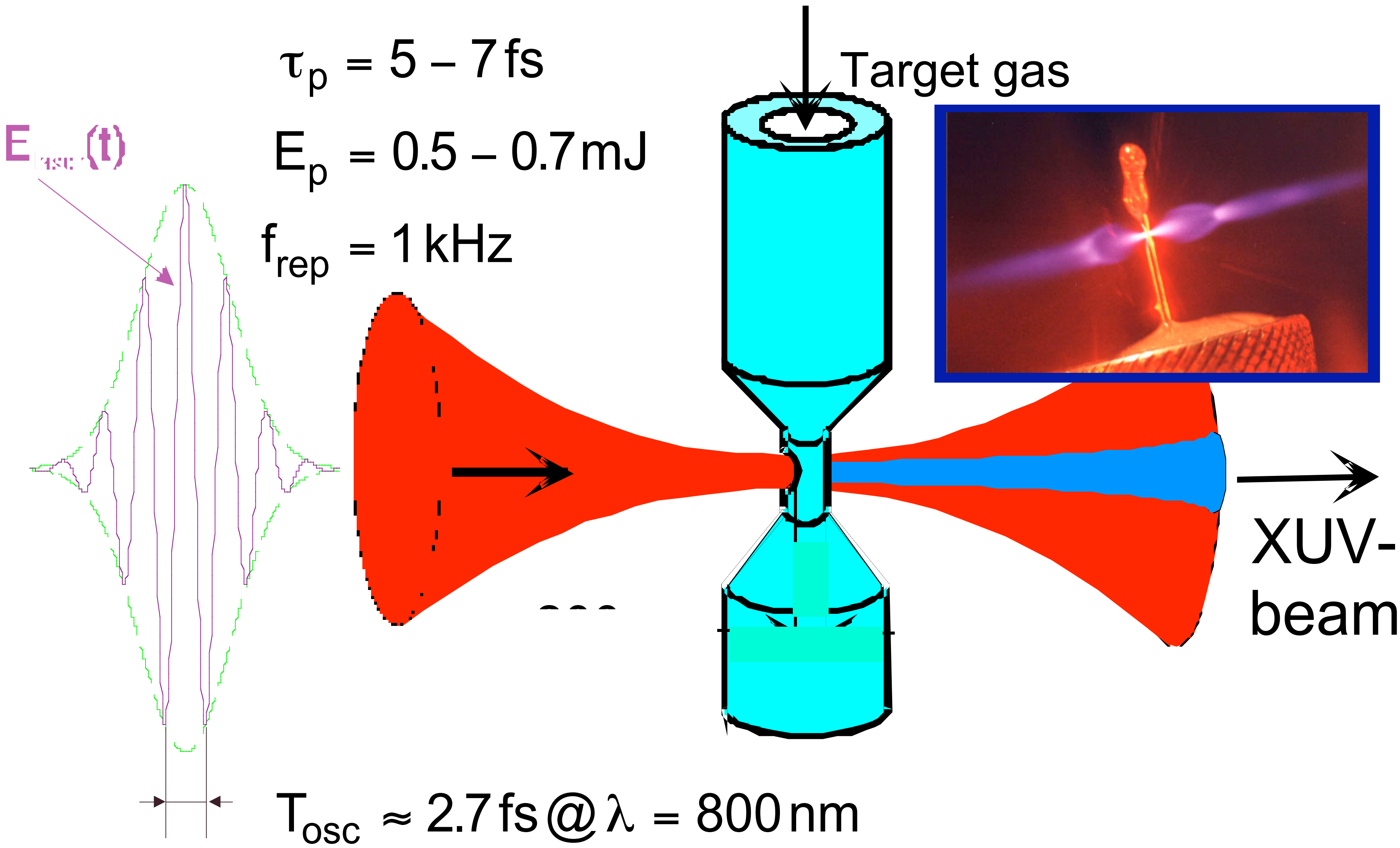
High-order Harmonic Generation

Attosecond XUV pulse generation



P.B. Corkum PRL **71**, 1994 (1993)

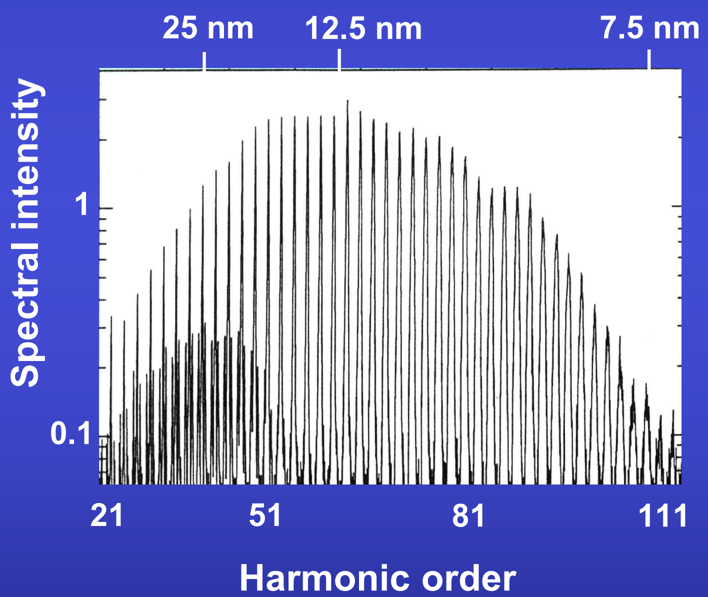
High-OrderHarmonicGeneration withSub-10-fsLaserPulses

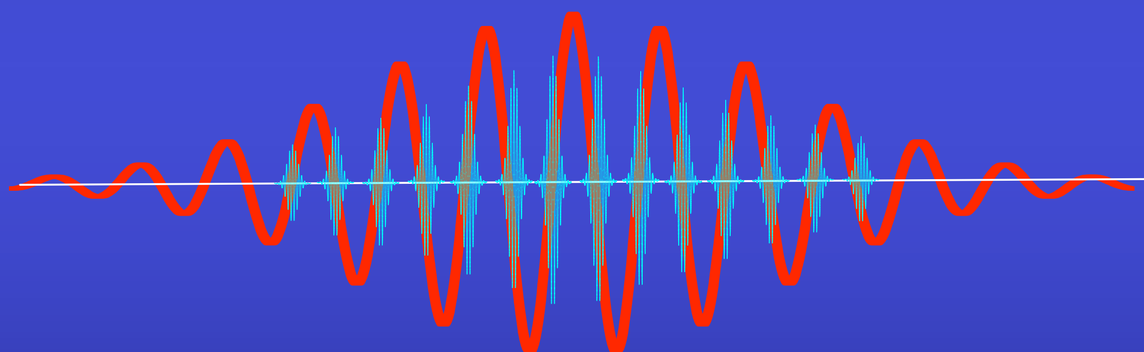


Recombination Emission from Strongly-Driven Atoms

Multi-cycle driver pulse : $\tau_p \gg T_o$

High-order odd harmonics of the driver laser



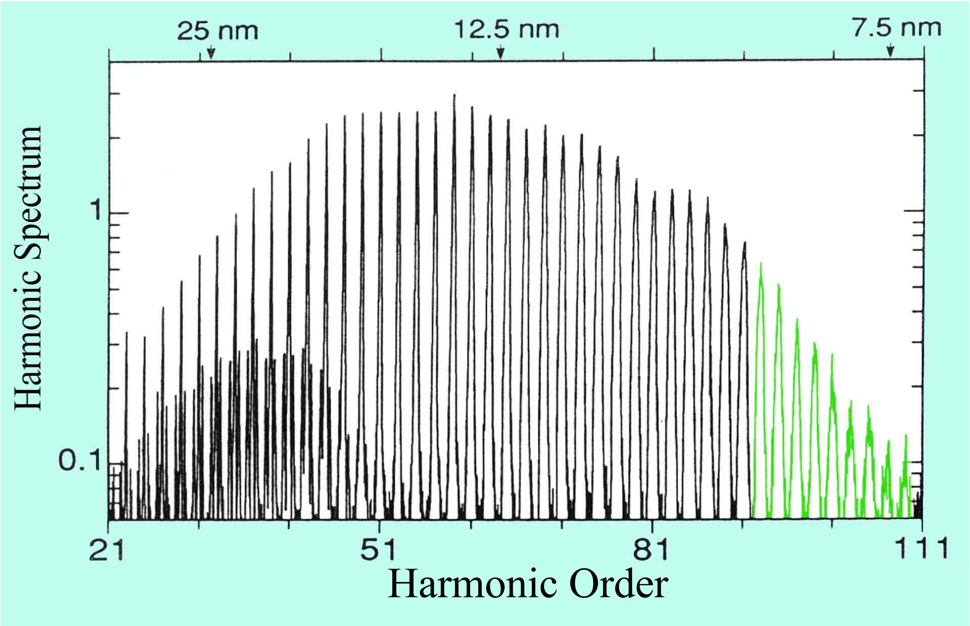


Cut-off harmonics: train of attosecond bursts

L'Huillier, Balcou, 1993, *PRL* **70**, 774
Macklin *et al,* 1993, *PRL* **70**, 766

Paul *et al,* Science **292**, 1689 (2001)
Tsakiris, Charalambidis *et al,* 2003

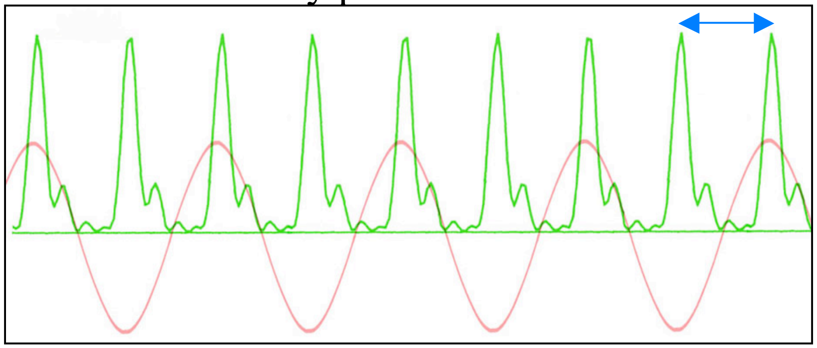
High Harmonic Generation



← J. Macklin *et al.*, PRL **70**, 766 (1993)
A. L'Huillier, P. Balcou, PRL **70**, 774 (1993)

Multi-100-THz trains of
sub-fs XUV/X-ray pulses

$\Delta T = \pi/\omega_L \approx 1.35$ fs @ $\lambda_L = 800$ nm

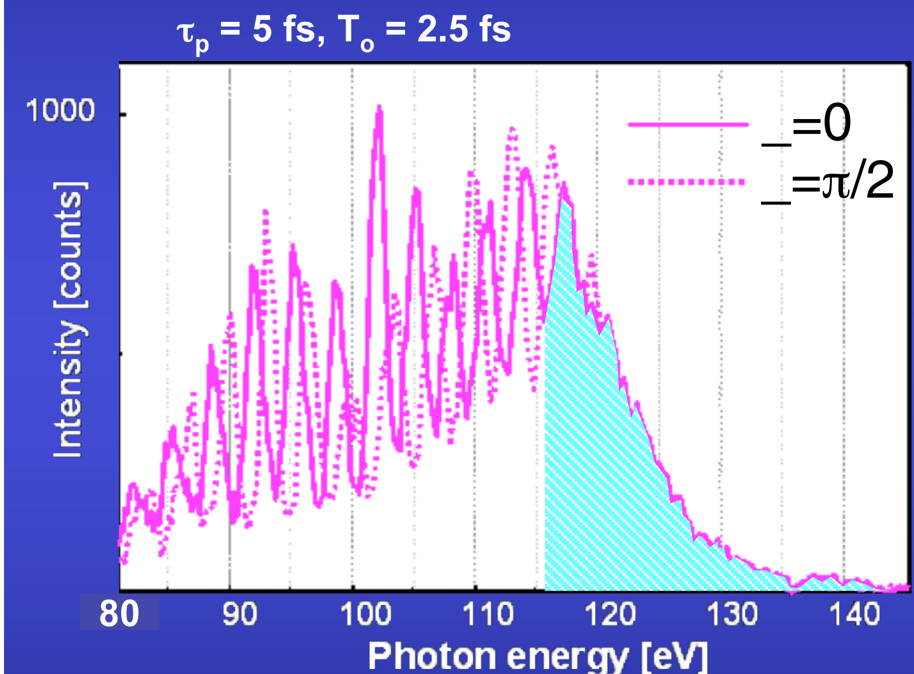


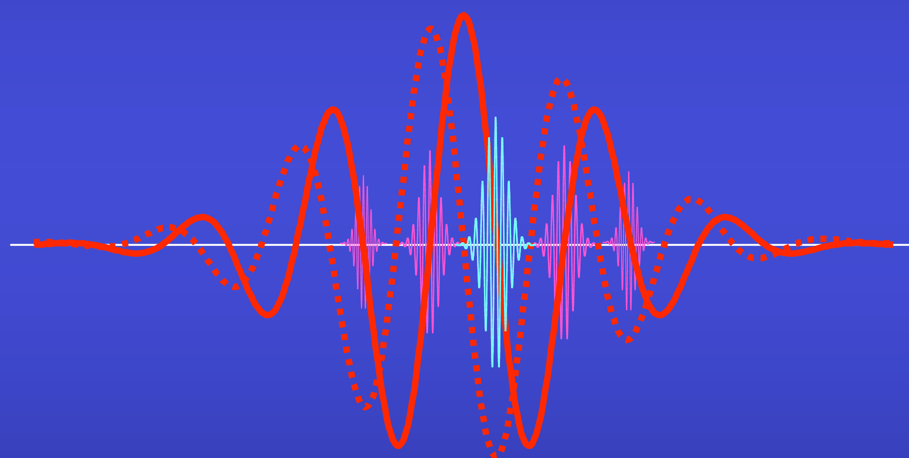
P. Antoine *et al.*, PRL **77**, 1234 (1996)
K. Schäfer, K. Kulander, PRL **78**, 638 (1997)
I. Christov *et al.*, PRL **78**, 1251 (1997)
N. Papadogiannis *et al.*, PRL **83**, 4289 (1999)

P. M. Paul *et al.*, Science **292**, 1689 (2001)

Recombination Emission from Strongly-Driven Atoms

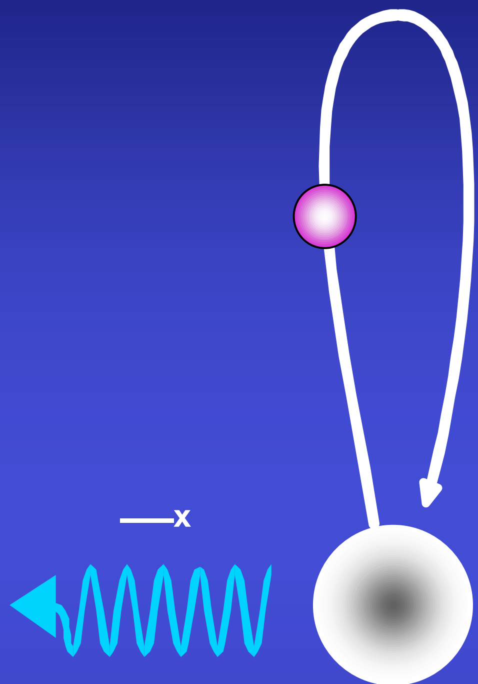
Few - cycle driver : $\tau_p < 3T_o$



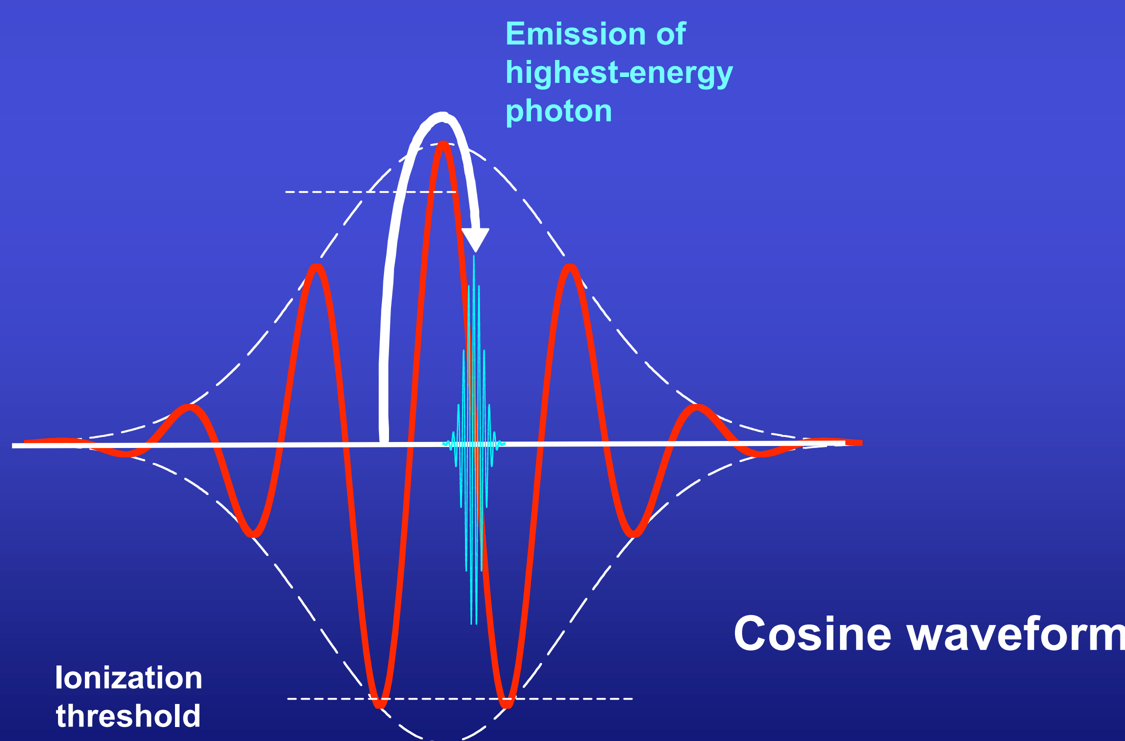


- **No genuine harmonics of the laser radiation**
- **Cosine waveform with $\tau_p \sim 2T_o$ (5 fs @ 750 nm) offers the potential for single sub-femtosecond X-ray pulse generation**

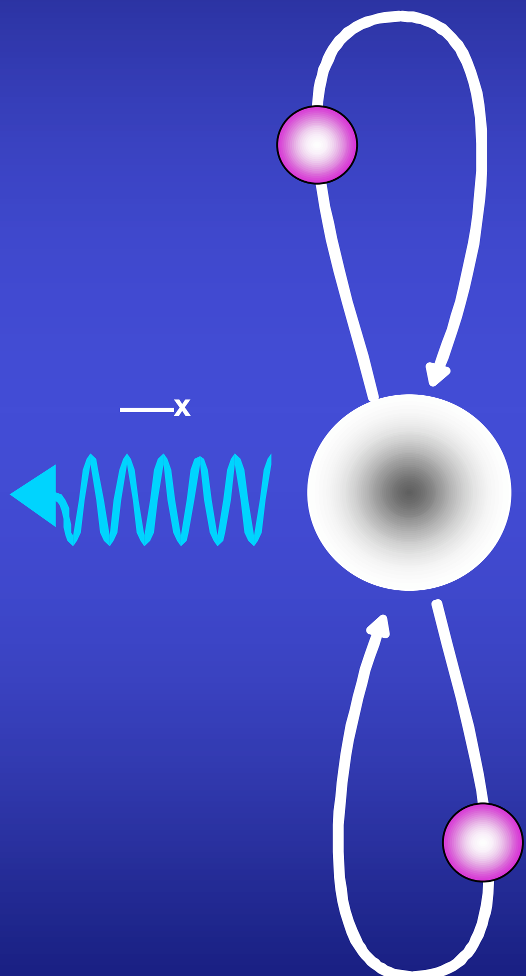
Recombination emission of a cosine pulse



Recombination emission:
soft-X-ray photon emission upon the electron recombining into its ground state

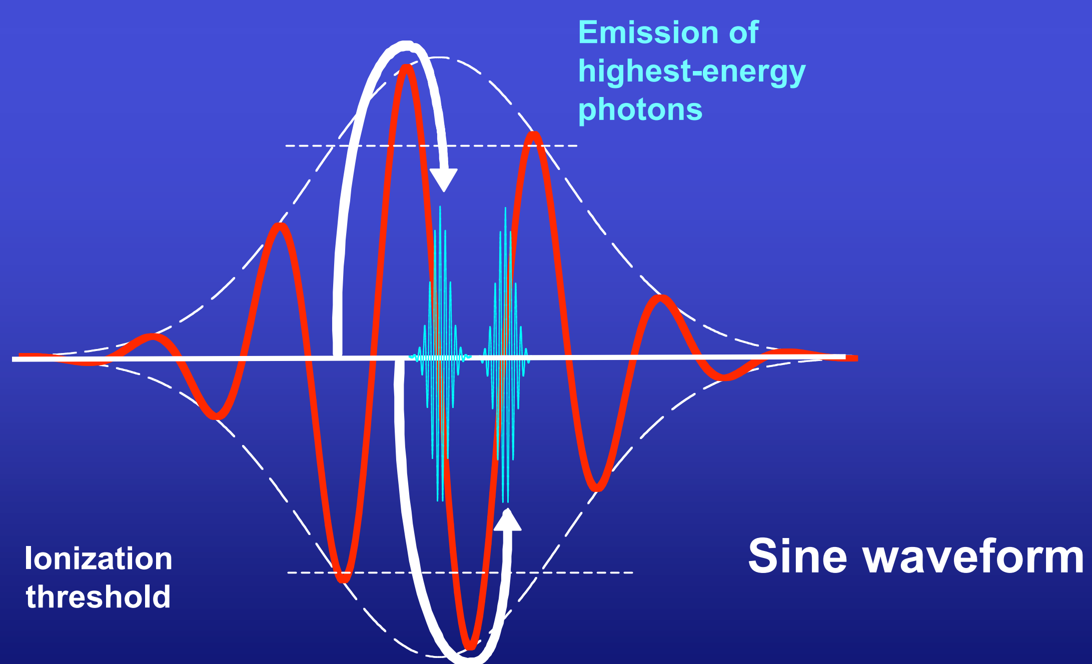


Recombination emission of a sine pulse



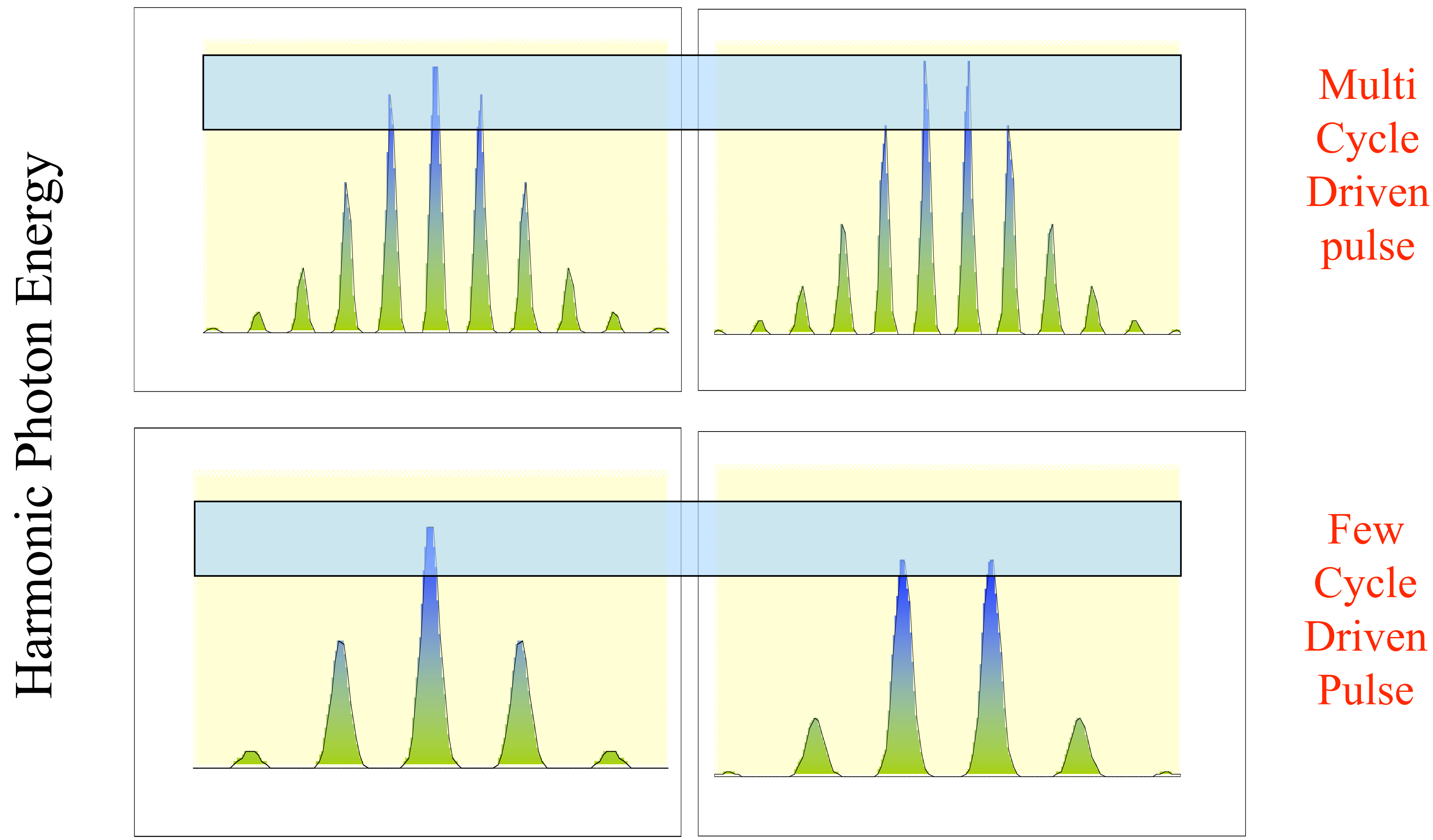
Recombination emission:

soft-X-ray photon emission upon the
electron recombining into its ground state

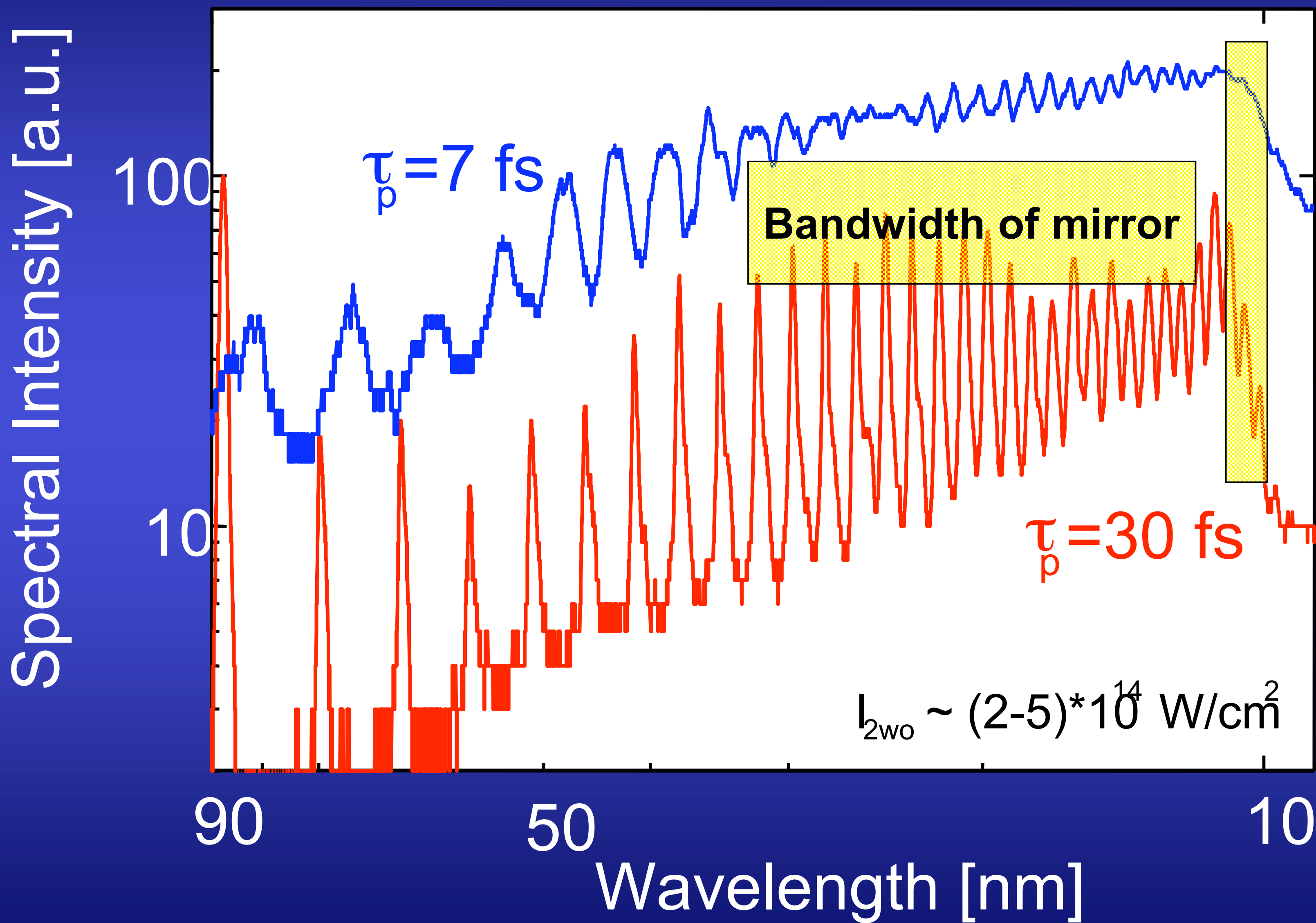


Harmonics in the time-domain

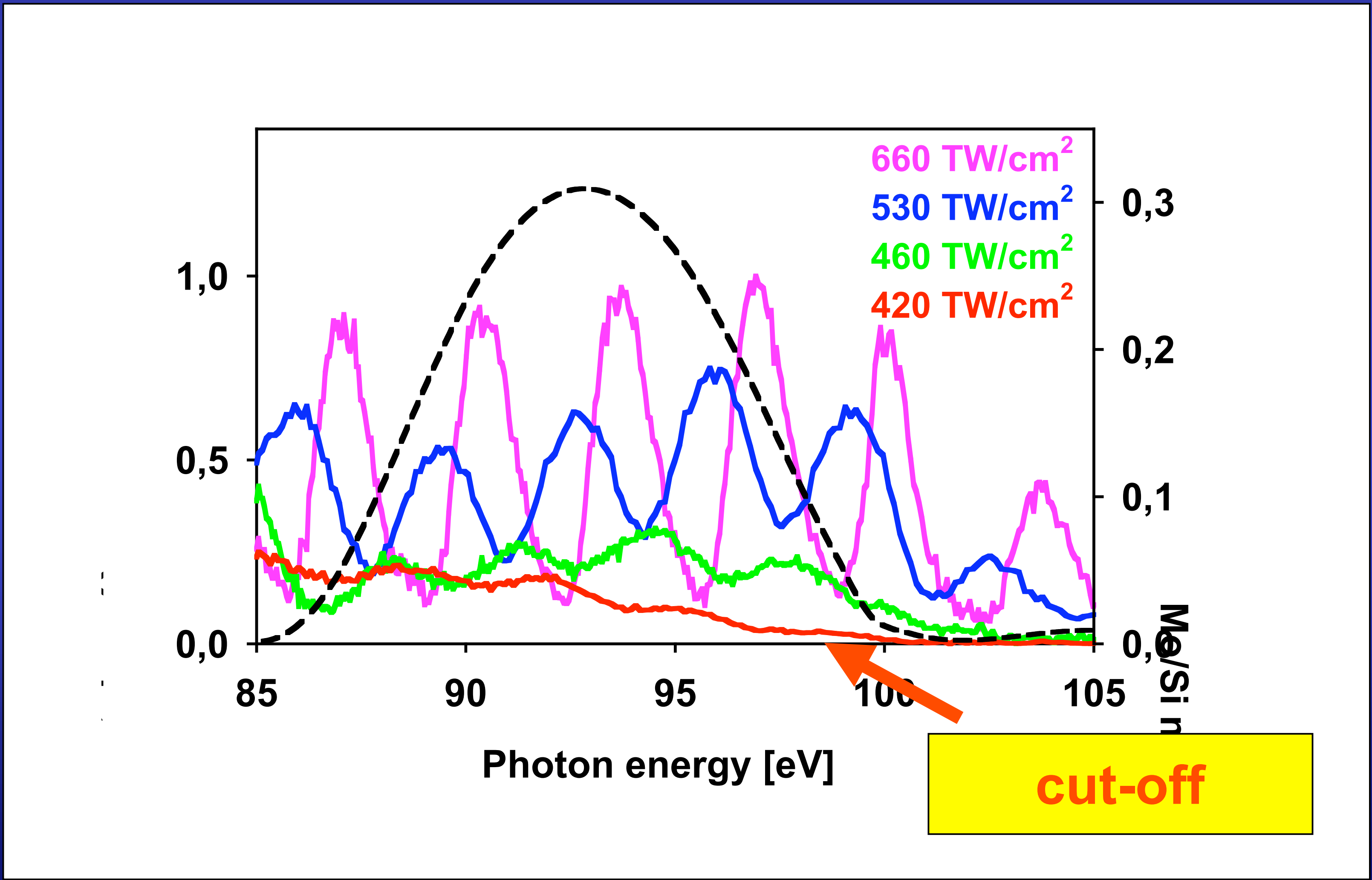
$E(t) = A(t) \cos \omega_L t$ $\qquad$ $E(t) = A(t) \cos \omega_L t$



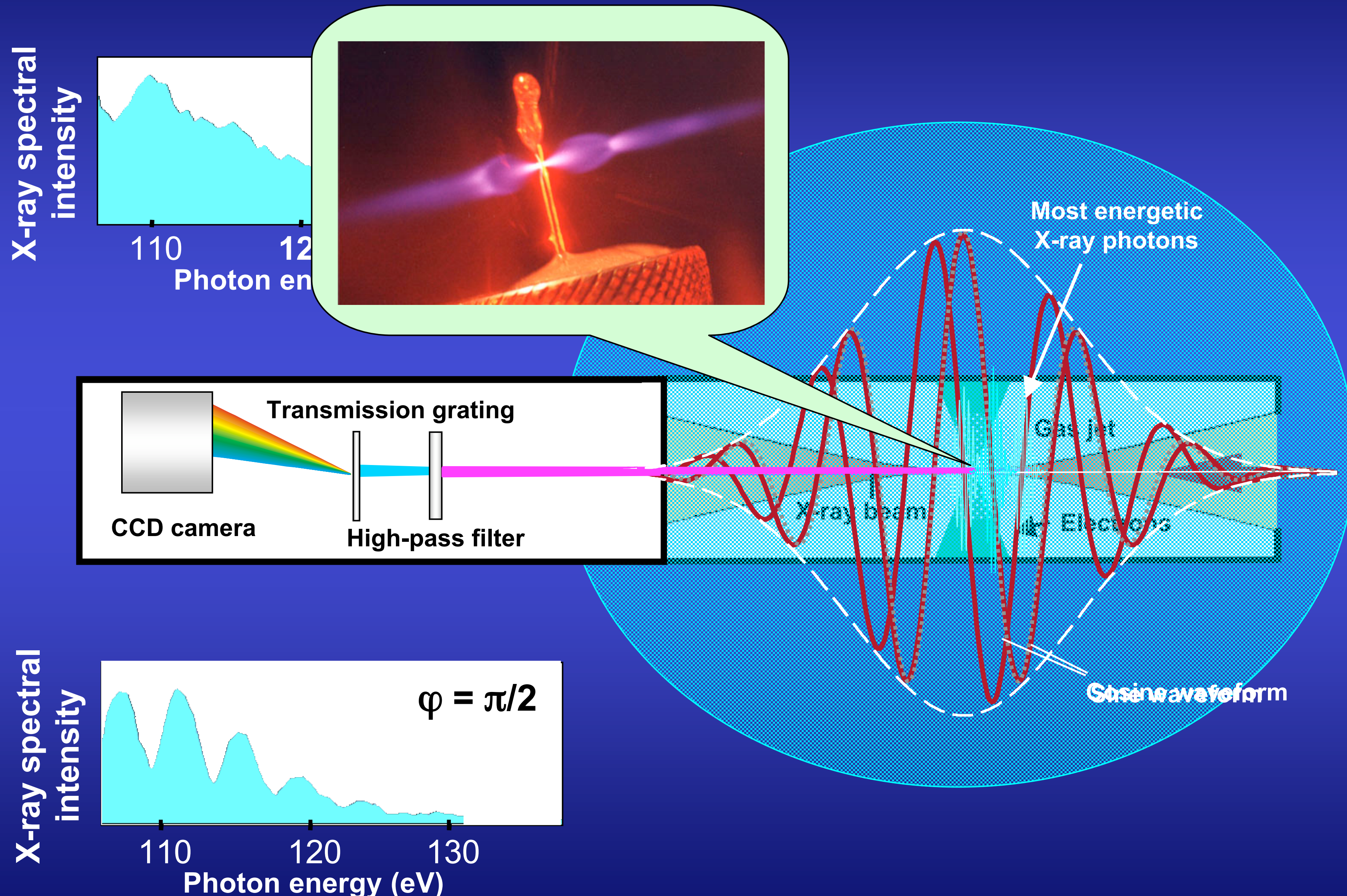
XUV Harmonics from Neon



Harmonic spectra depending on generating laser intensity



Recombination Emission from Ionizing Atoms



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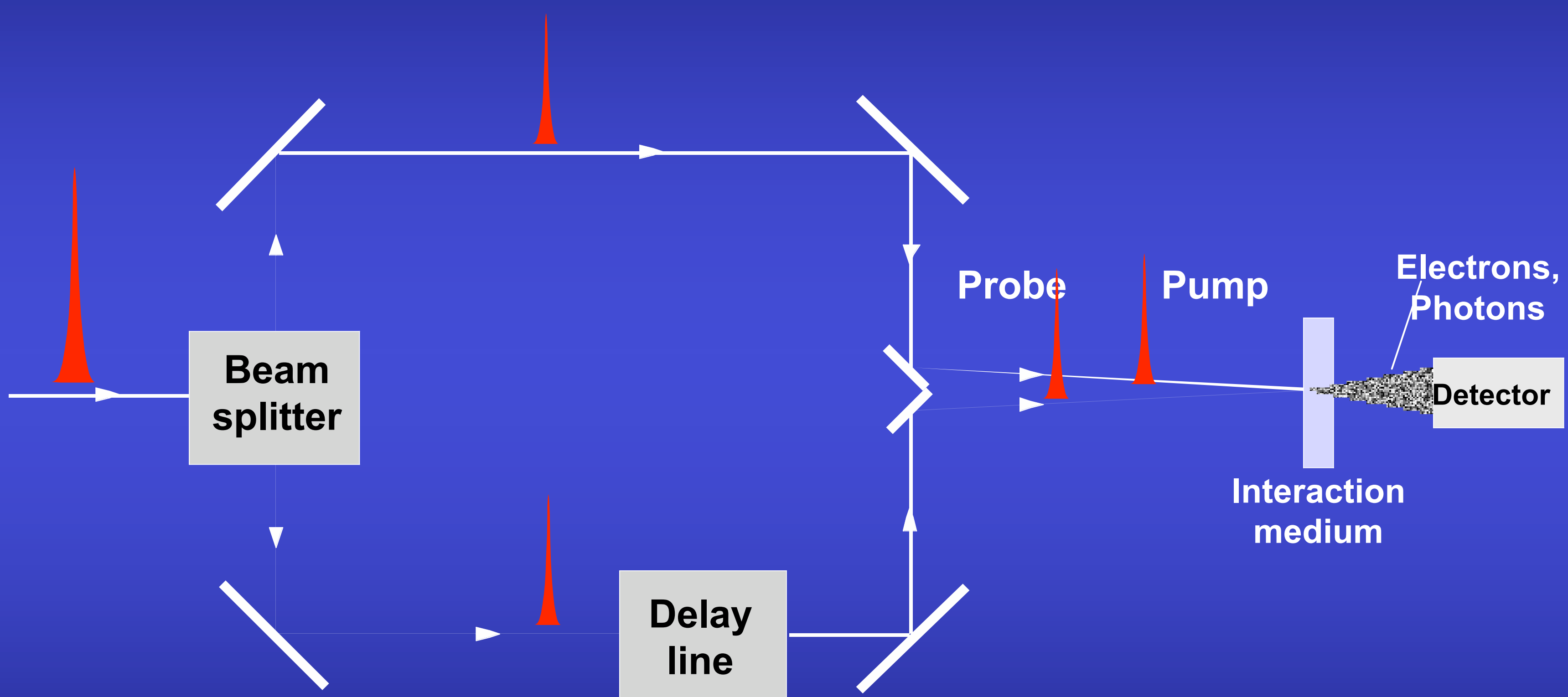
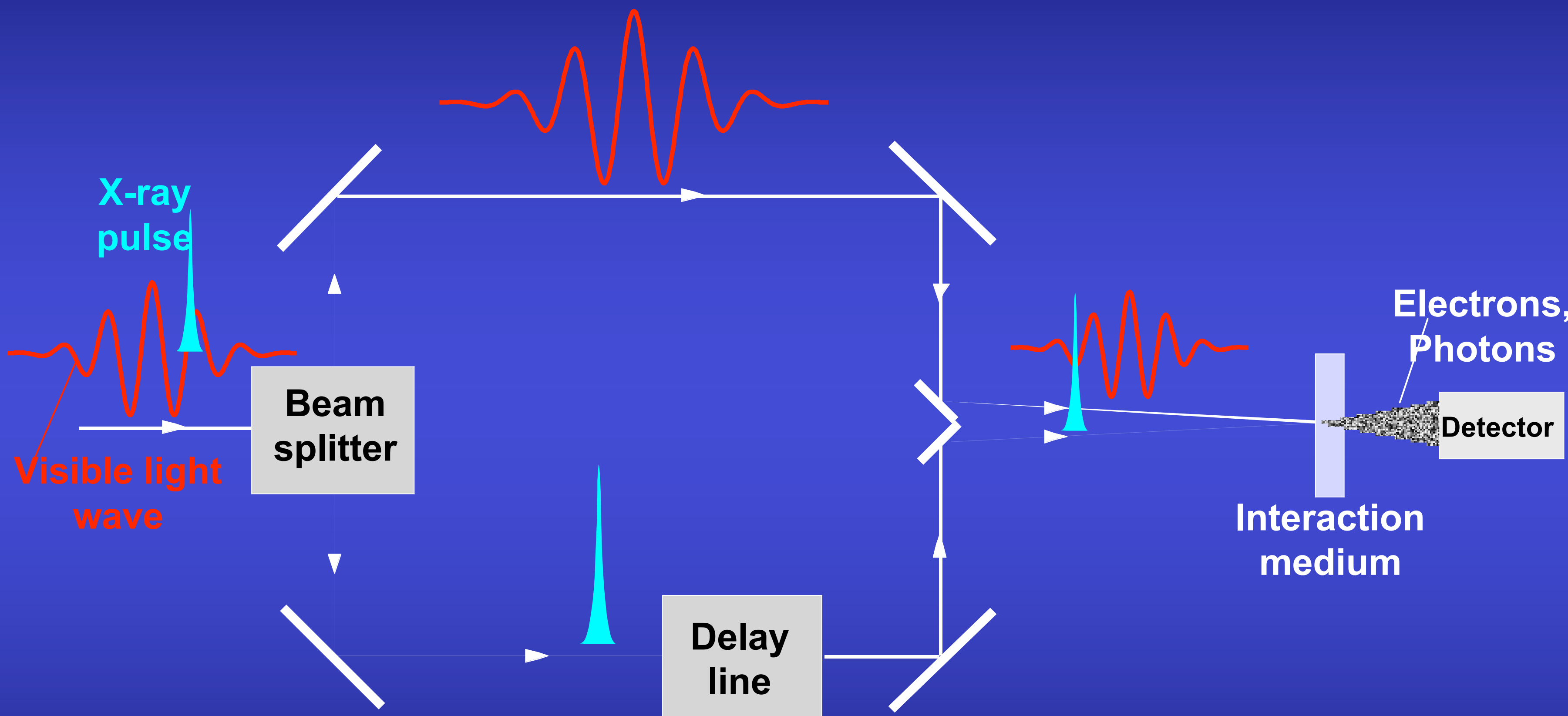
Sampling a Pulse with Itself: Autocorrelation



Frustrated by low two-photon transition probability at X-ray photon energies

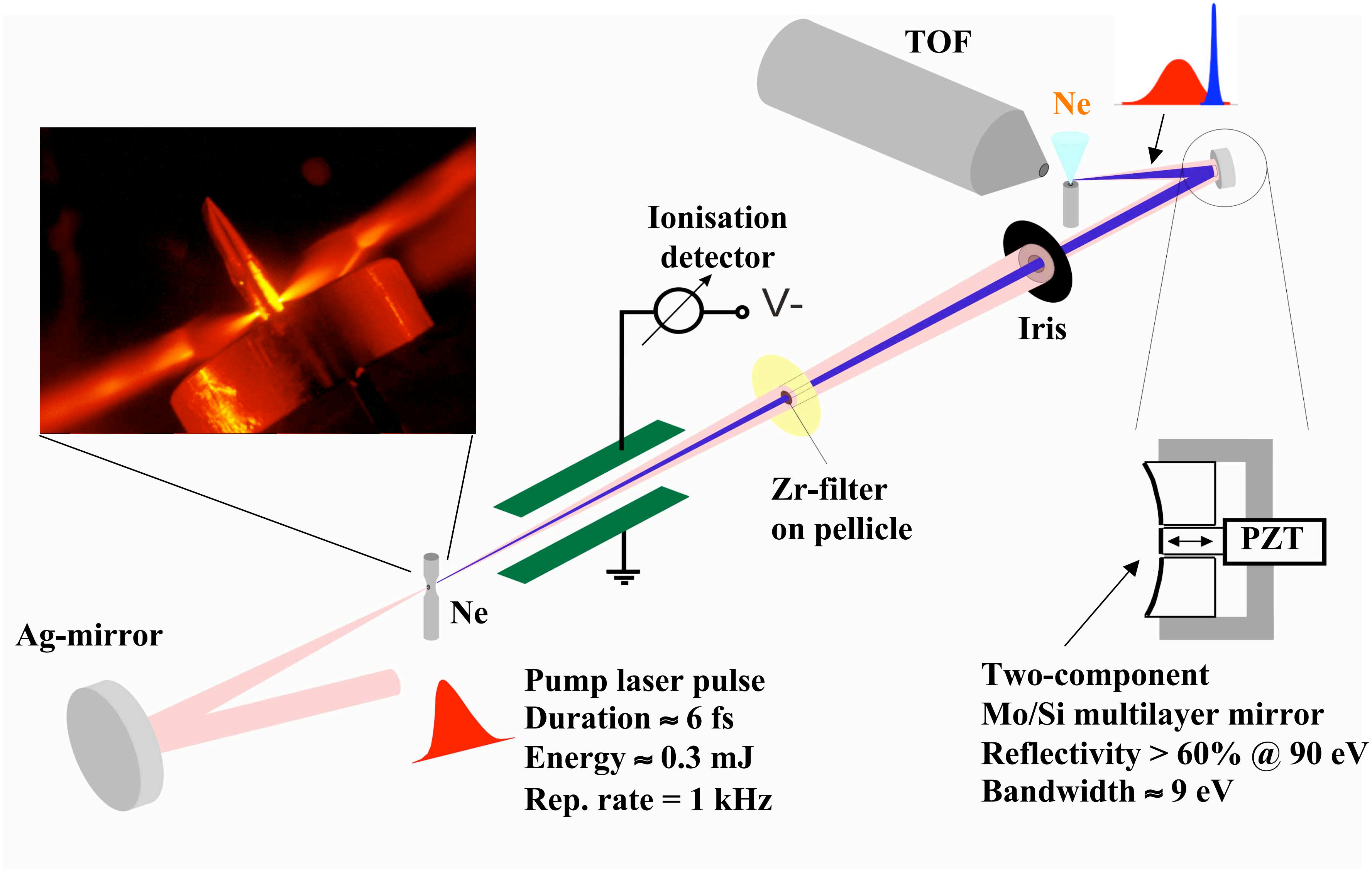
Measuring a Sub-Femtosecond X-Ray Pulse with Laser Light?



Sampling must be performed by the laser *field* rather than the pulse envelope

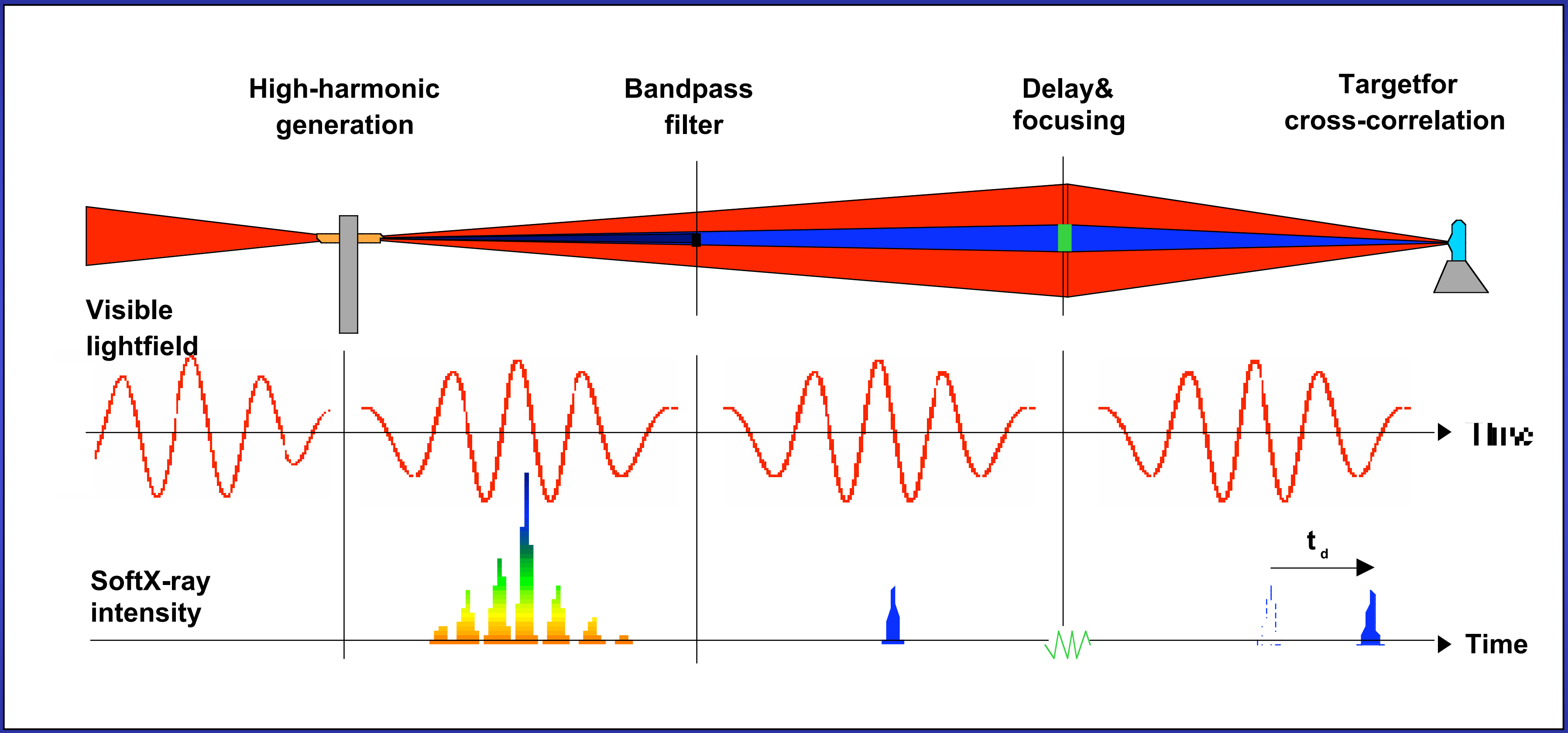
Hentschel *et al.*, Nature **414**, 509 (2001)

The Measurement: Experimental Setup

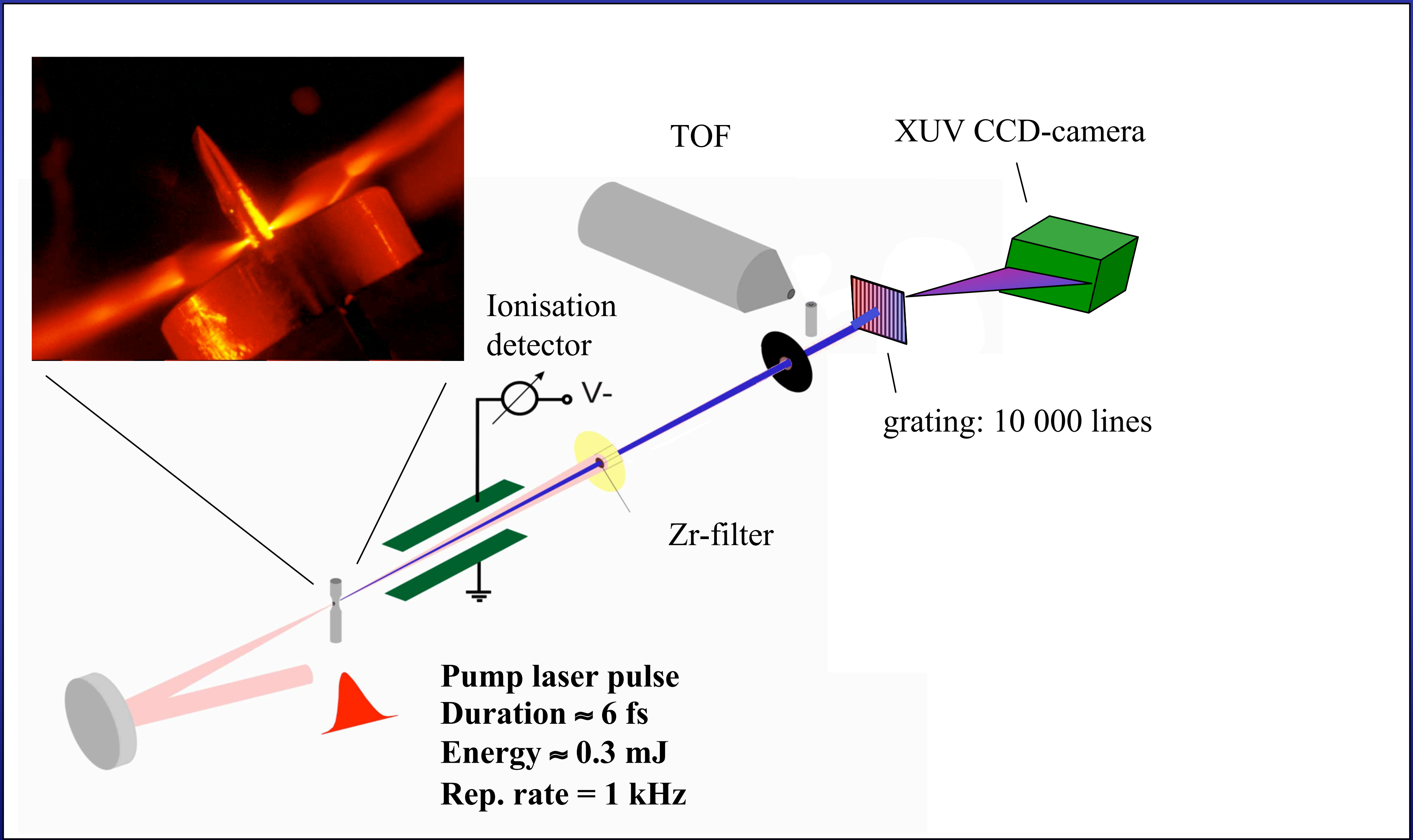


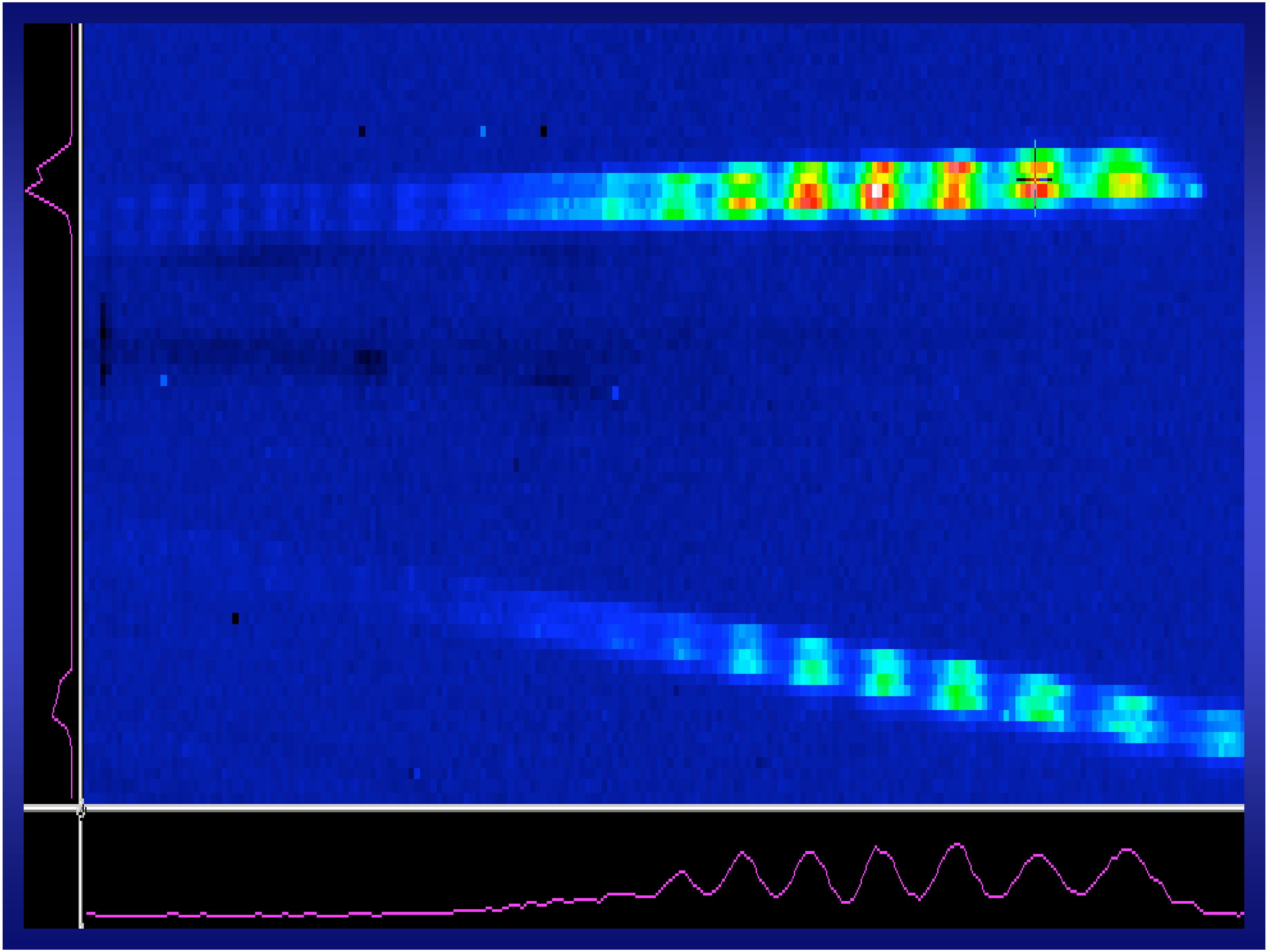
M. Drescher *et al.*, Science **291**, 1923 (2001)

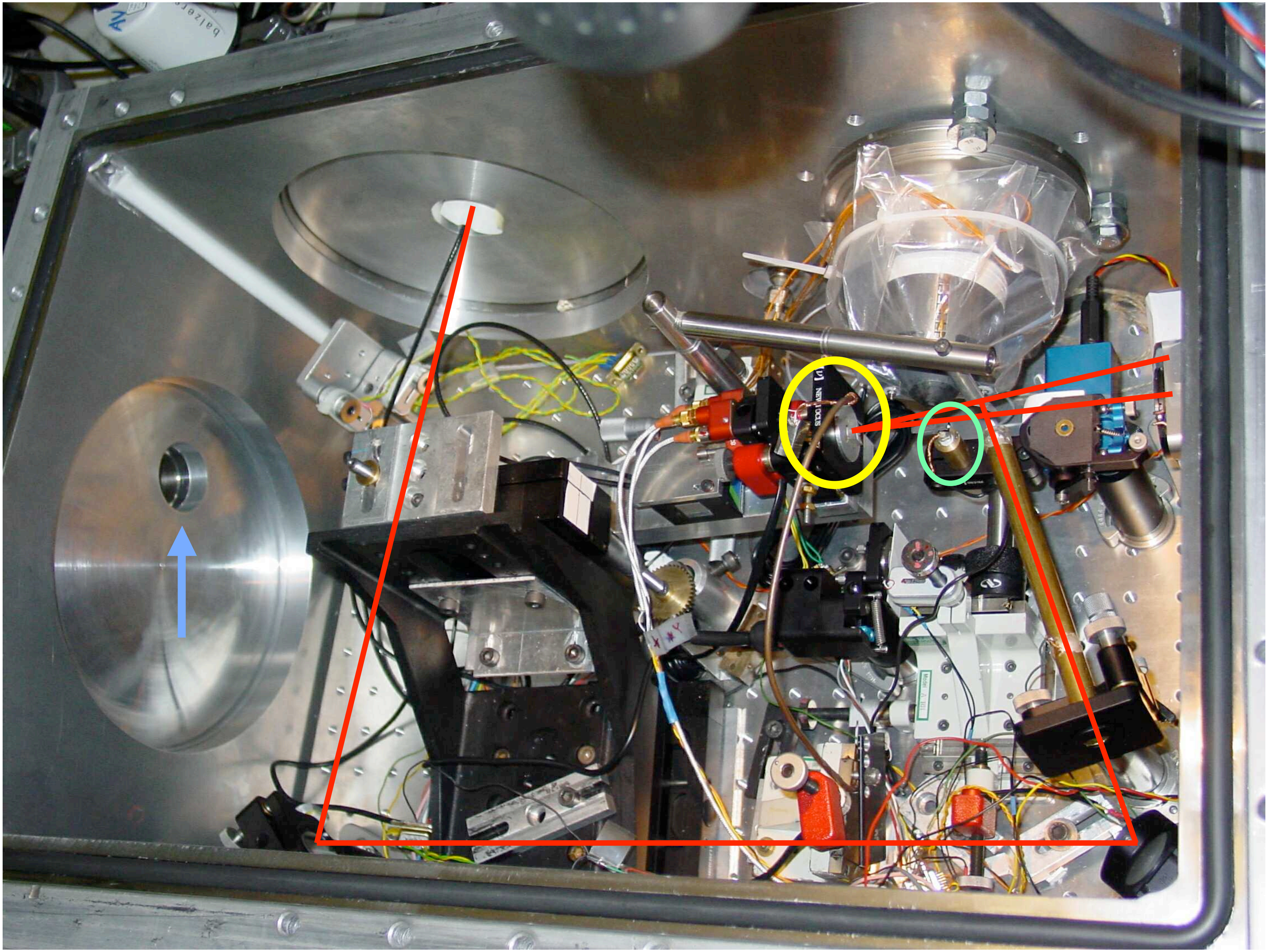
Schematic of the Experiment



Setup for measuring ‚online' spectra

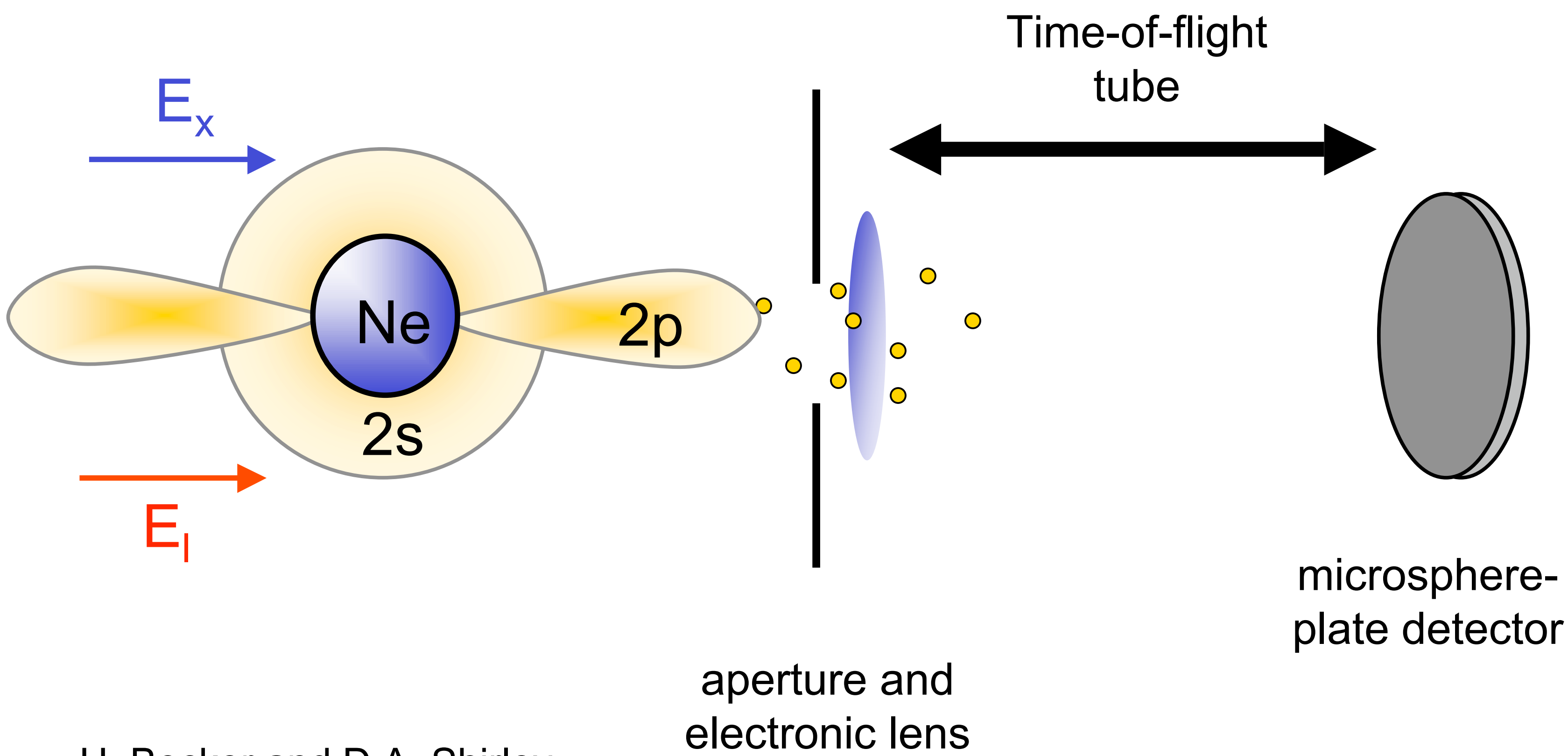






VIS / XUV X-Correlation: Principle of Excitation and Detection

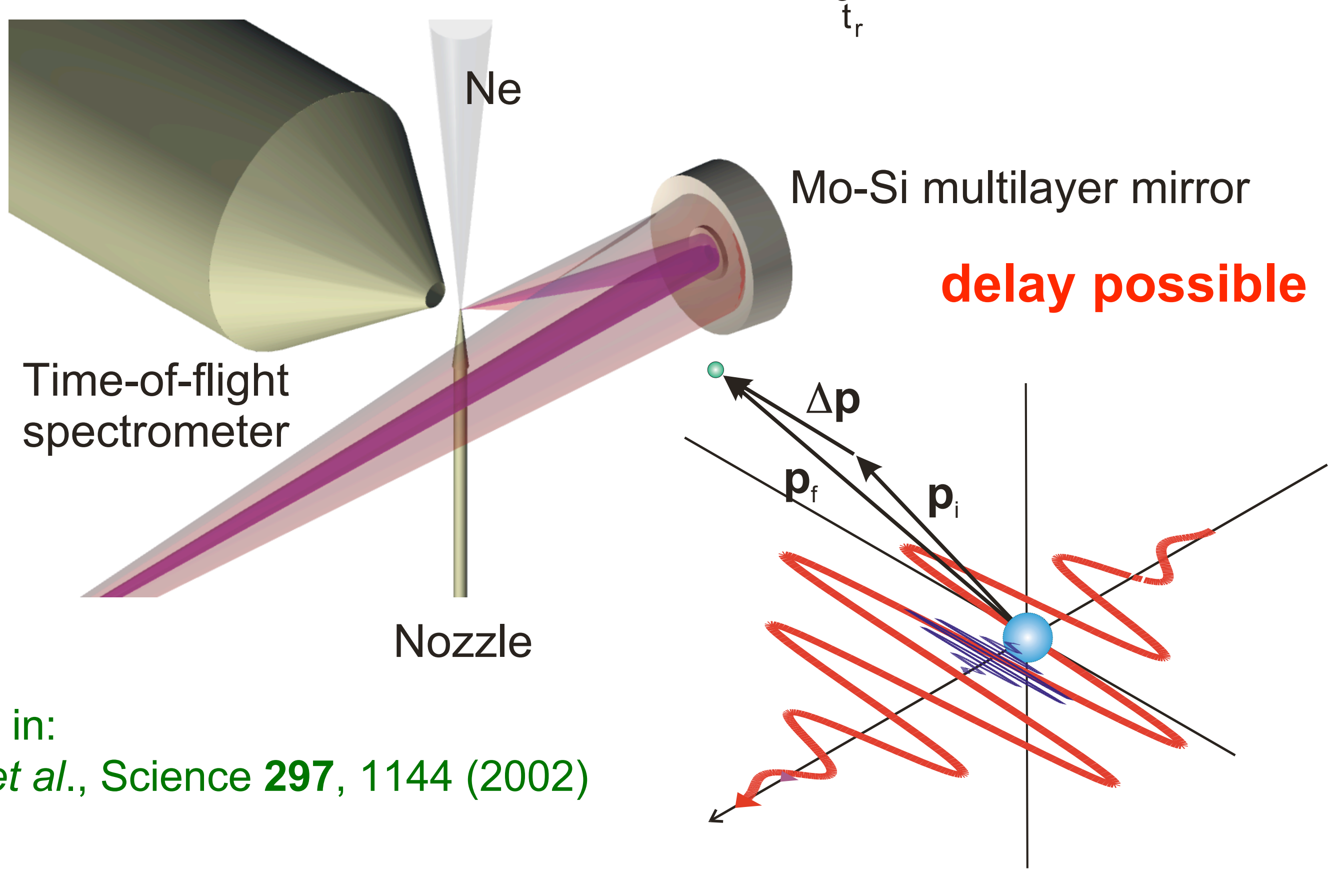
parallel geometry!!



U. Becker and D.A. Shirley,
VUV and Soft X-ray Photoionization, p. 152

Ionization with an Isolated Attosecond Pulse

$$\Delta p(t)_r = e\int_{t_r}^{\infty} E_L(t')\,dt'$$

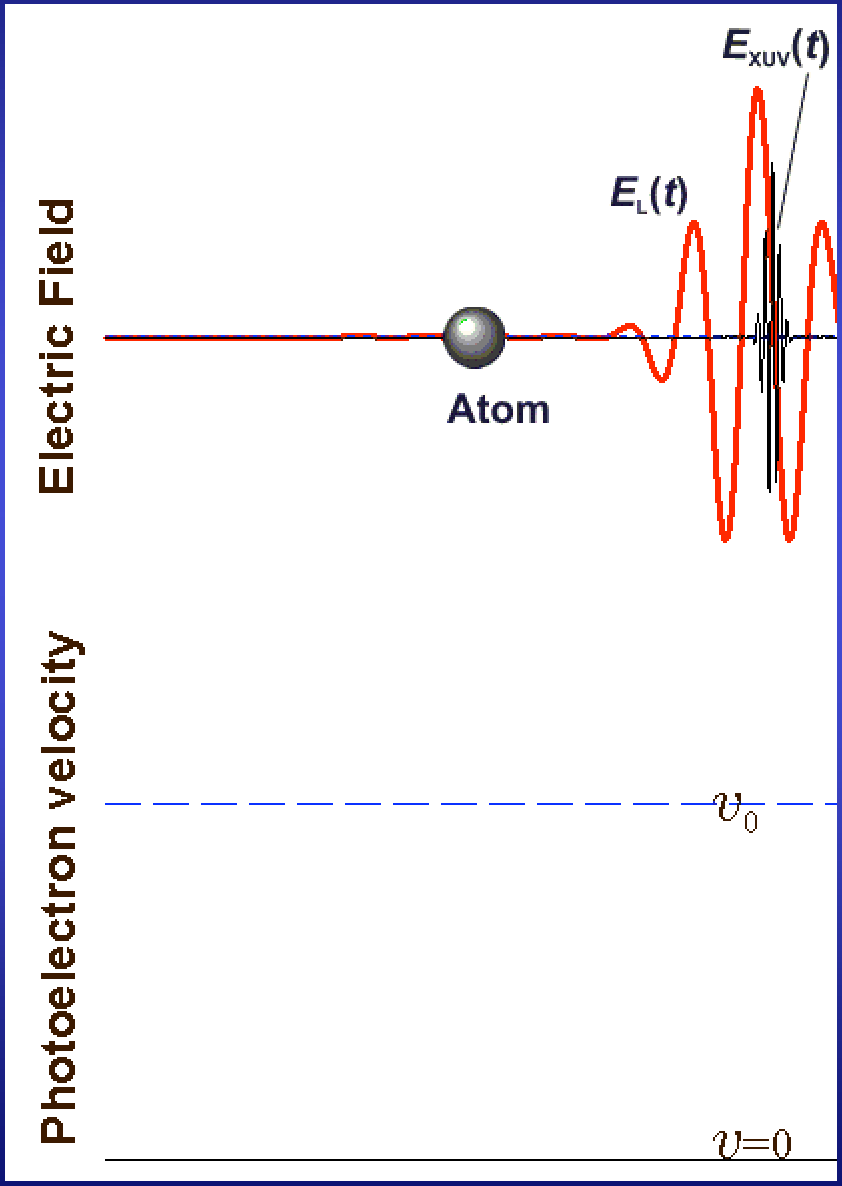


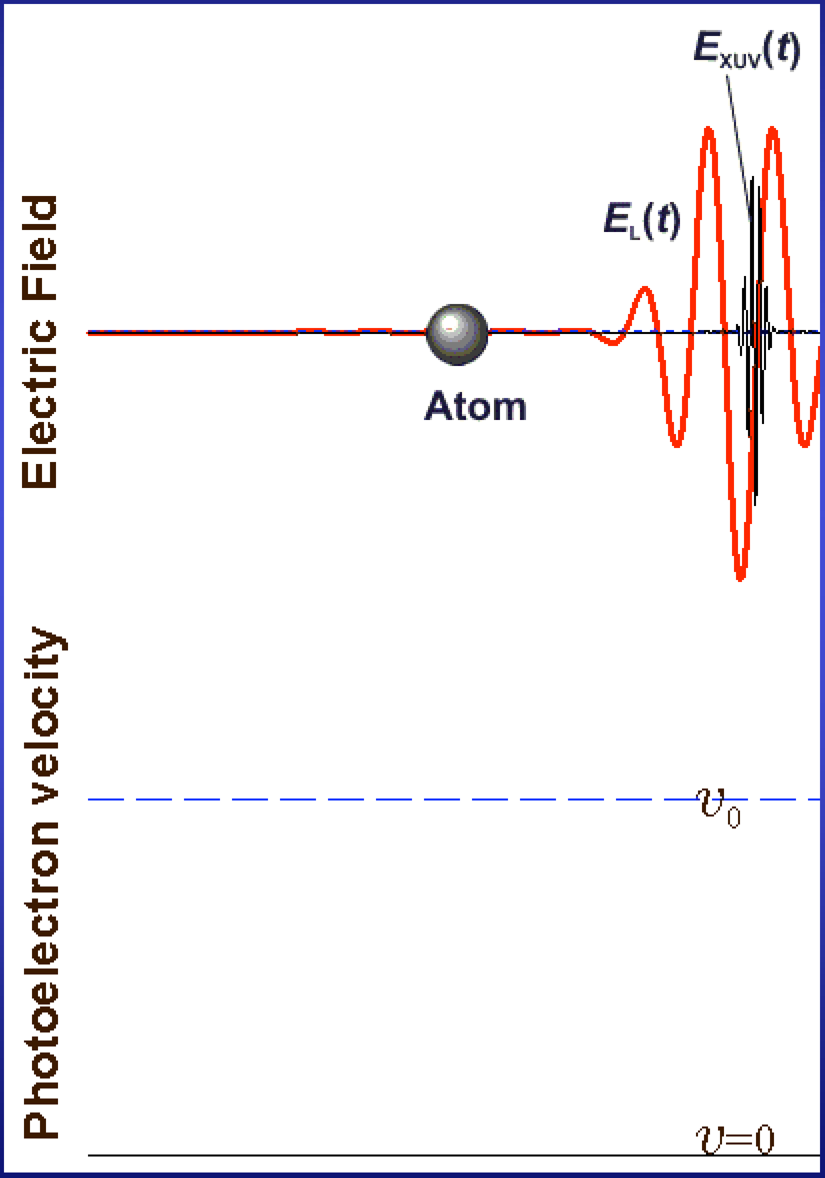
Detection as in:
Kienberger *et al.*, Science **297**, 1144 (2002)

Gas: Ne
Electrons: 2p
W_b= 21.46 eV

XUV cut-off energy: ~95 eV
Mirror reflectivity bandwidth: ~9 eV (FWHM)

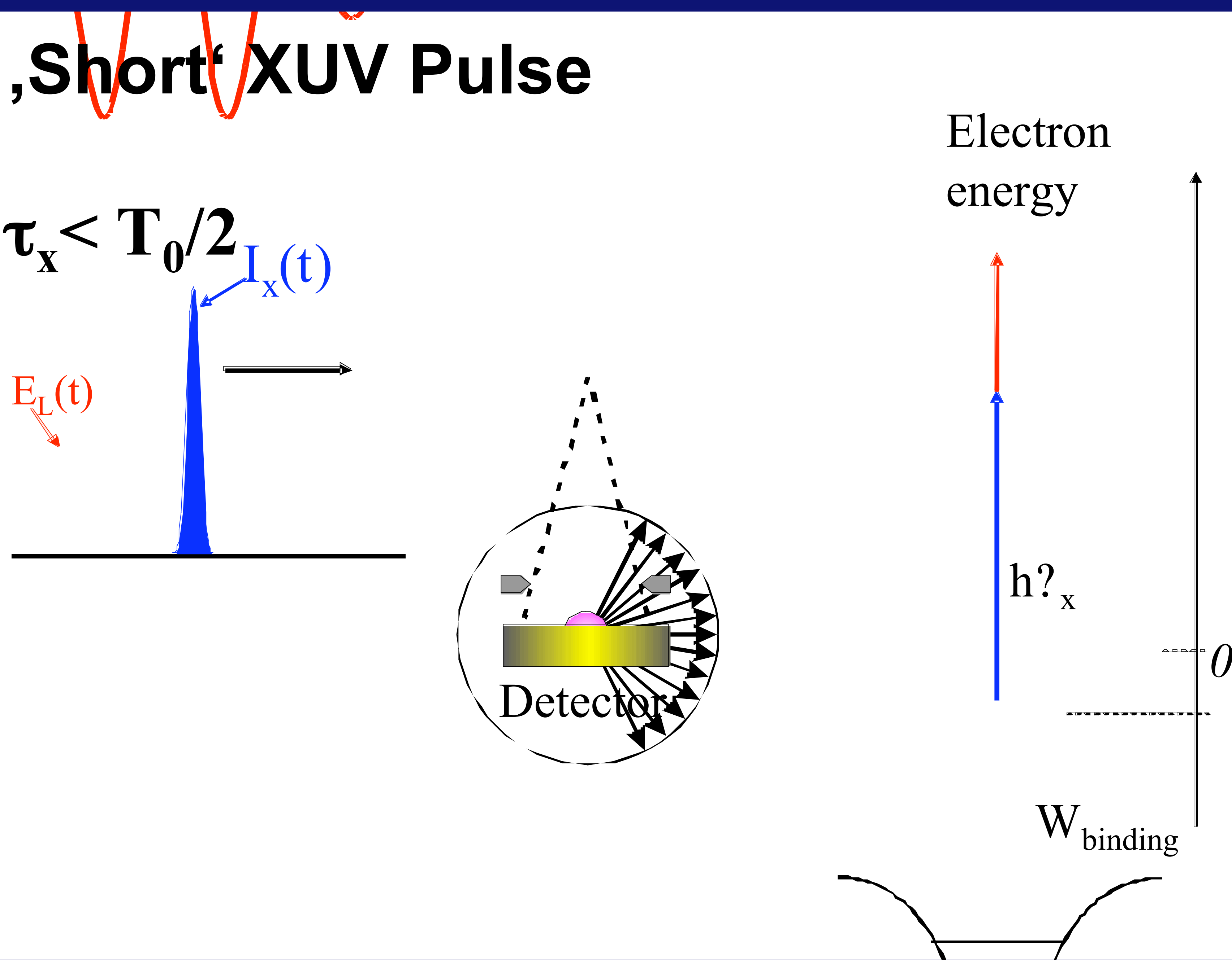
Photoelectron Acceleration/Deceleration



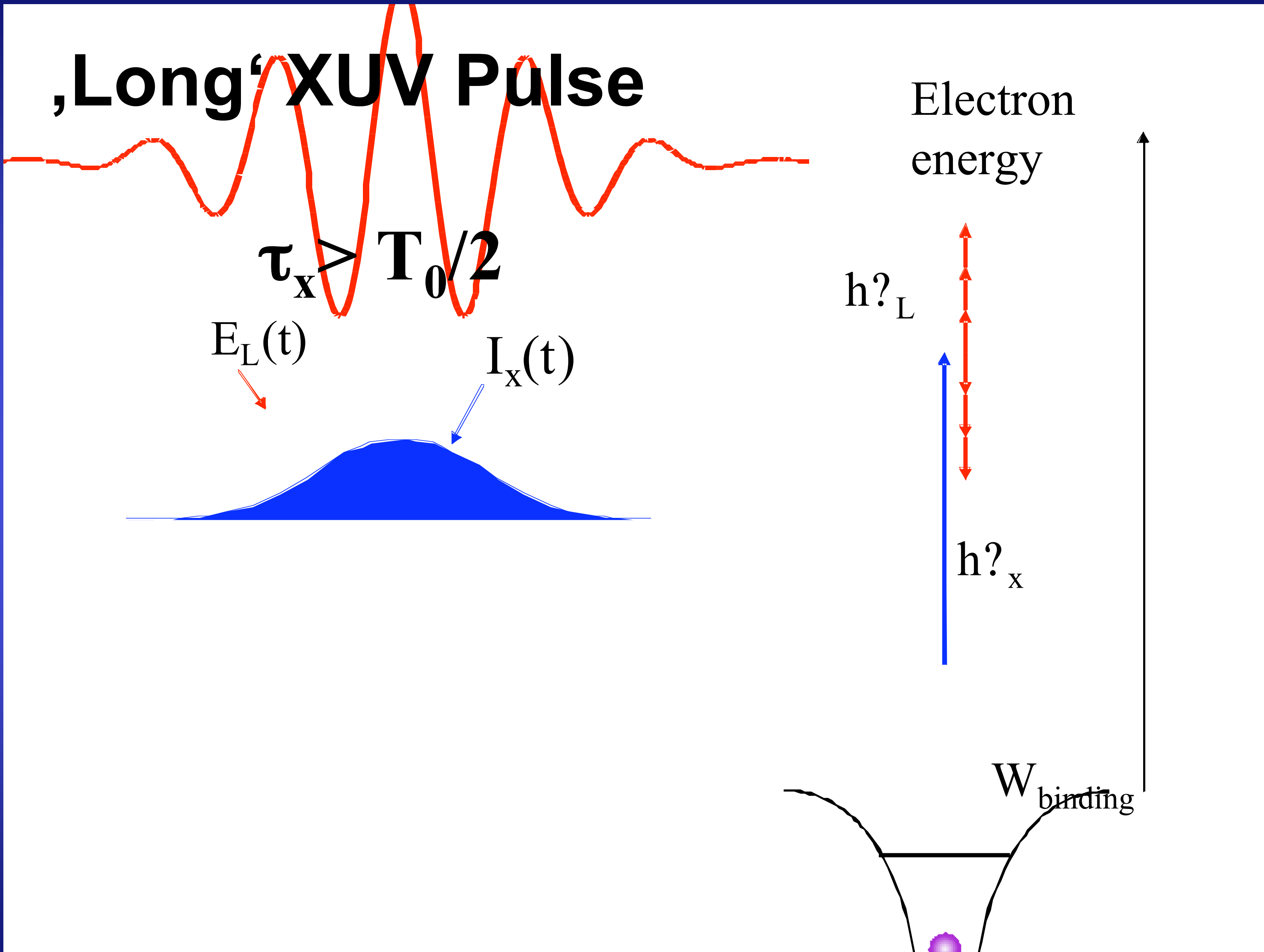


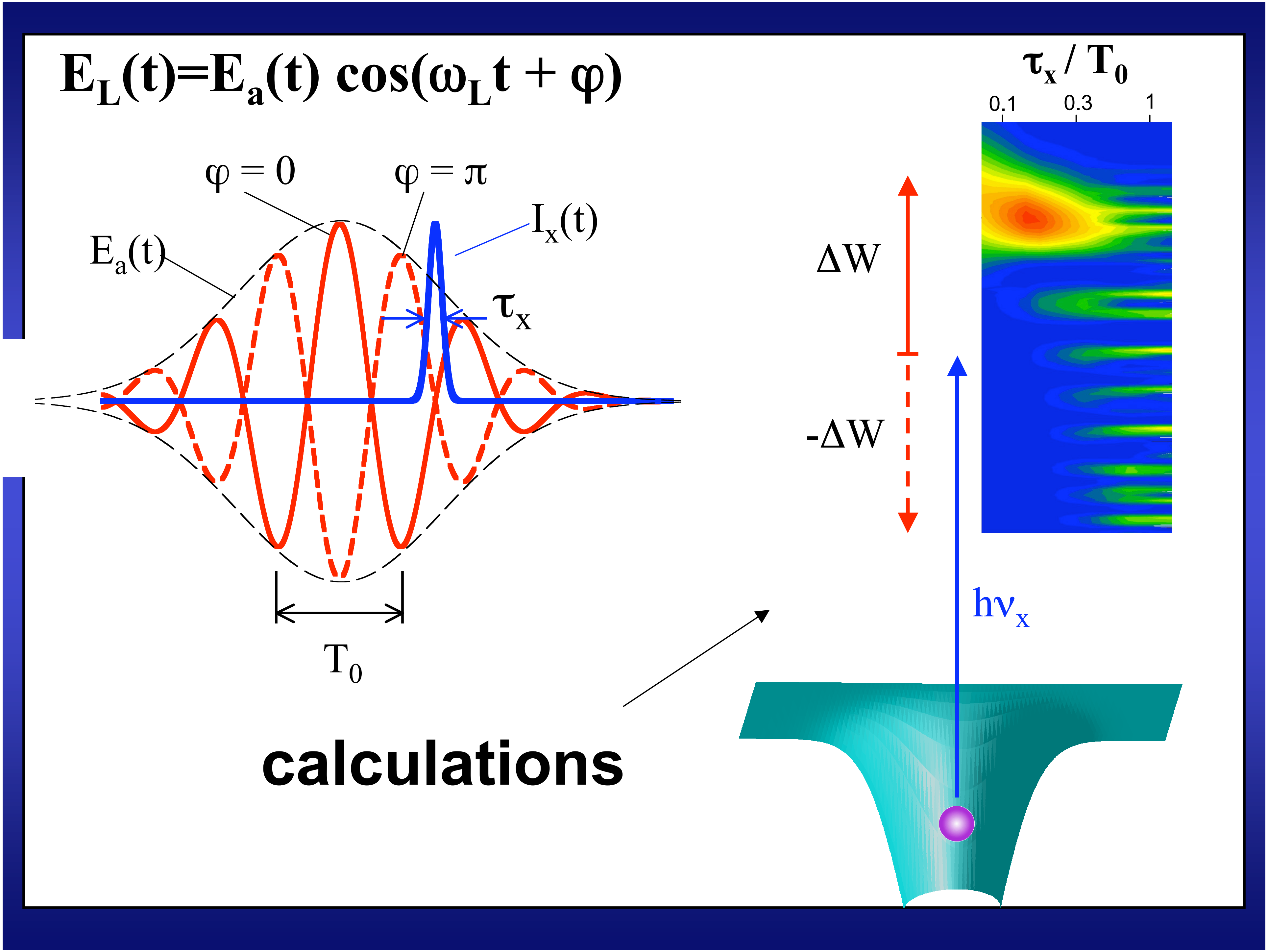
‚Short‘ XUV Pulse

$\tau_x < T_0/2$



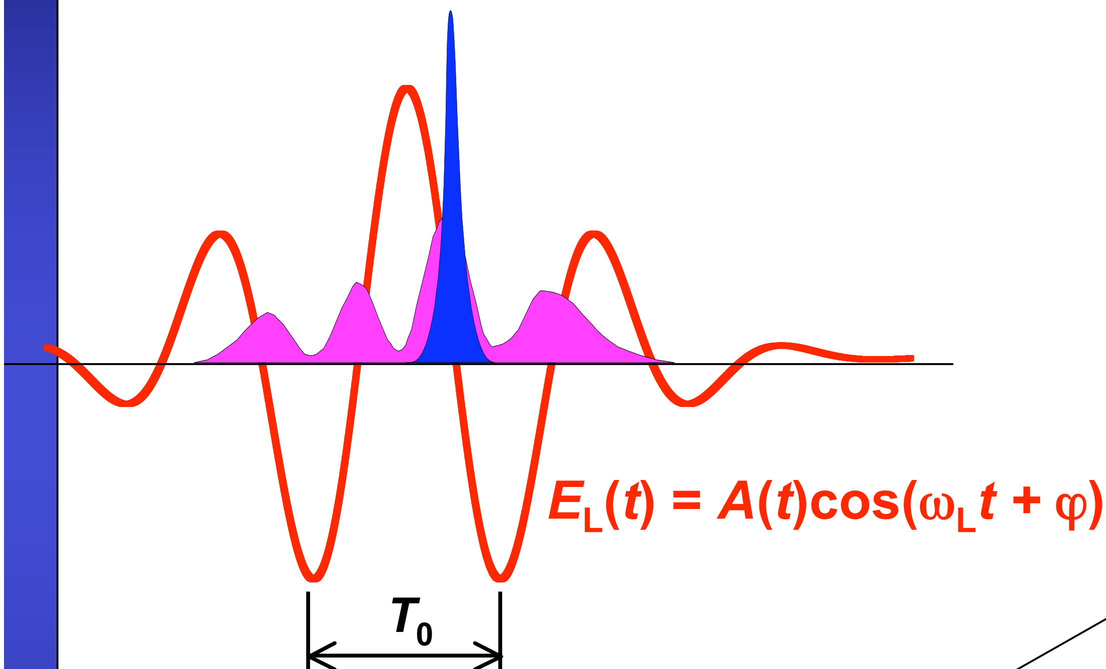
‚Long‘ XUV Pulse

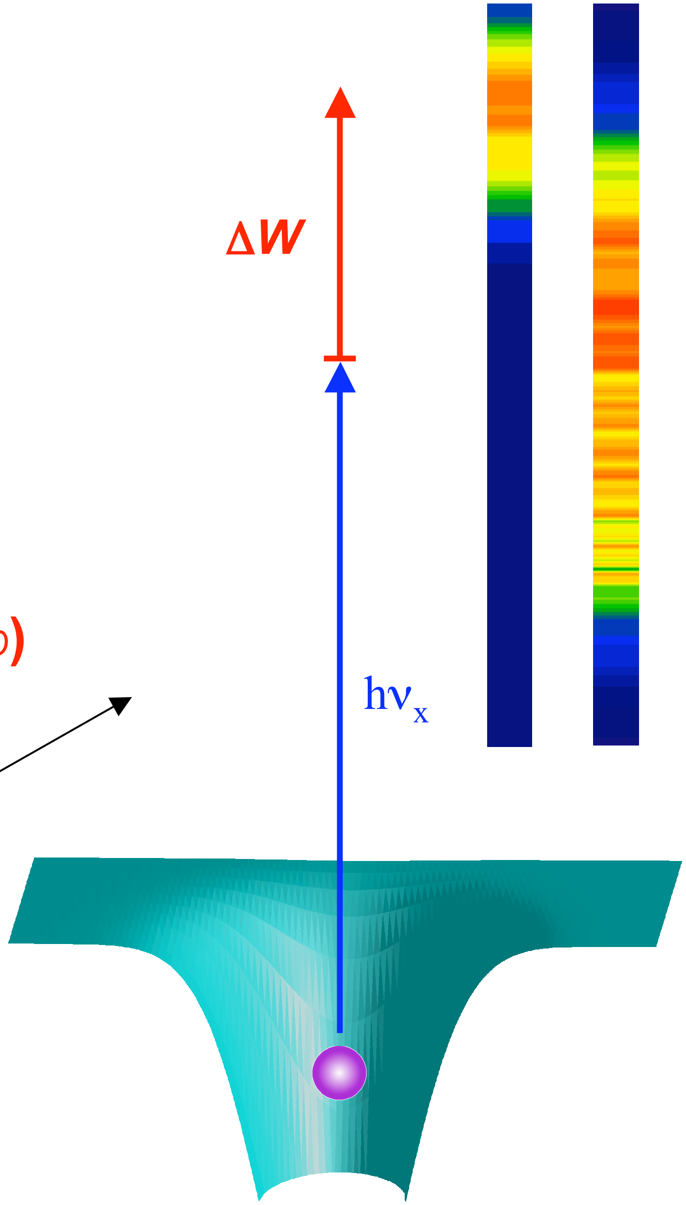




$E_L(t)=E_a(t)\cos(\omega_L t + \varphi)$
$\varphi = 0$
$\varphi = \pi$
$I_x(t)$
$E_a(t)$
τ_x
T_0
calculations
τ_x / T_0
0.1
0.3
1
ΔW
-ΔW
$h\nu_x$

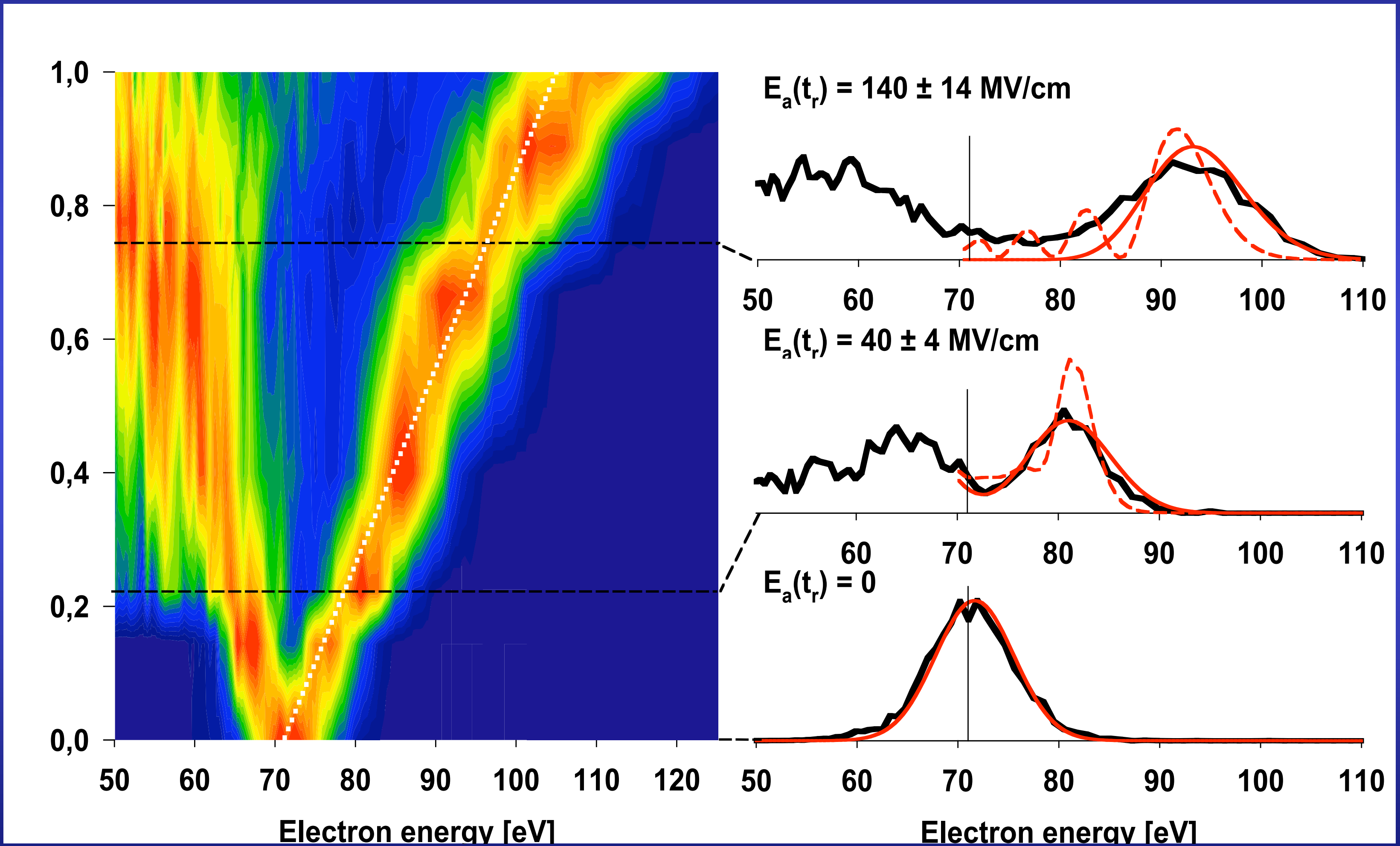
Cross-check: Attosecond Diagnostics





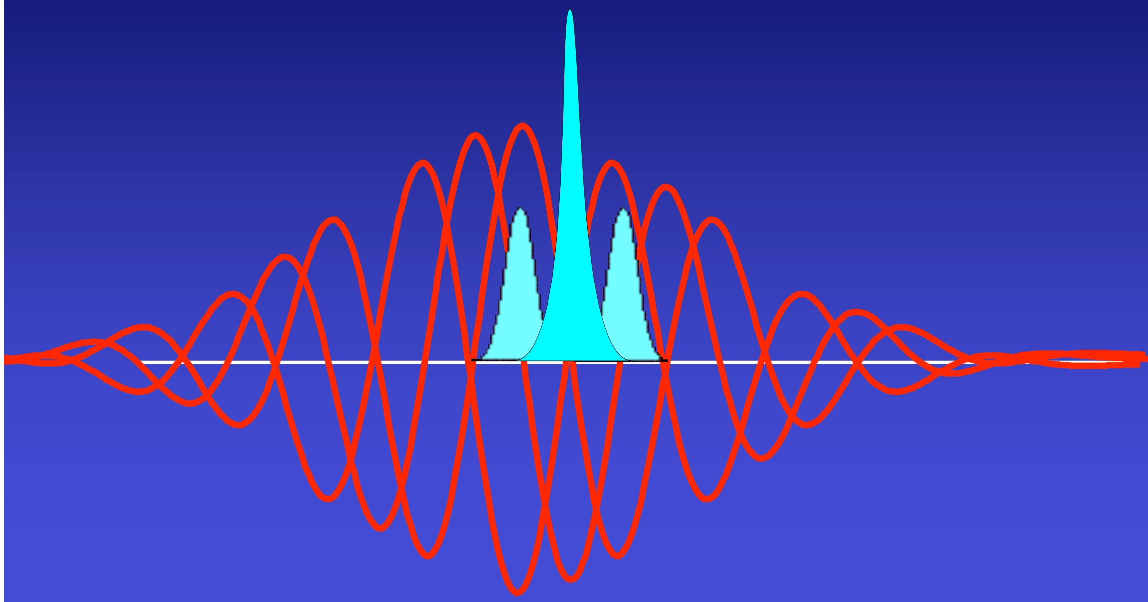
measurement

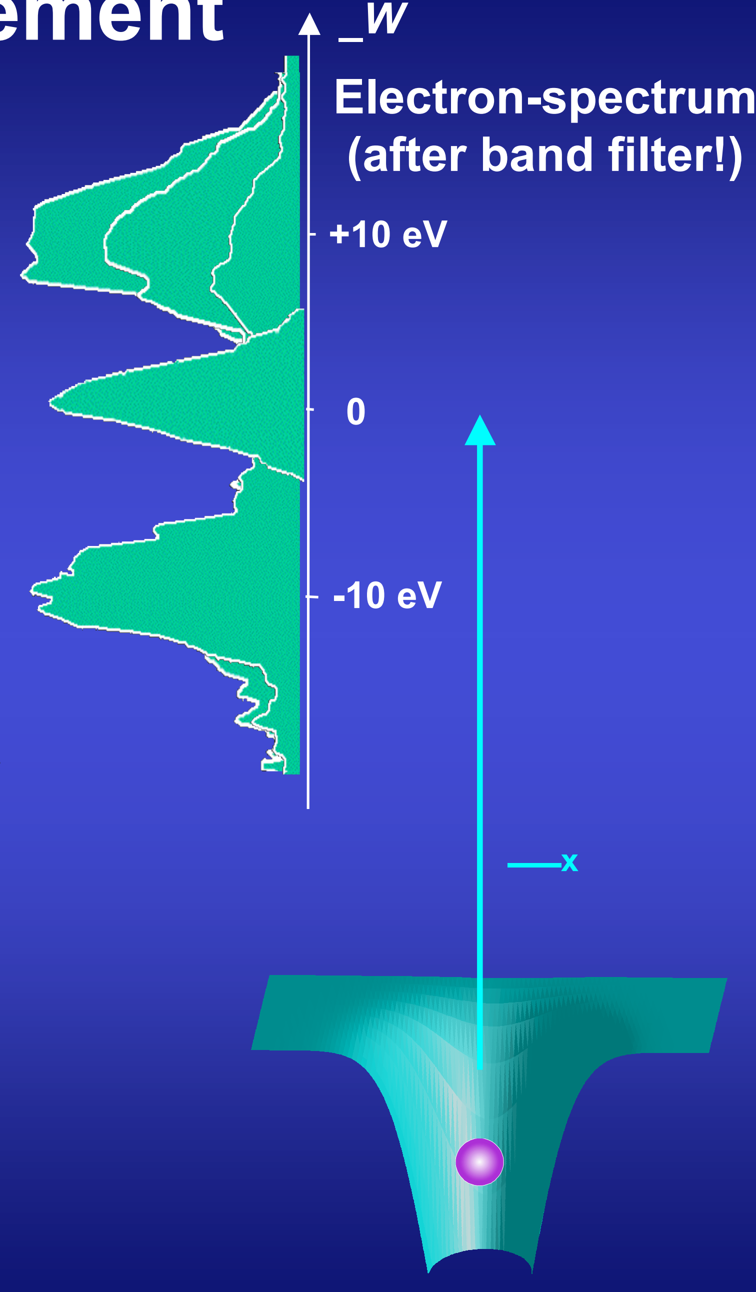
Electric Field Scan

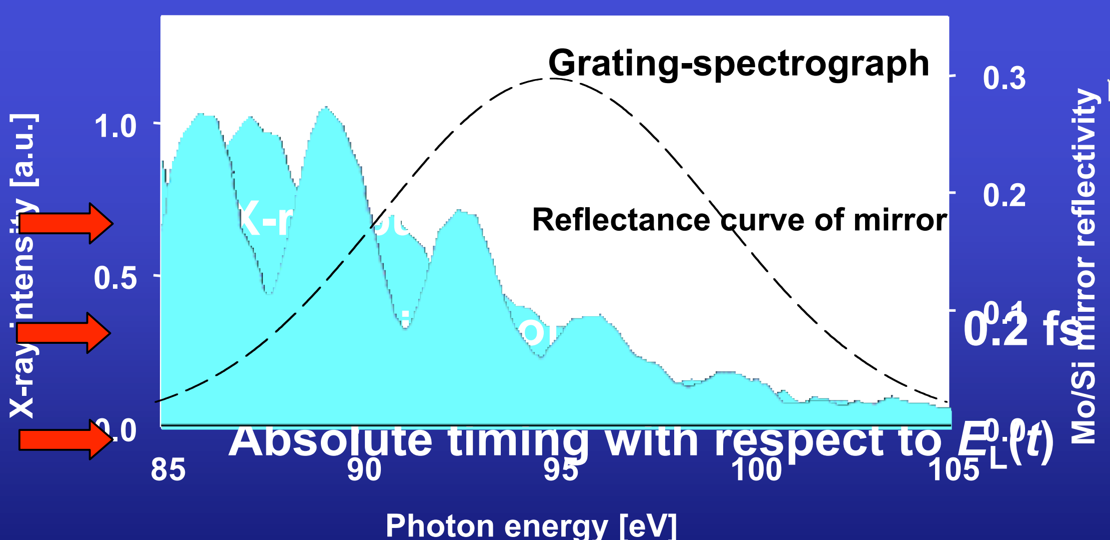


Kienberger *et al.*, Science **297**, 1444 (2002)

Advanced as Pulse Measurement

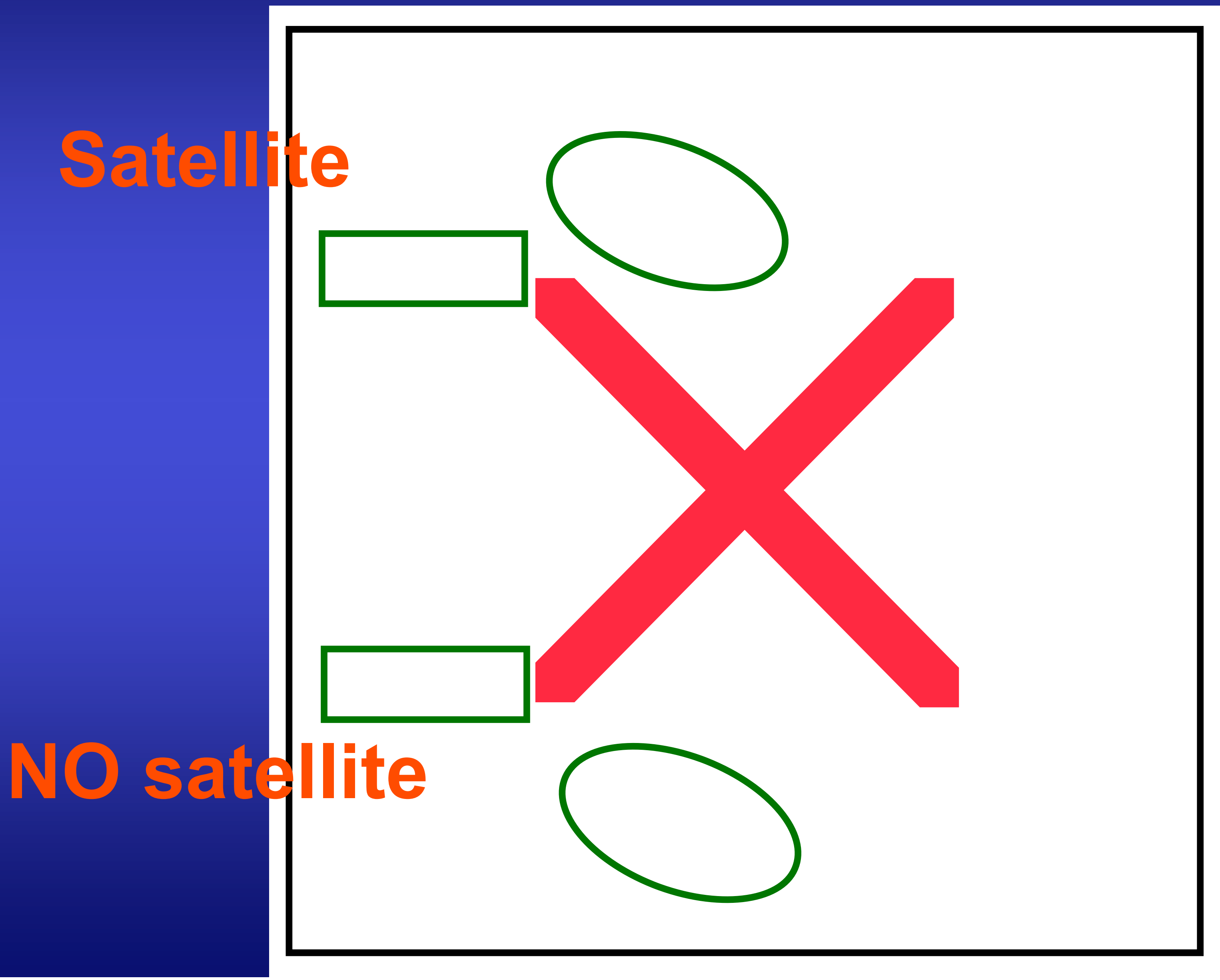






Kienberger *et al.*, Nature **427**, 817 (2004)

Streak Records of Sub-Femtosecond XUV Pulses



Outline

1.) The tools:

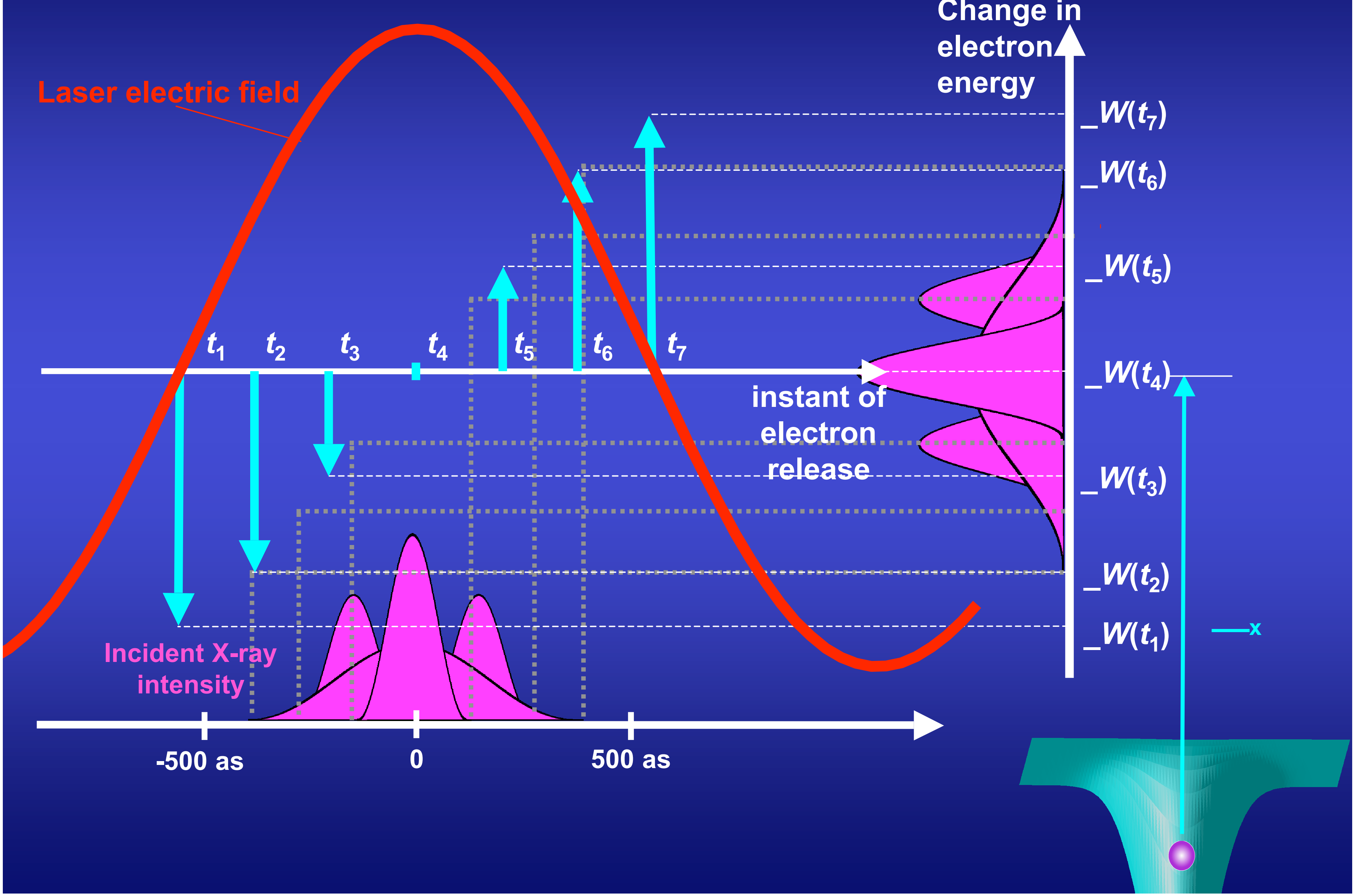
- Phase-stabilized few-opt.-cycle laser pulses
- Single as pulses: High-order Harmonic Generation

2.) Attosecond pulse measurement

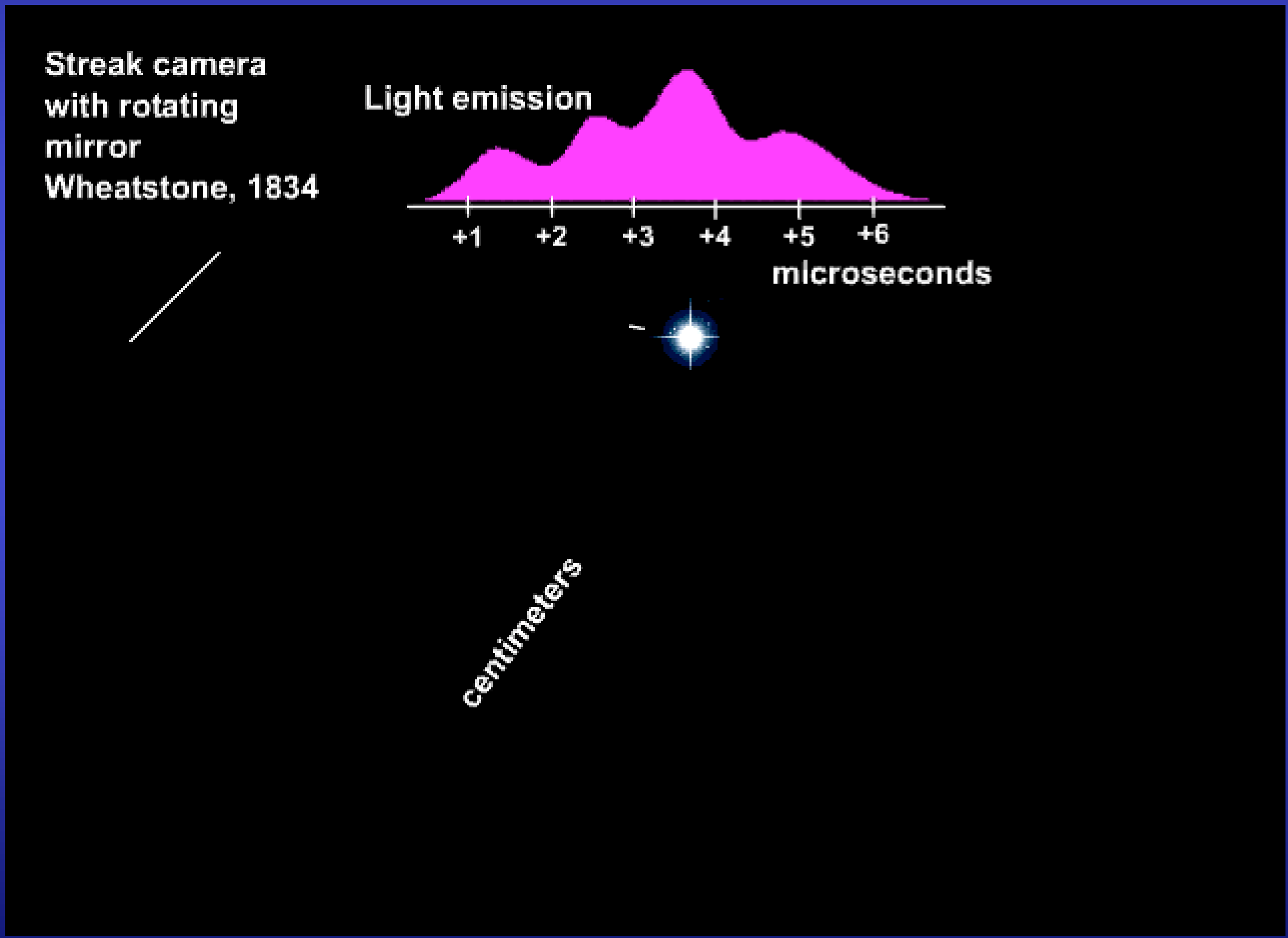
- Photoelectron spectra
- Attosecond streak camera

3.) Application: Spectroscopy

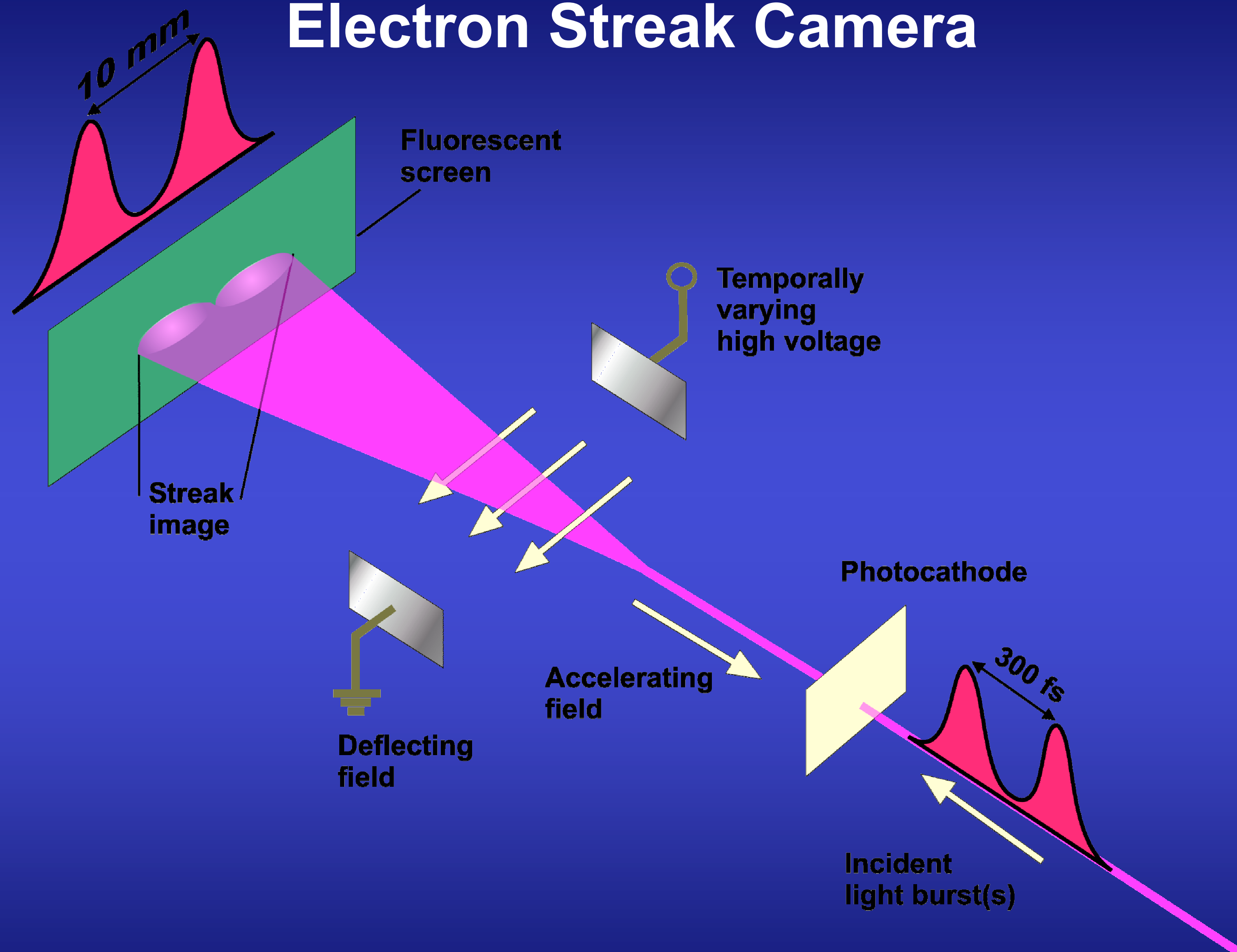
Mapping Time to Energy



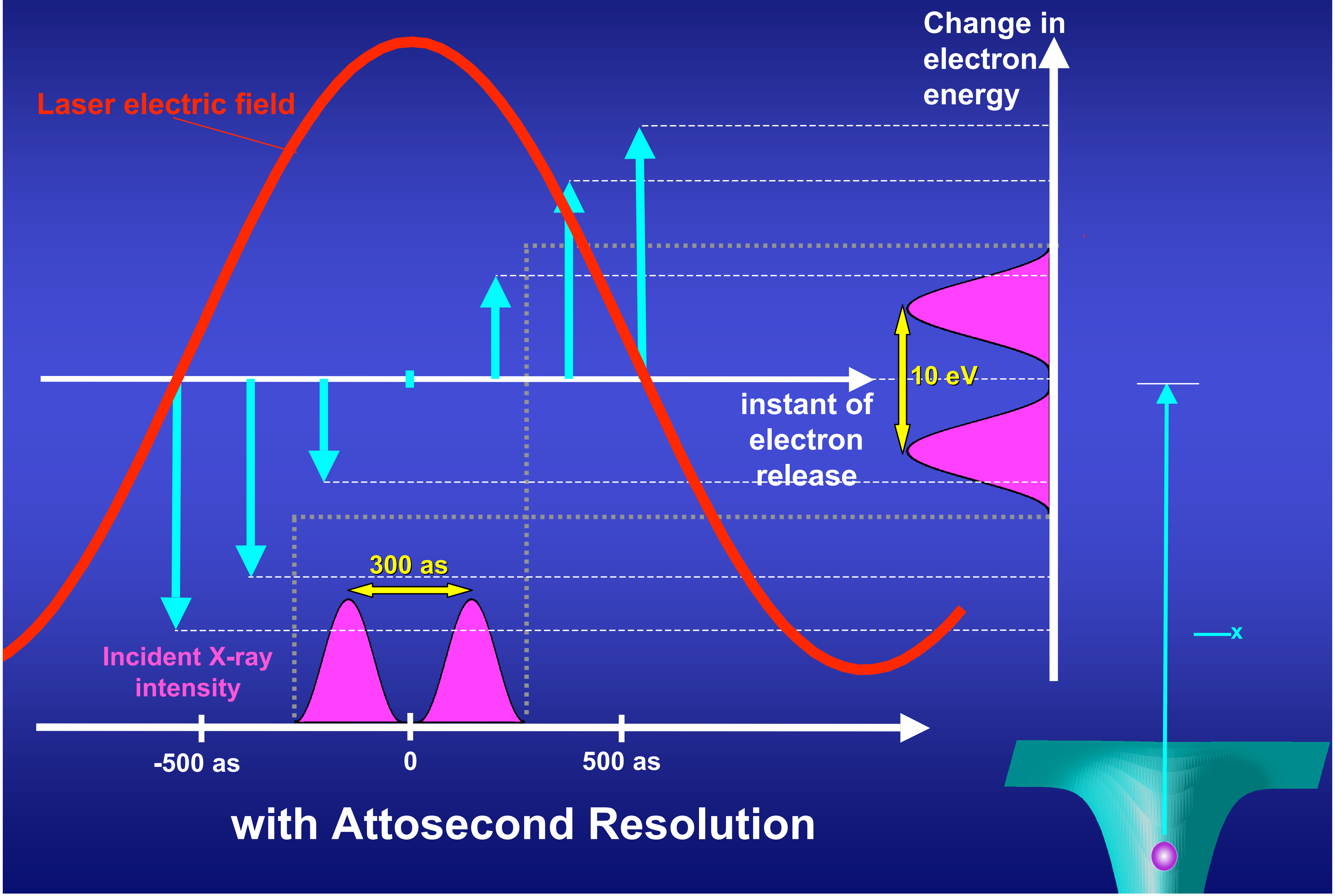
Optical Streak Camera, 1834



Electron Streak Camera

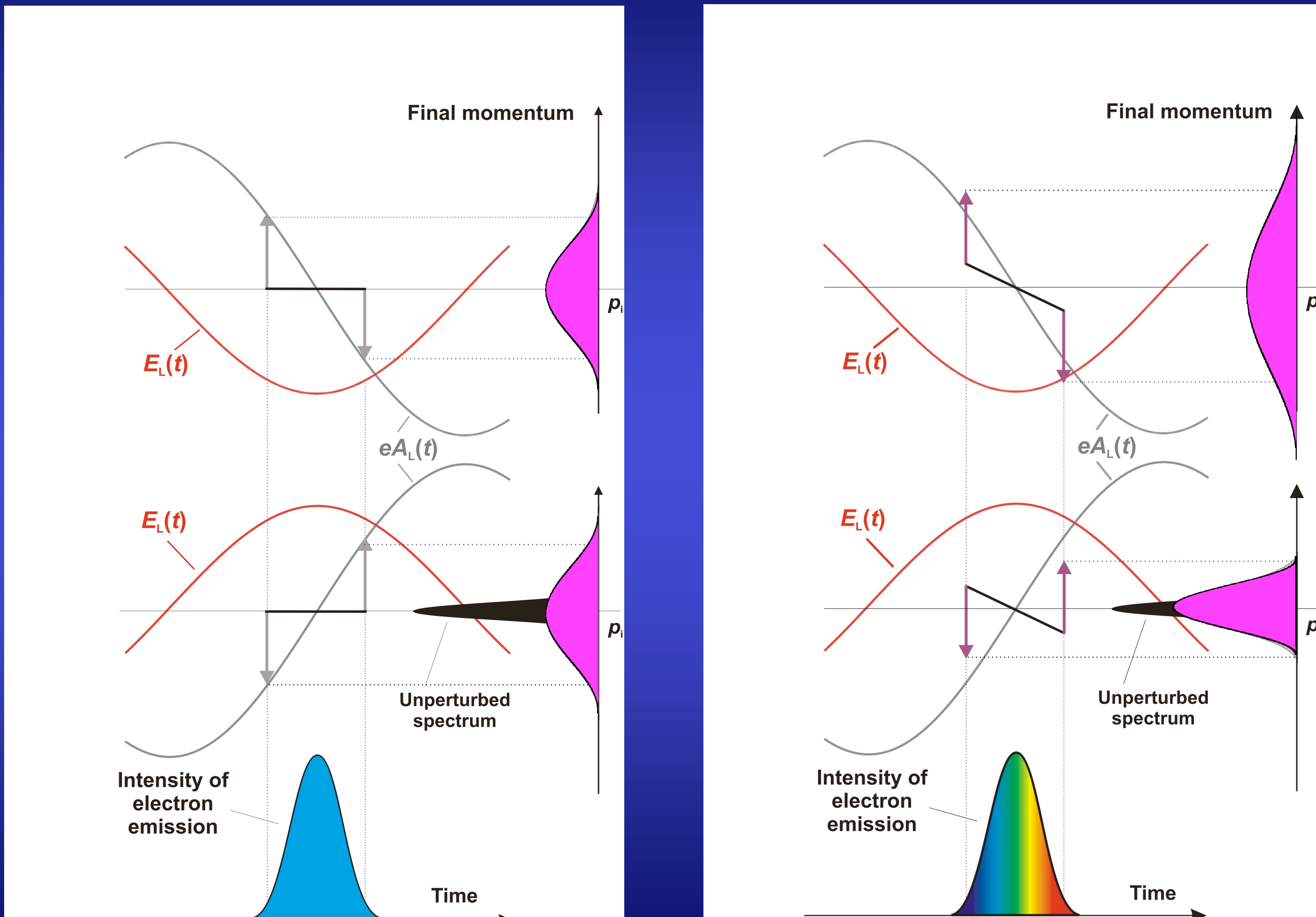


Optical-Field-Driven Streak Camera



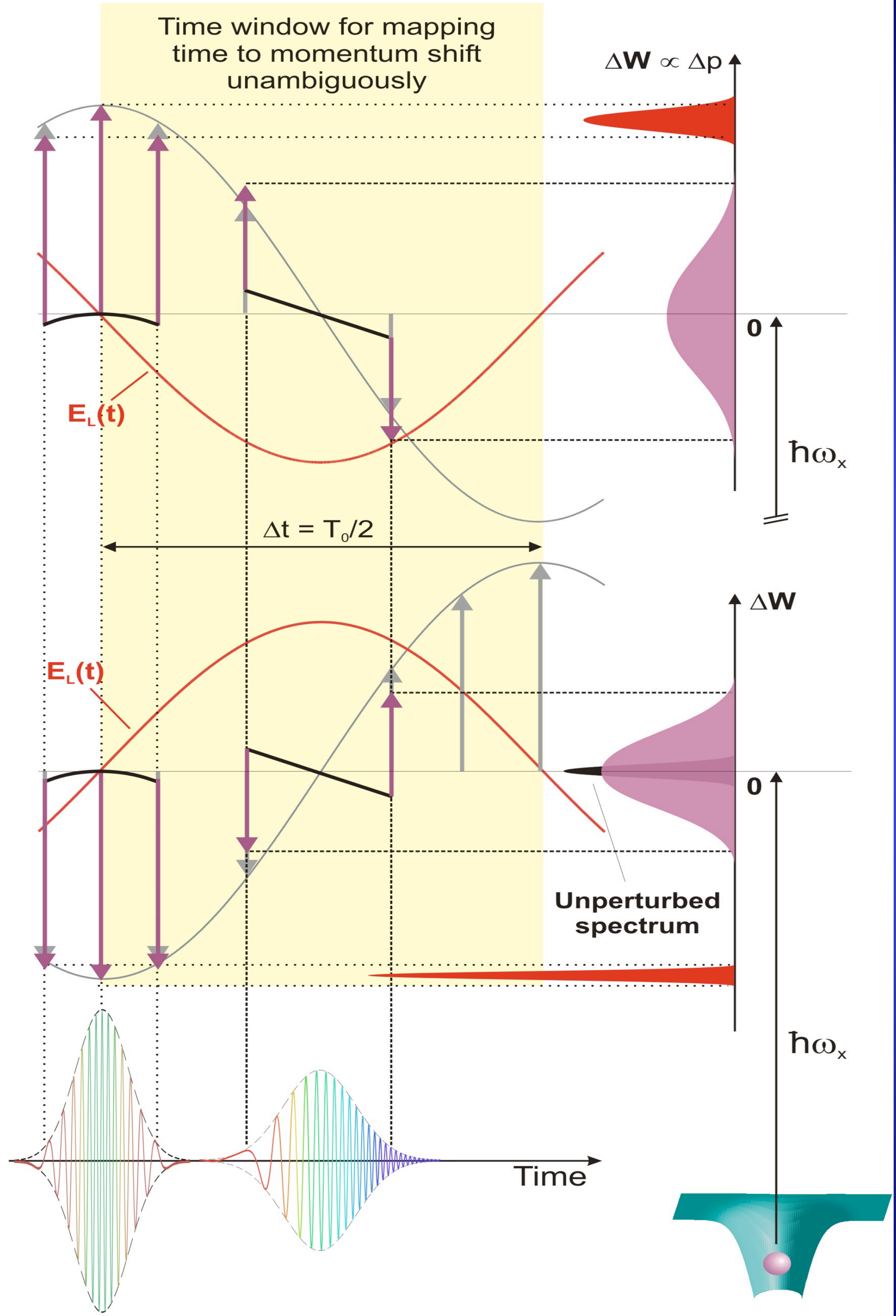
with Attosecond Resolution

Atomic Transient Recorder Concept



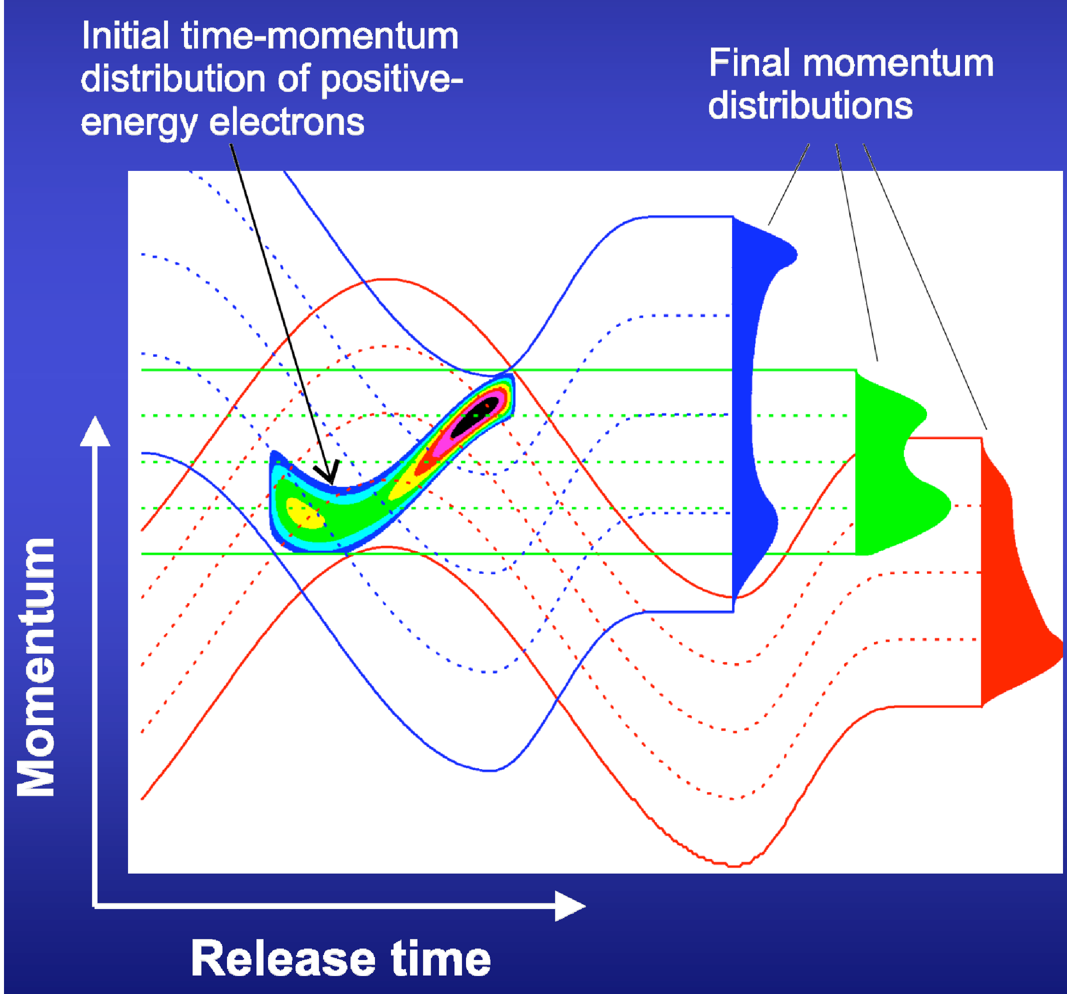
Fourier transform limited pulse (el. bunch)

Linearly chirped case



Time window for mapping
time to momentum shift
unambiguously
ΔW ∝ Δp
0
$E_L(t)$
ħω$_x$
Δt = $T_0/2$
ΔW
$E_L(t)$
0
Unperturbed
spectrum
ħω$_x$
Time

Atomic Transient Recorder:
tomographic images of electron distribution

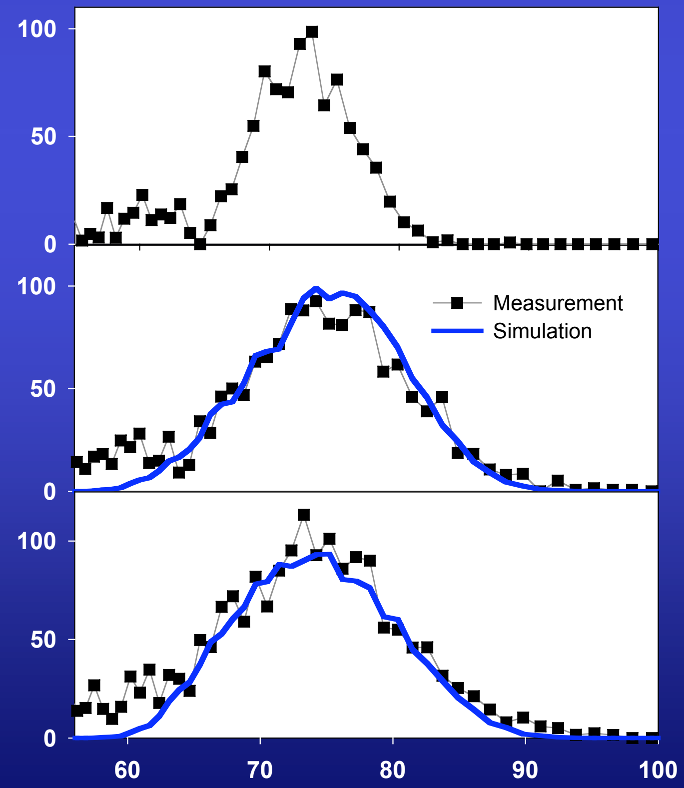


Reconstructs the time-momentum distribution of atomic electron emission confined to $T_0/2$ from "tomographic images" recorded by a strong, phase-controlled light field

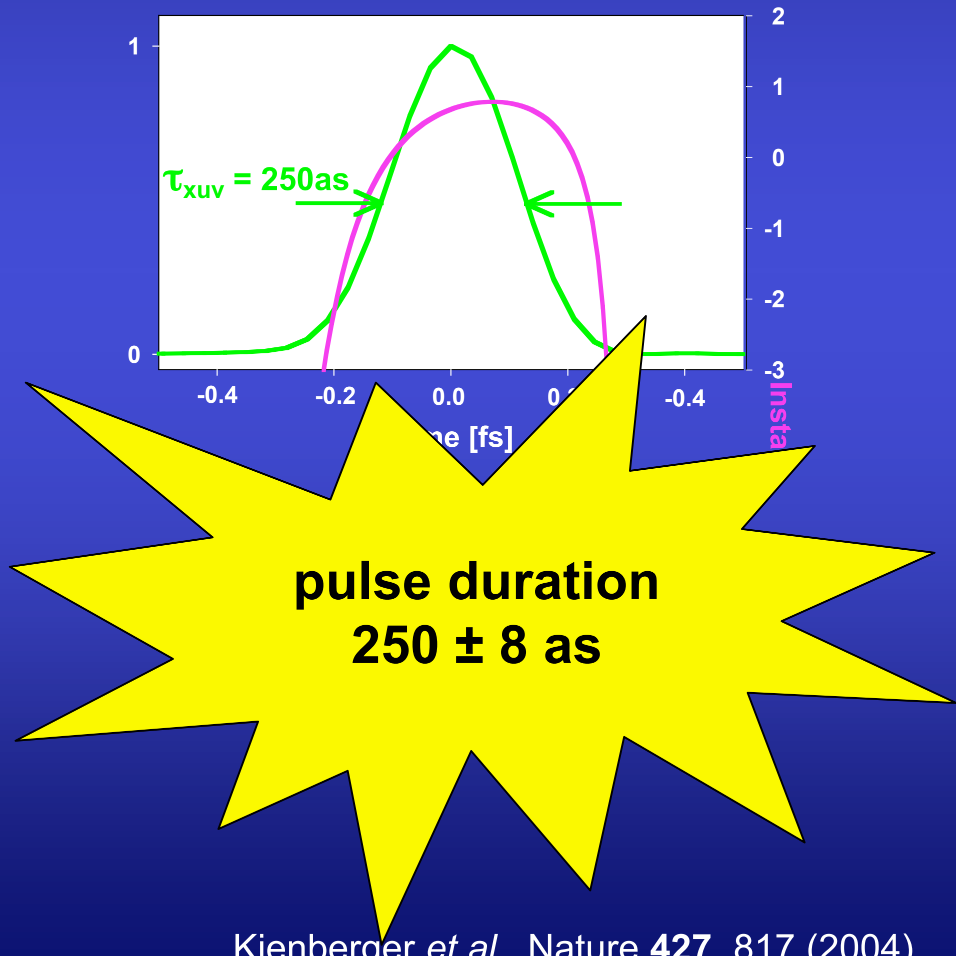
Resolves the time evolution of atomic excitation and relaxation processes on an attosecond time scale by probing primary (photo) or secondary (Auge electron emission, respectively.

Full Characterization of a Sub-Femtosecond XUV Pulse

“Streak images“ of photoelectrons emitted at adjacent field oscillation maxima of the probing field $E_L(t)$



Reconstructed temporal intensity profile and chirp of the XUV excitation pulse

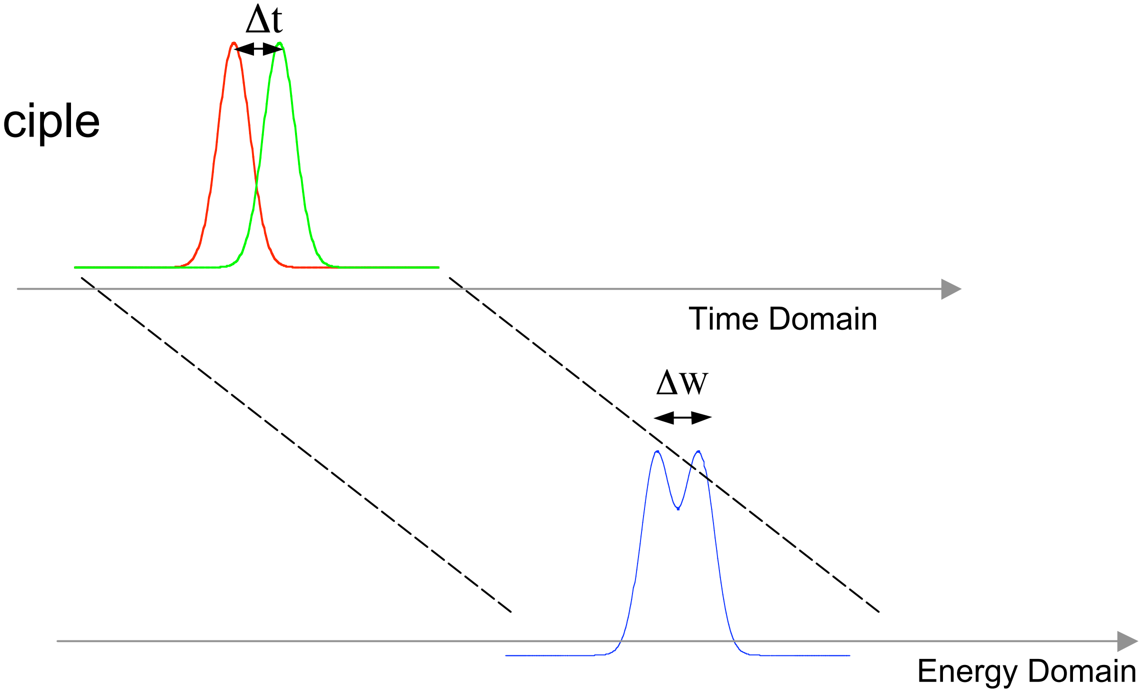


Kienberger *et al.*, Nature **427**, 817 (2004)

Resolution of measurement

Time - energy uncertainty principle

$\Delta E \Delta t$=_



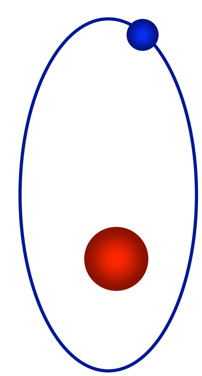
$$\delta t = \frac{T_0}{2\pi}\sqrt{\frac{\hbar\omega_L}{\Delta W_{max}}}$$

Current Experiment

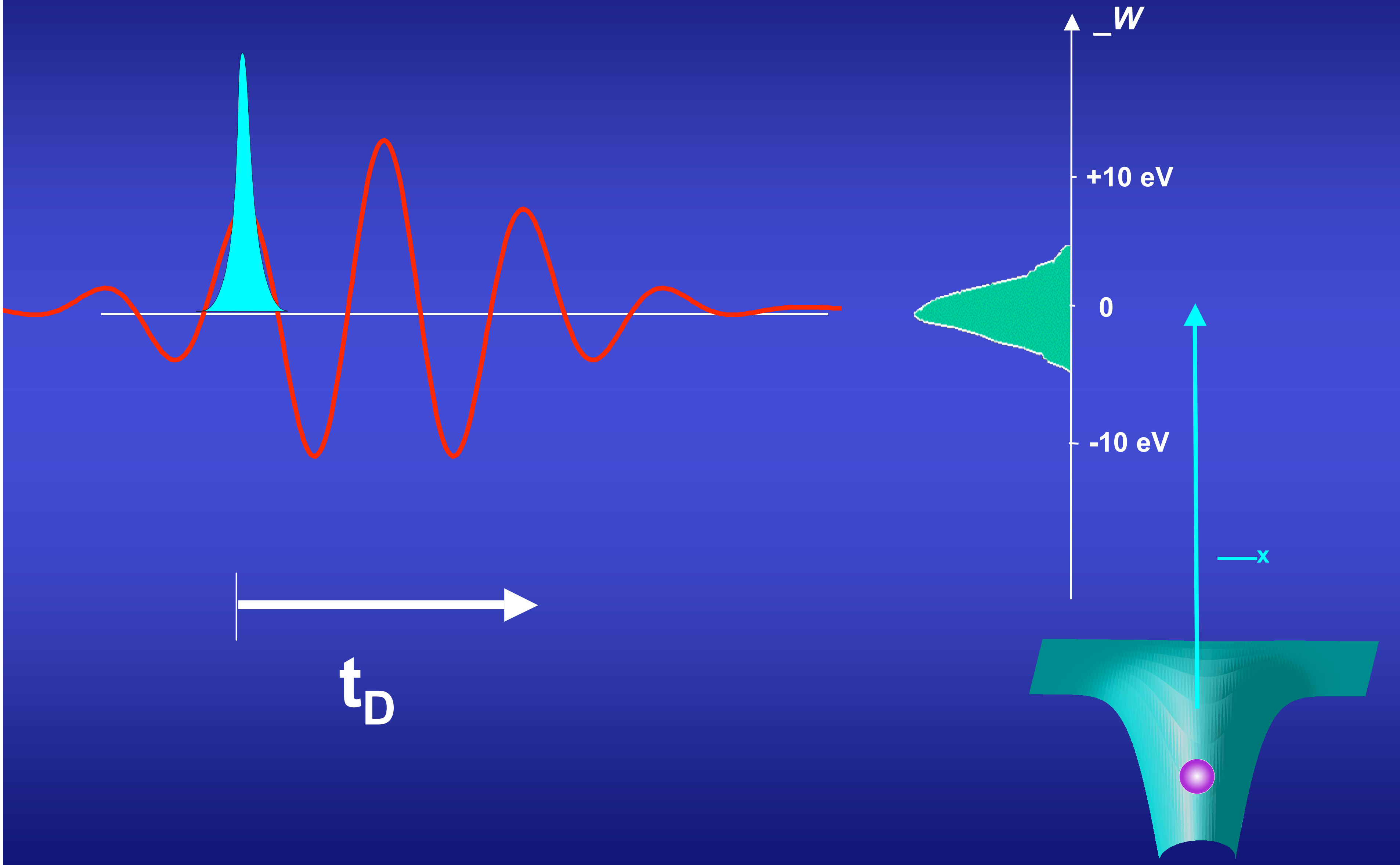
ΔW= 20 eV

T=2 fs

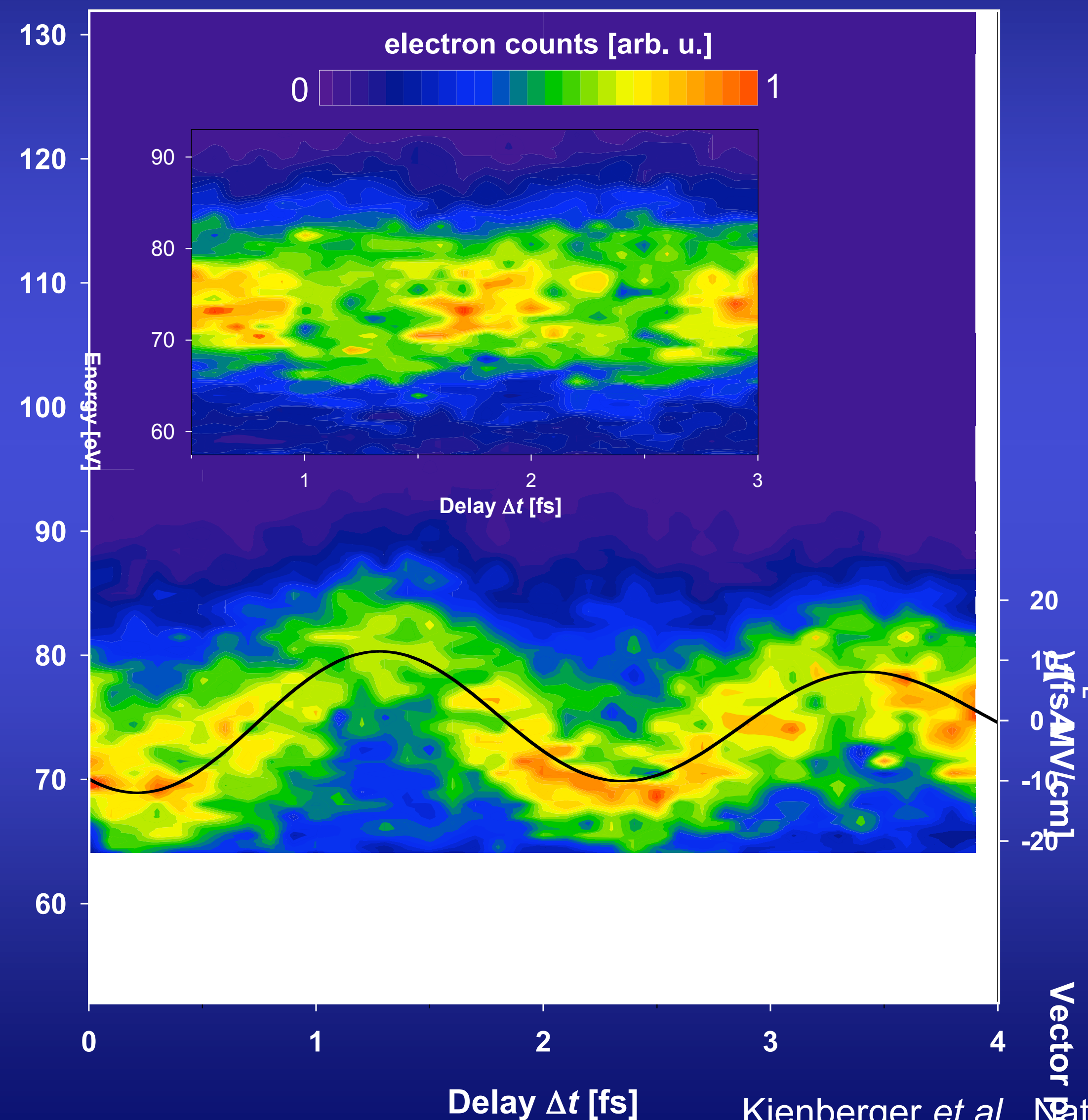
Δt ~ 100 as



Sampling Field Oscillations

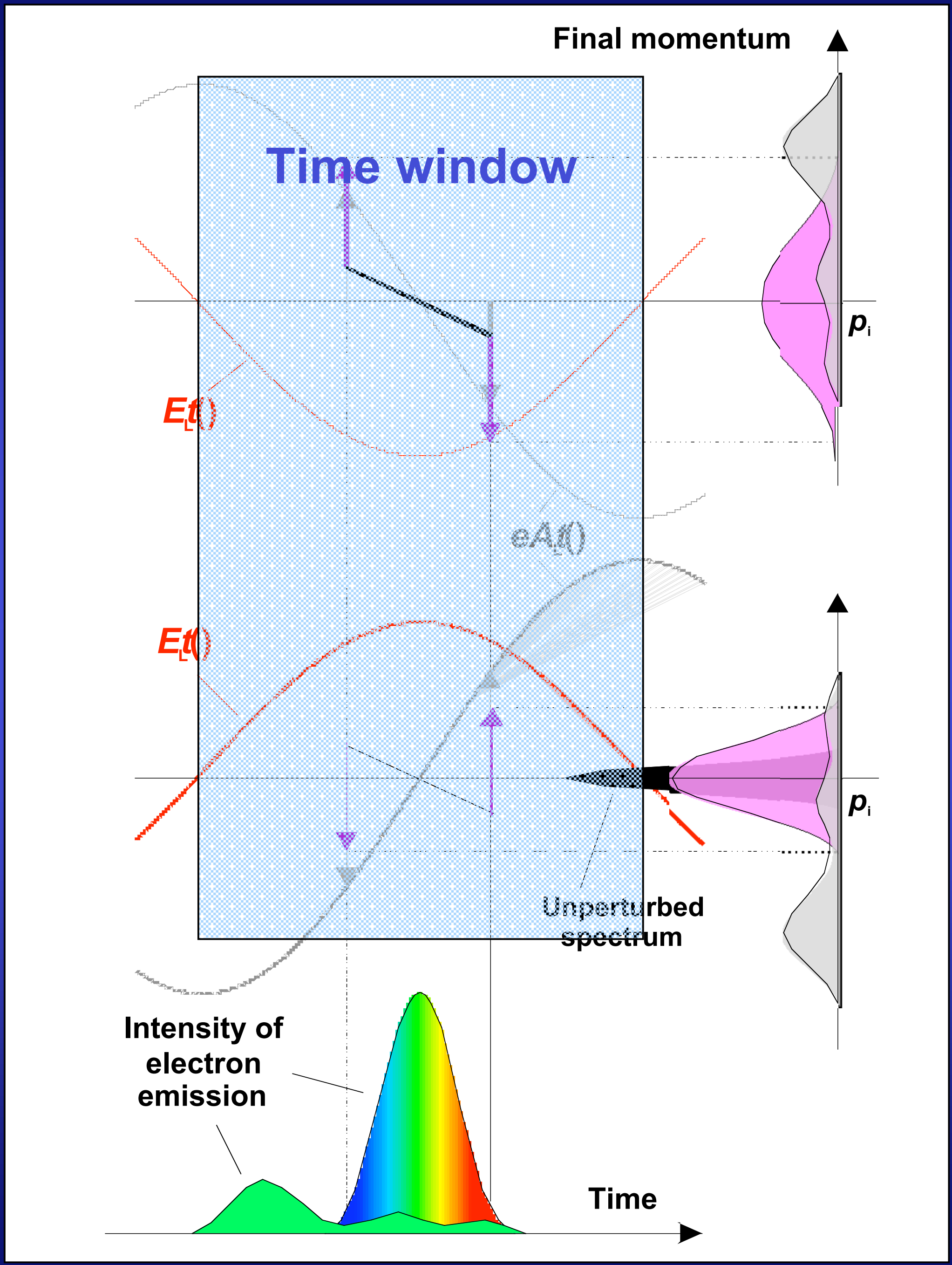


Sub-Femtosecond X-Ray Pulse Resolves Field Oscillations of Visible Light



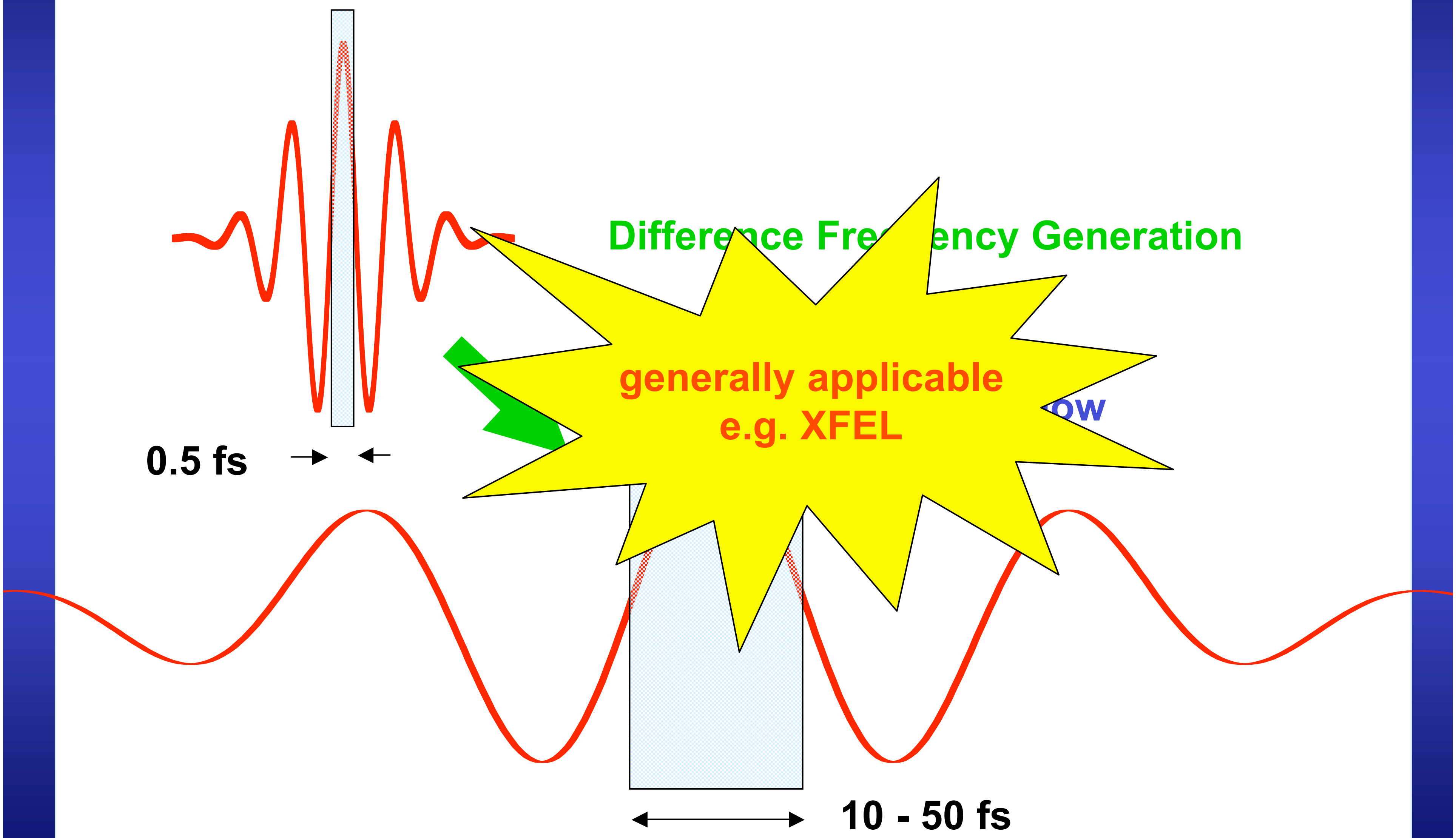
Kienberger *et al.*, Nature **427**, 817 (2004)

Outlook



Final momentum
Time window
p_i
$E_L(t)$
$eA_L(t)$
$E_L(t)$
p_i
Unperturbed spectrum
Intensity of electron emission
Time

Expanding the time window



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3. Application: From Attosecond Diagnostics to Attosecond Spectroscopy

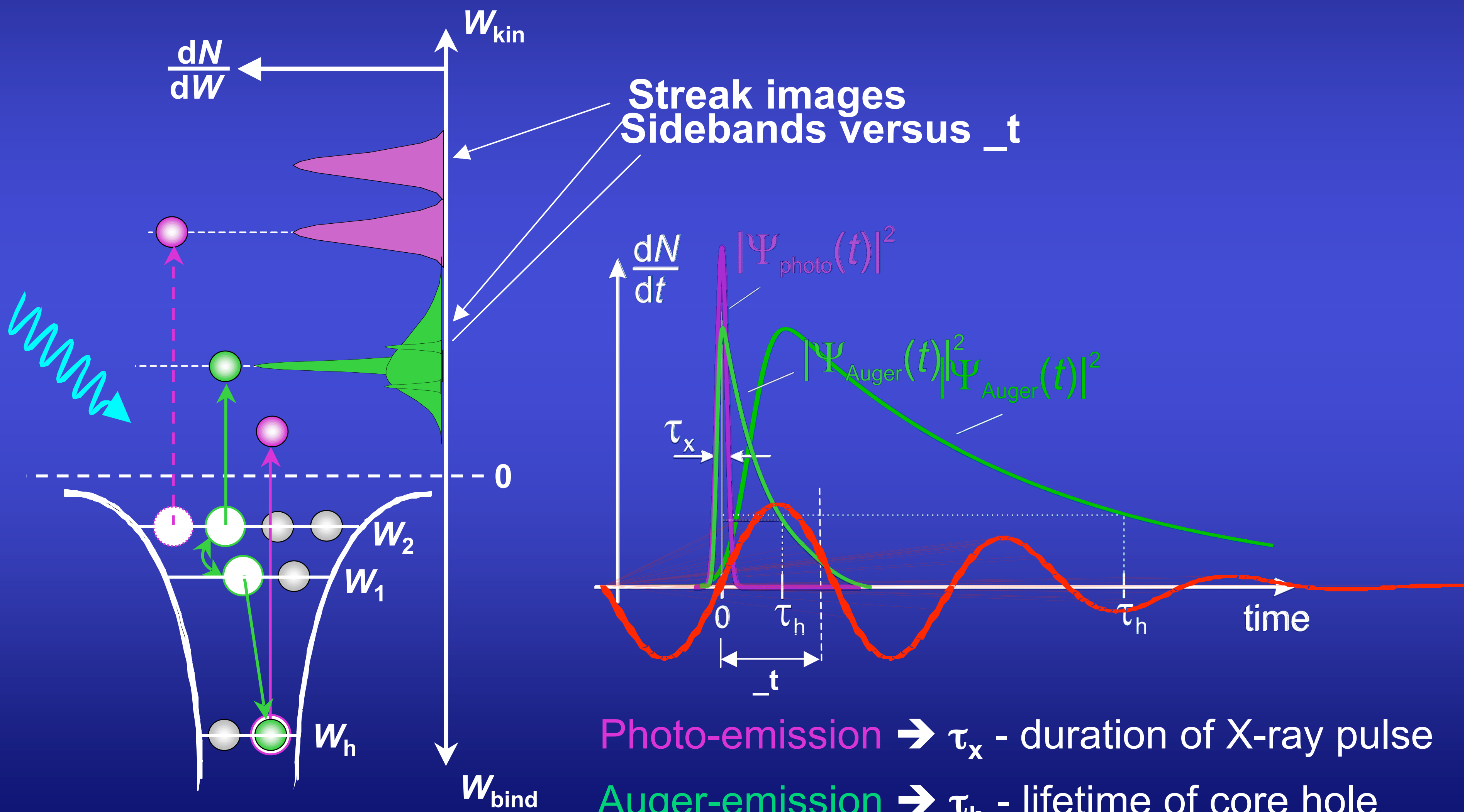
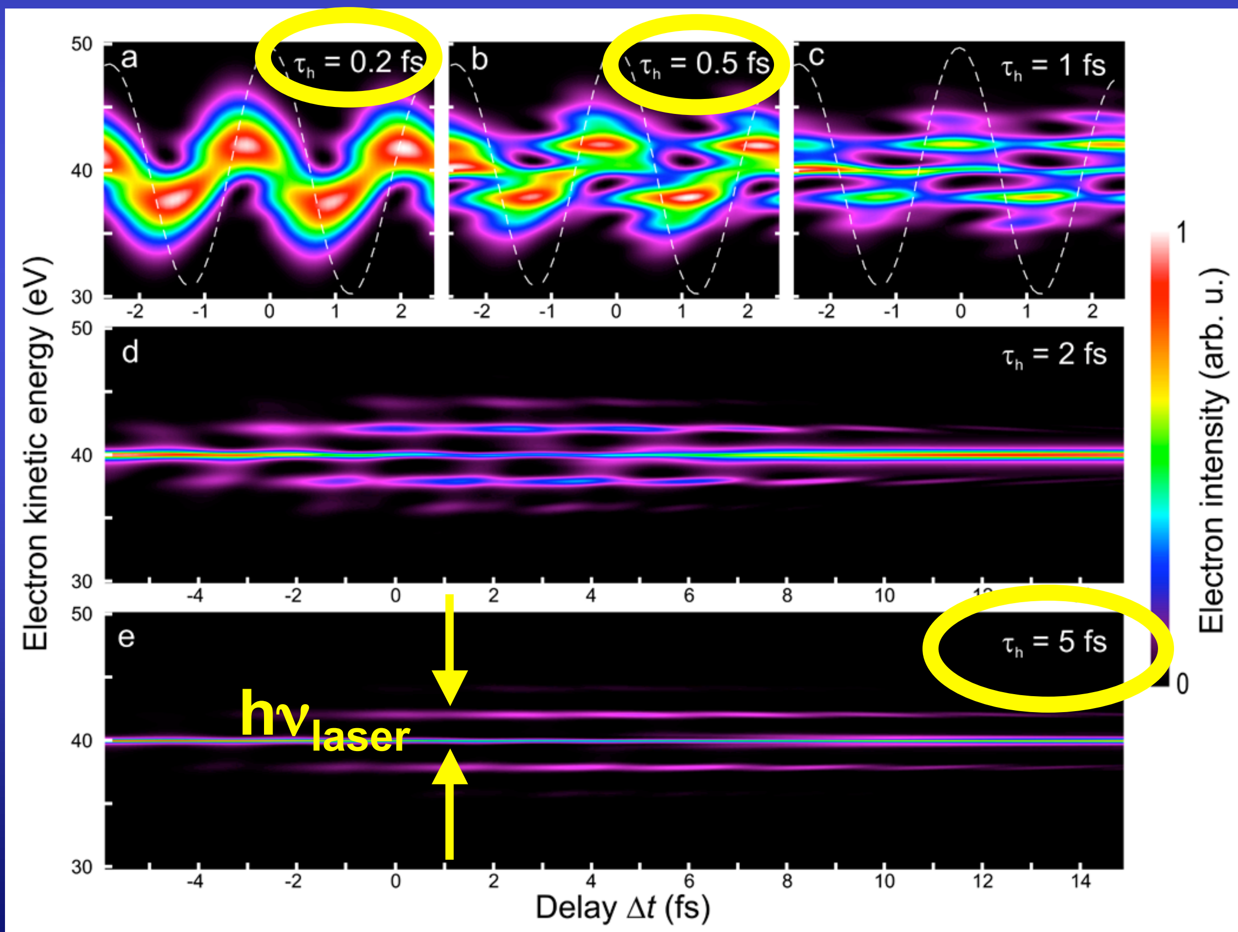


Photo-emission ➔ τ_x - duration of X-ray pulse

Auger-emission ➔ τ_h - lifetime of core hole

Sampling Auger Electron Emission in Parallel Detection Geometry: Simulations

Pump X-ray pulse, $\tau_X = 0.5$ fs
Probe laser, $T_0 = 2.5$ fs, $\tau_L = 5$ fs



$\tau_h < T_o/2$

Sampling by laser field

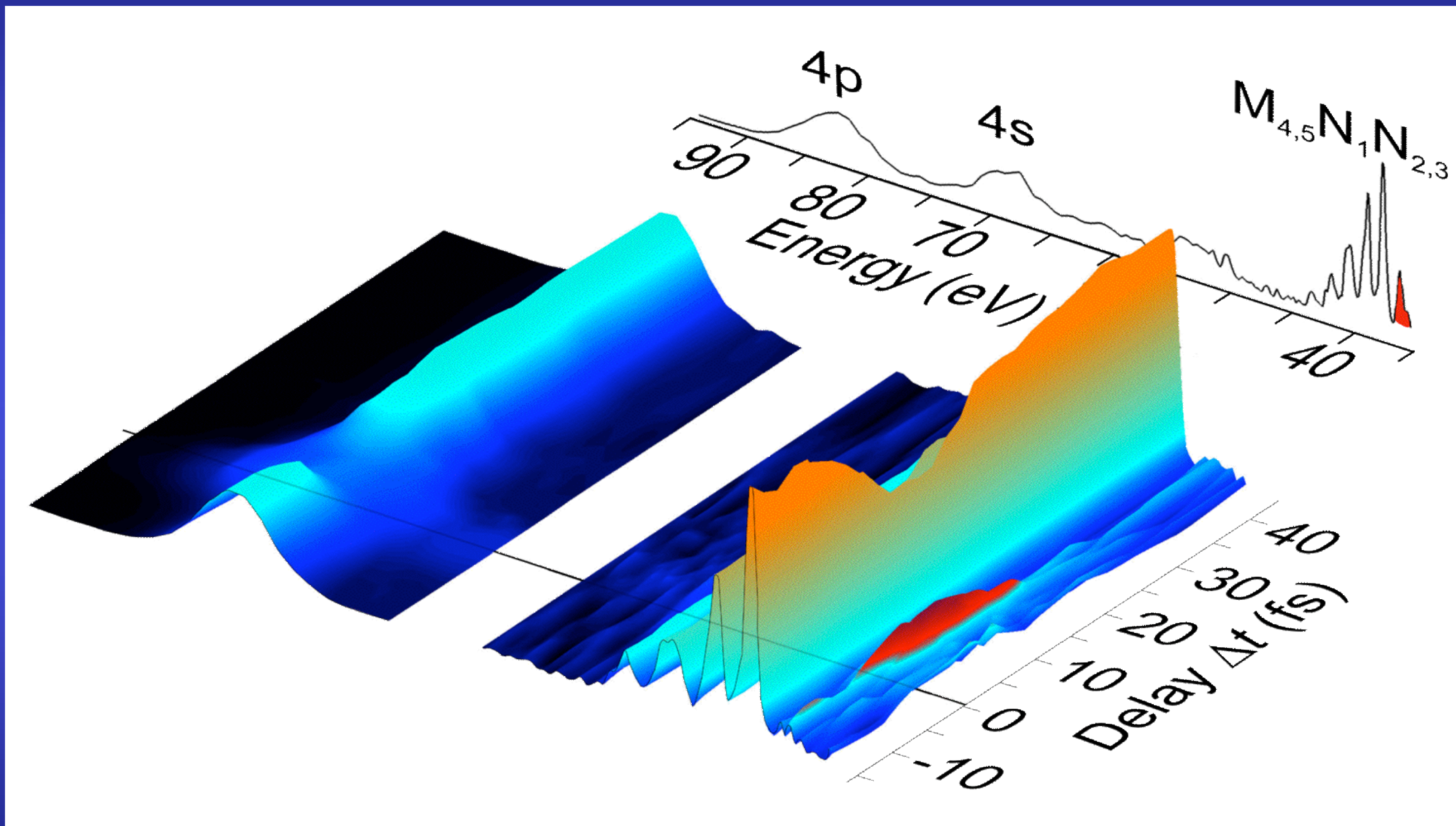
Resolution: $T_o/20 \approx 100$ as

$\tau_h > T_o/2$

Sampling by pulse envelope

Resolution: $\tau_L/5 \approx 1$ fs

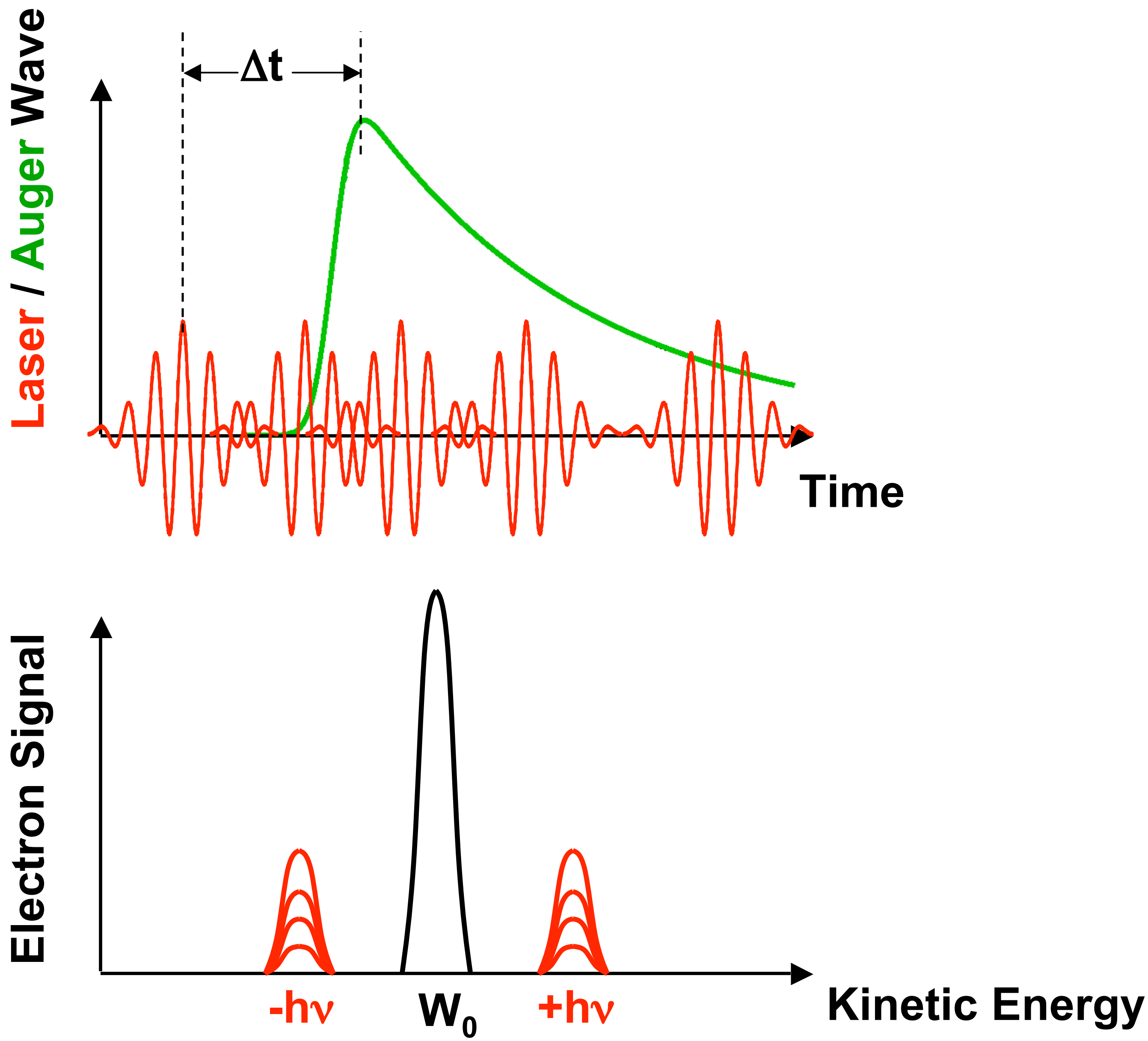
Snapshots of Electron Emission from Kr Following Core-Hole Excitation by a Sub-fs X-Ray Pulse



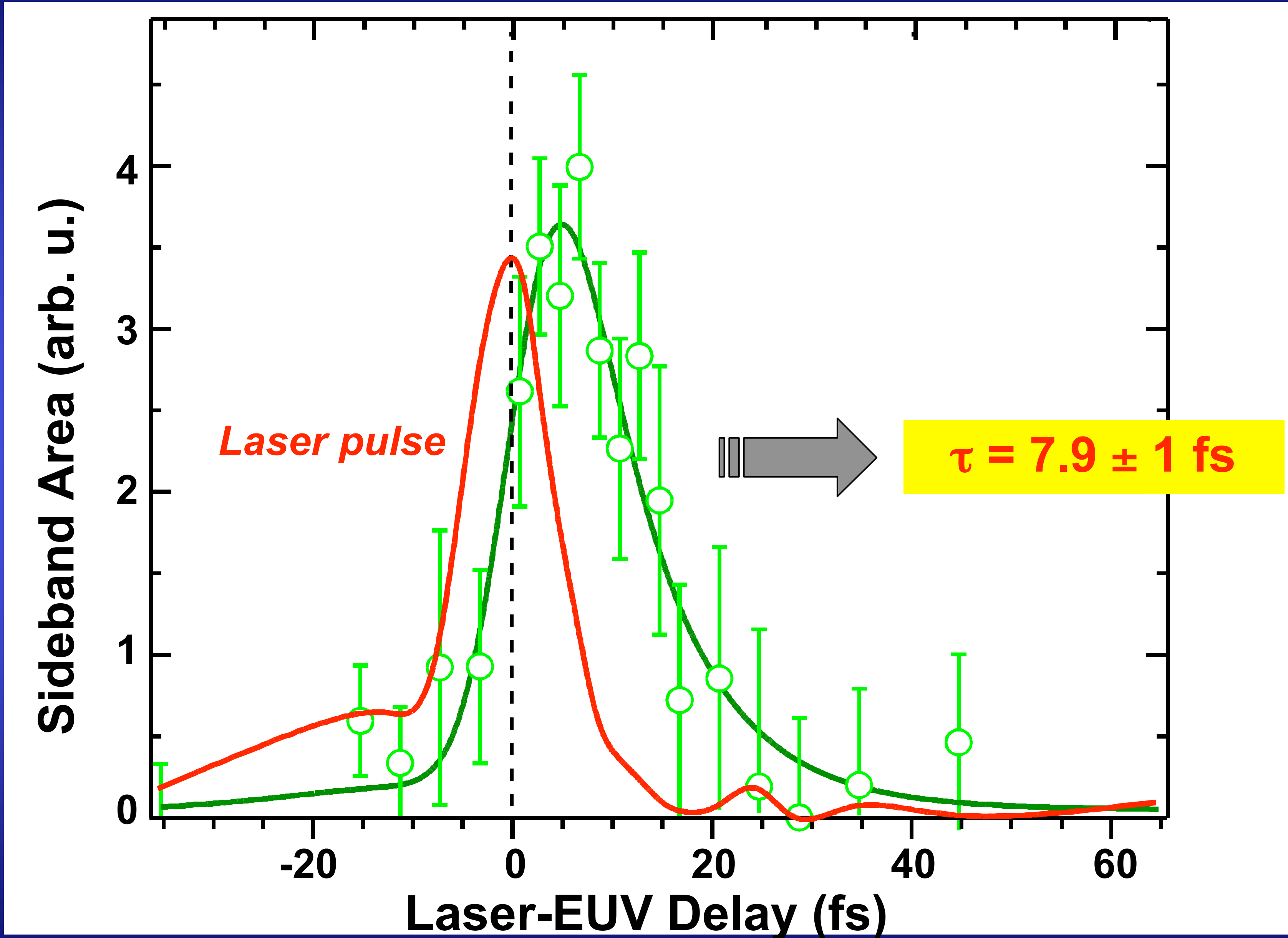
➡ **Tracing core-hole decay directly in time**

M. Drescher *et al.*, Nature **419**, 803 (2002)

Sidebands map electron wave packet

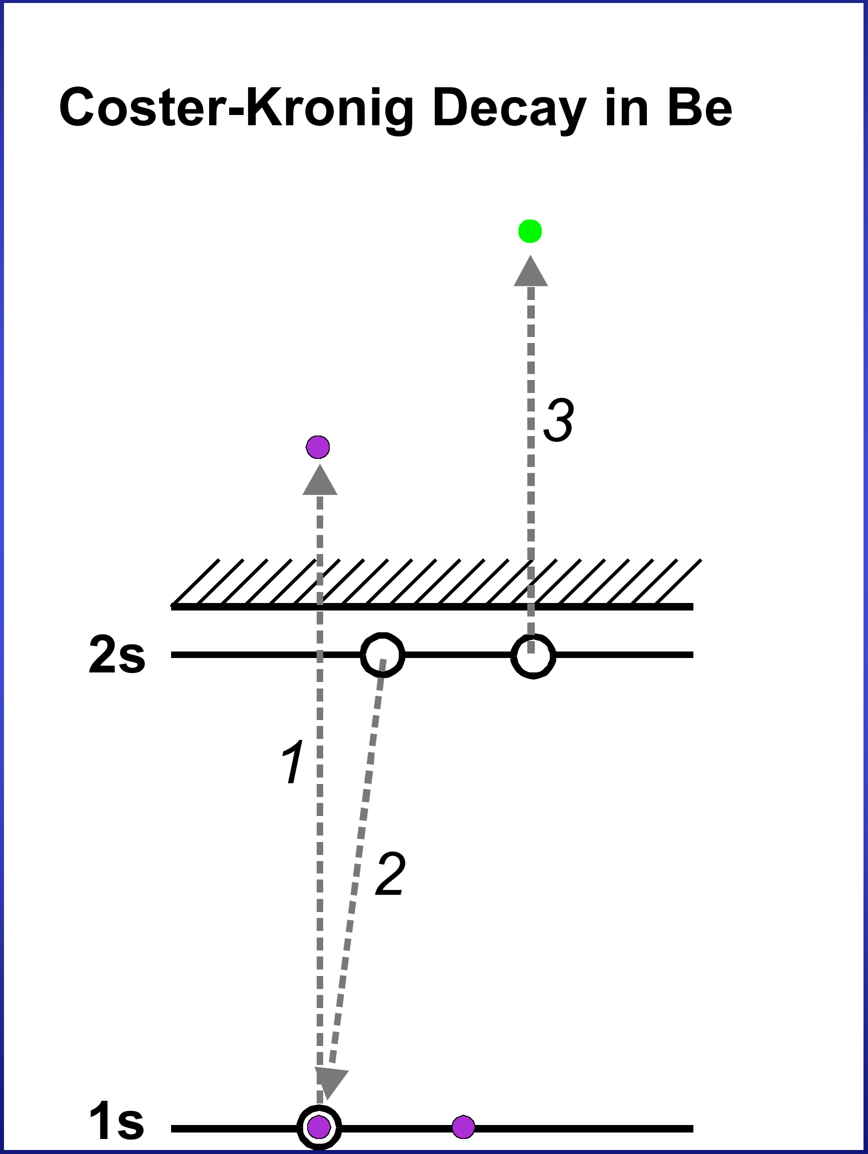


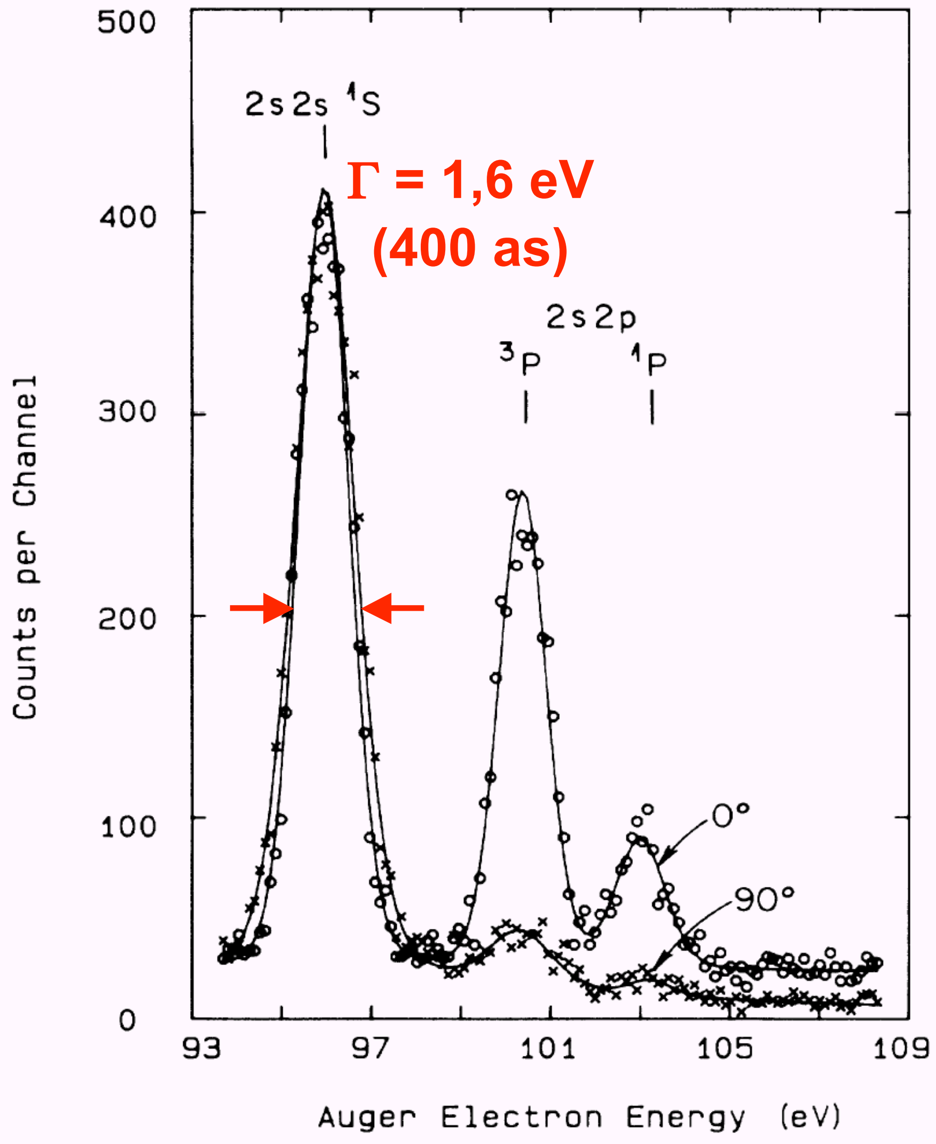
Time-dependent sideband-area



Drescher *et al.*, Nature **419**, 803 (2002)

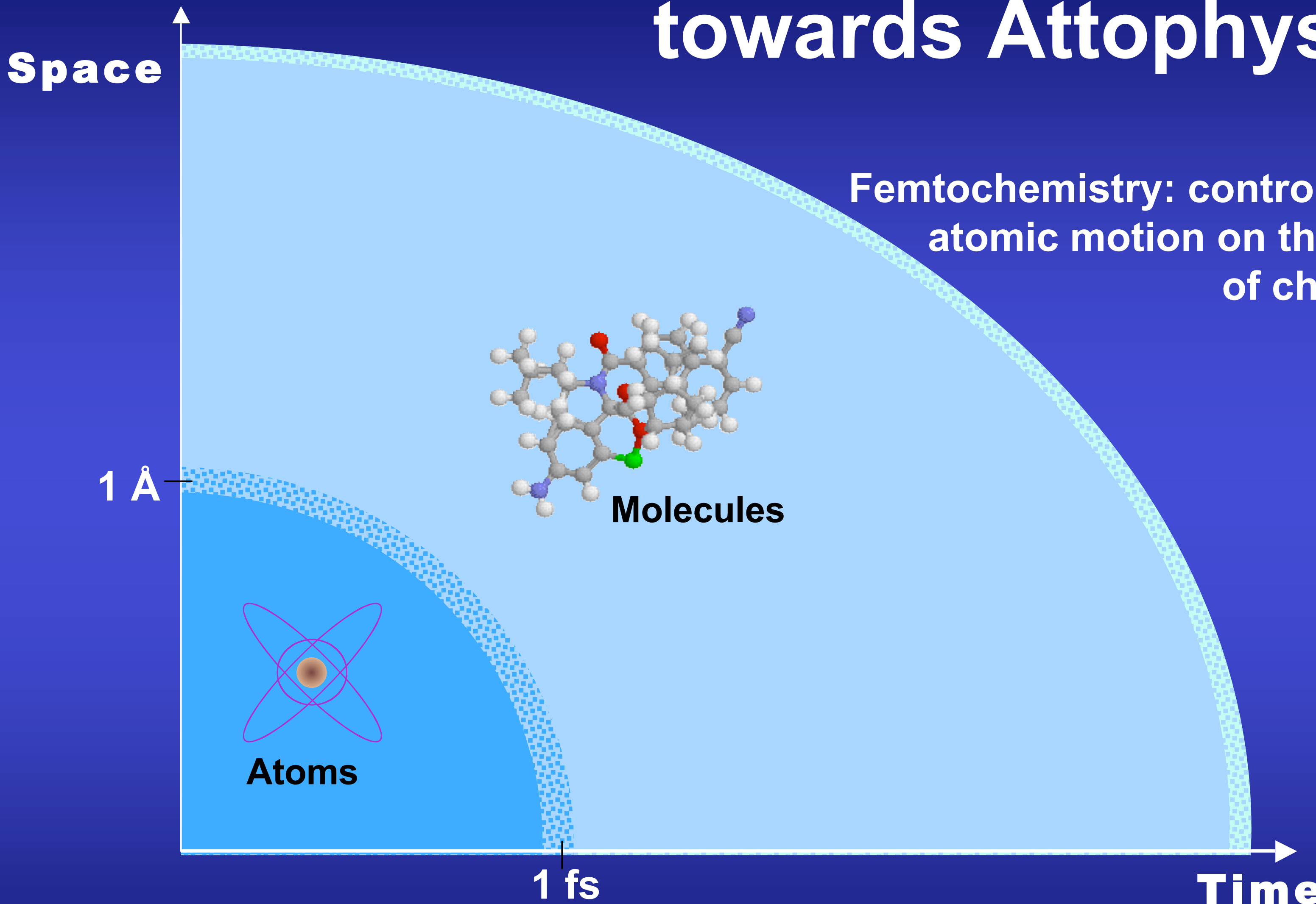
Future prospect: even faster processes





M.O. Krause et al., PRL 59, 2736 (1987)

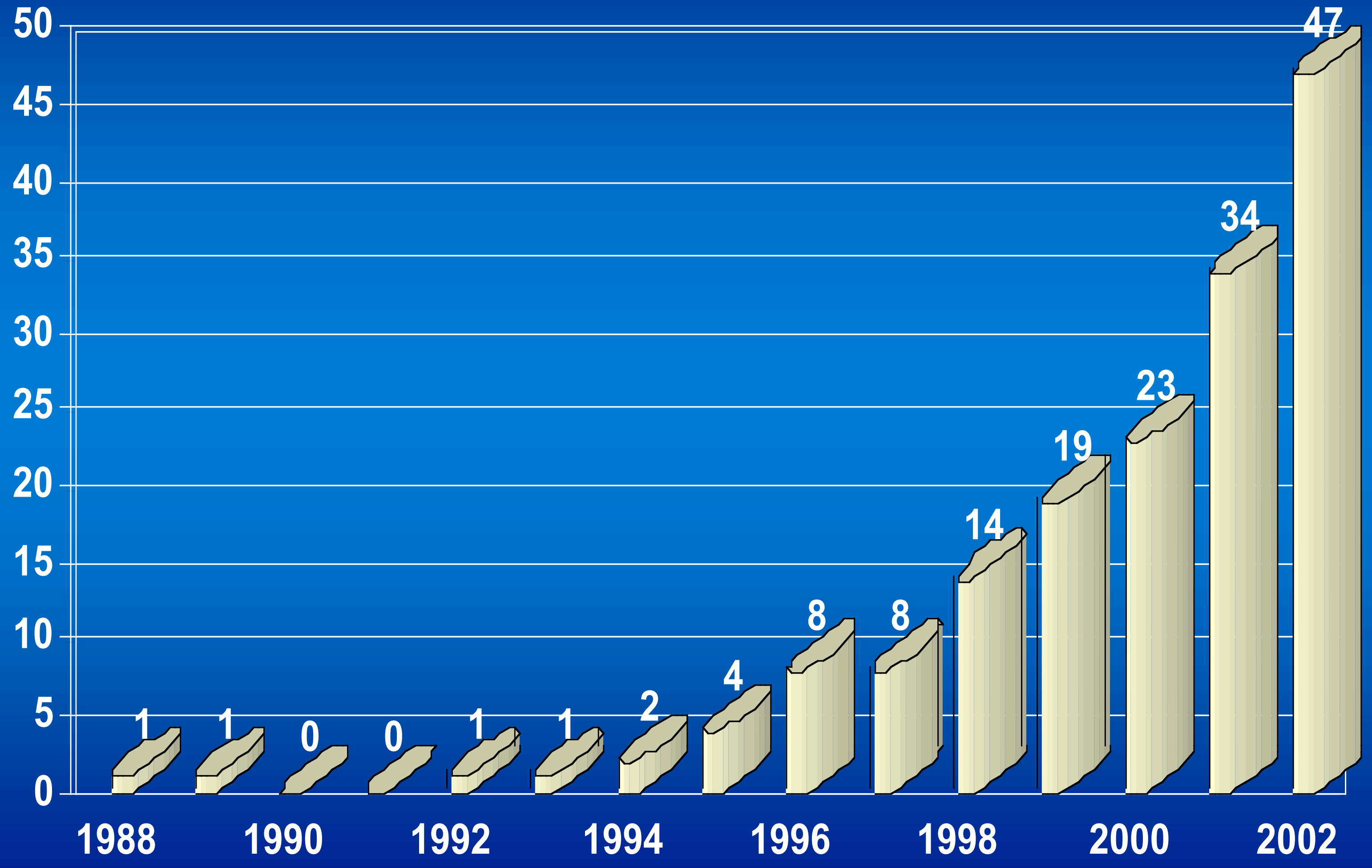
From Femtochemistry towards Attophysics



Femtochemistry: controlling & tracing atomic motion on the length scale of chemical bonds

Attophysics: controlling & tracing electronic motion on a sub-atomic scale

Number of published "attosecond" papers



A brothel, for women only

Agence France-Presse
LEIBSTADT, SWITZERLAND

A FORMER MALE stripper has decided to cast off centuries of tradition and open the country's first women-only brothel, according to a Swiss newspaper report published yesterday.

Swiss newspaper Blick said the 31-year-old man, who was only named as Calvin, decided to open "Angels" because he realised there was a real demand for the services he offers.

Calvin has hired six young "good-time boys" to entertain his female customers with massages, striptease shows and more should they so wish.

The brothel owner's doctor ordered him to hang up his g-string after 10 years on the striptease circuit because of severe back problems.

"Trojan horse" of the Americans, the report said, adding that it was widely considered that Britain could not reconcile itself to a future in the European community.

But British character, culture and traditions were not without virtue, the report concluded.

A refusal to become flustered, the Beatles, James Bond and "crafty" diplomacy were all much admired.

World's fastest laser appears to 'freeze chemistry in time'

Agence France-Presse
PARIS

AUSTRIAN SCIENTISTS say they have developed the world's fastest laser, a device that opens the door to studying the movement of electrons and other atomic processes.

The laser works at 650 attoseconds (650 million trillionths of a second), a team led by Ferenc Krausz of Vienna's Technical University said in the British science weekly *Nature* published yesterday.

It marks a giant step forward in ultra-lasers, a field in which scientists strive to find exactly how a chemical or physical reaction works in order to develop new materials and drugs.

The achievement heralds "the dawn of attophysics", said Yaron Silberberg of the Weizmann Institute of Science in Israel, referring to an attosecond, the shortest unit of time for which we currently have a name.

An attosecond is to a second what a second is to 32 billion years.

"At these time scales, chemistry is essentially frozen in time," Silberberg said.

He hopes that enhancements could one day yield a laser to show how molecules gain and shed electrons themselves.

The laser first shines a pulse onto a gas of neon atoms. This splits the beam into an optical pulse and a "harmonic" pulse in the ultraviolet and X-ray ranges of the energy spectrum.

The two beams then pass through a zirconium filter, which causes the harmonic pulse to slow down but does not affect the speed of the optical pulse.

By directing the two beams at a cloud of krypton-gas atoms, the scientists were able to measure the difference in time between when the atoms were hit by the harmonic pulse and when they were hit by the optical pulse.

That enabled them to monitor the spectrum of energy released by electrons as they were expelled from the atoms.

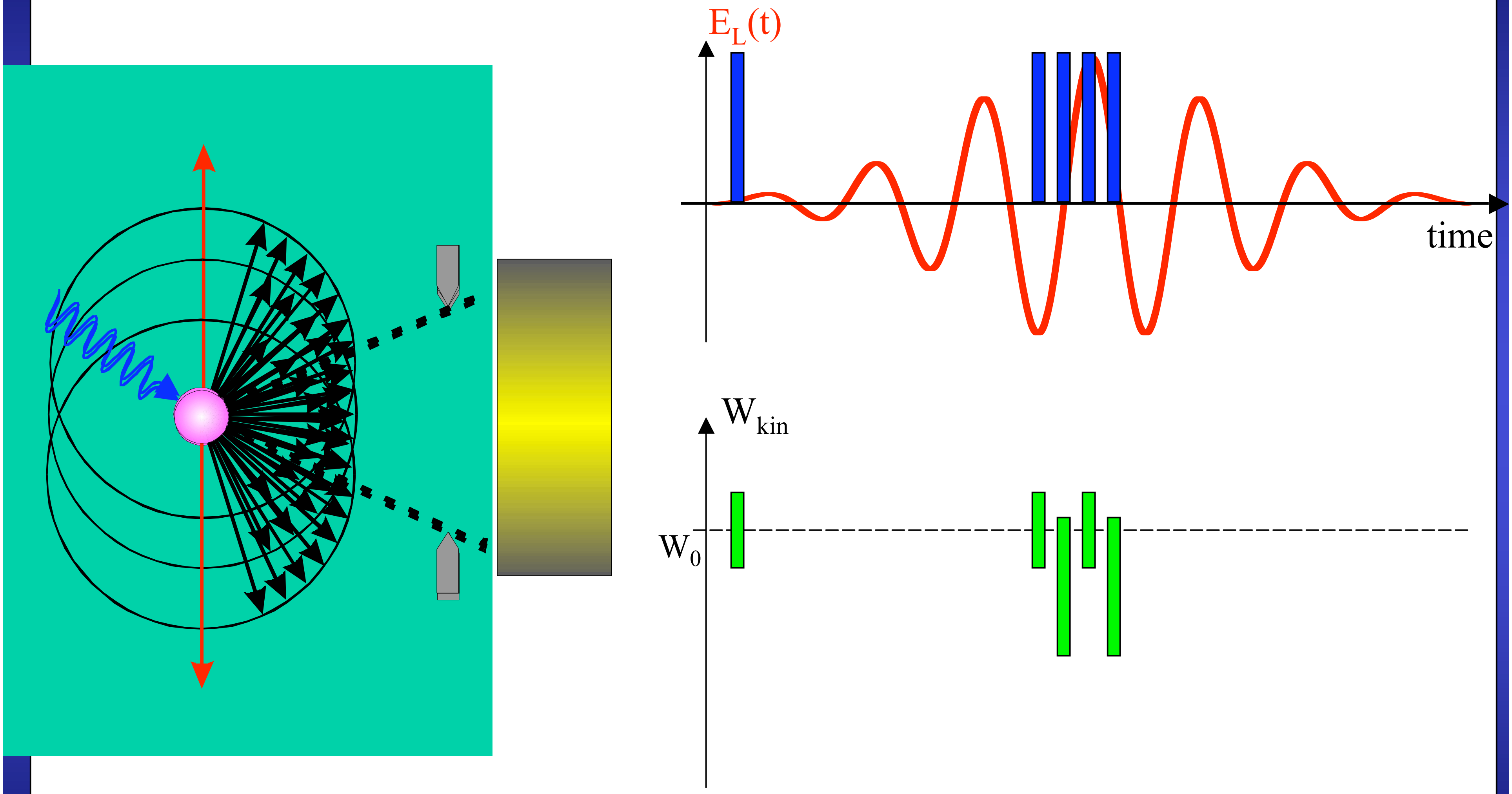
The attosecond laser builds on the femtosecond laser, a device that is fractionally slower and already used to study chemical reactions.

Femtochemistry leapt to prominence in 1999 when Ahmed Zewail of Egypt won the Nobel Prize for Chemistry for his work in the field.

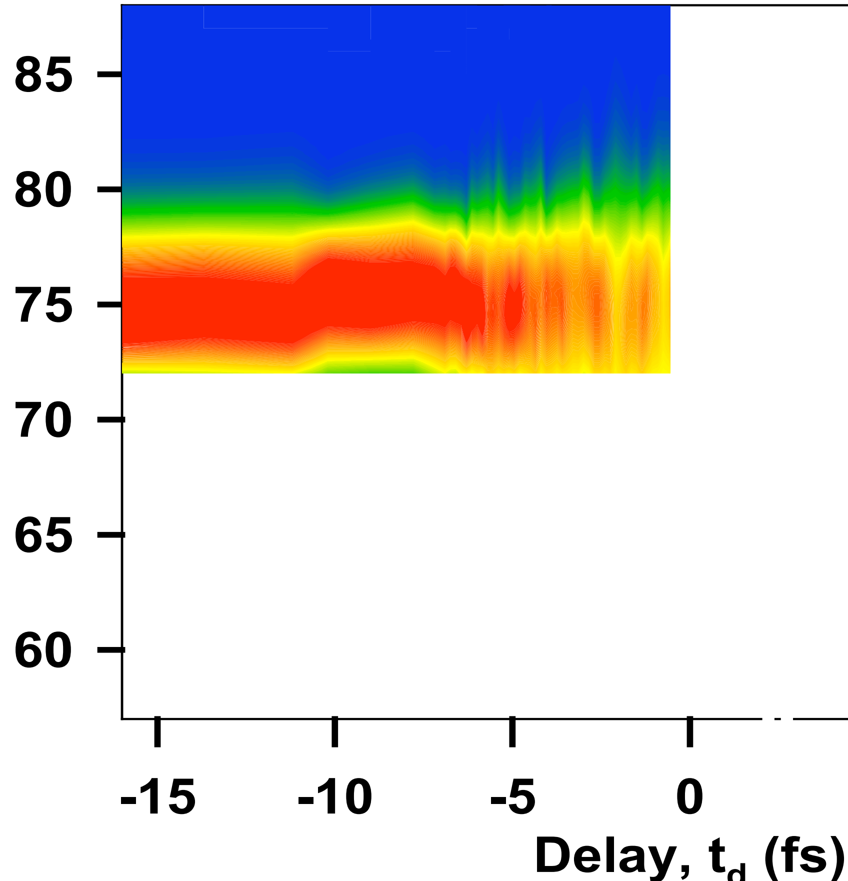
A femtosecond is 10 to the power of minus 15, while an attosecond is 10 to the power of minus 18.

Grants:
FWF (Austrian Science fund) F016 (ADLIS),Z63 (Wittgenstein), P15382 (phase-control);
RK: fellowship by Austrian Academy of Sciences

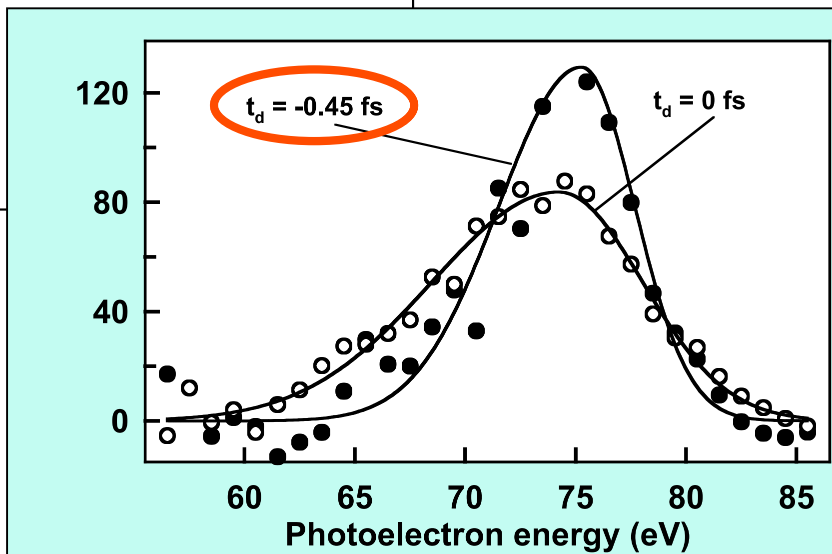
Laser assisted X-ray photoionization



Subfemtosecond XUV Pulse Resolves Light Field Oscillations

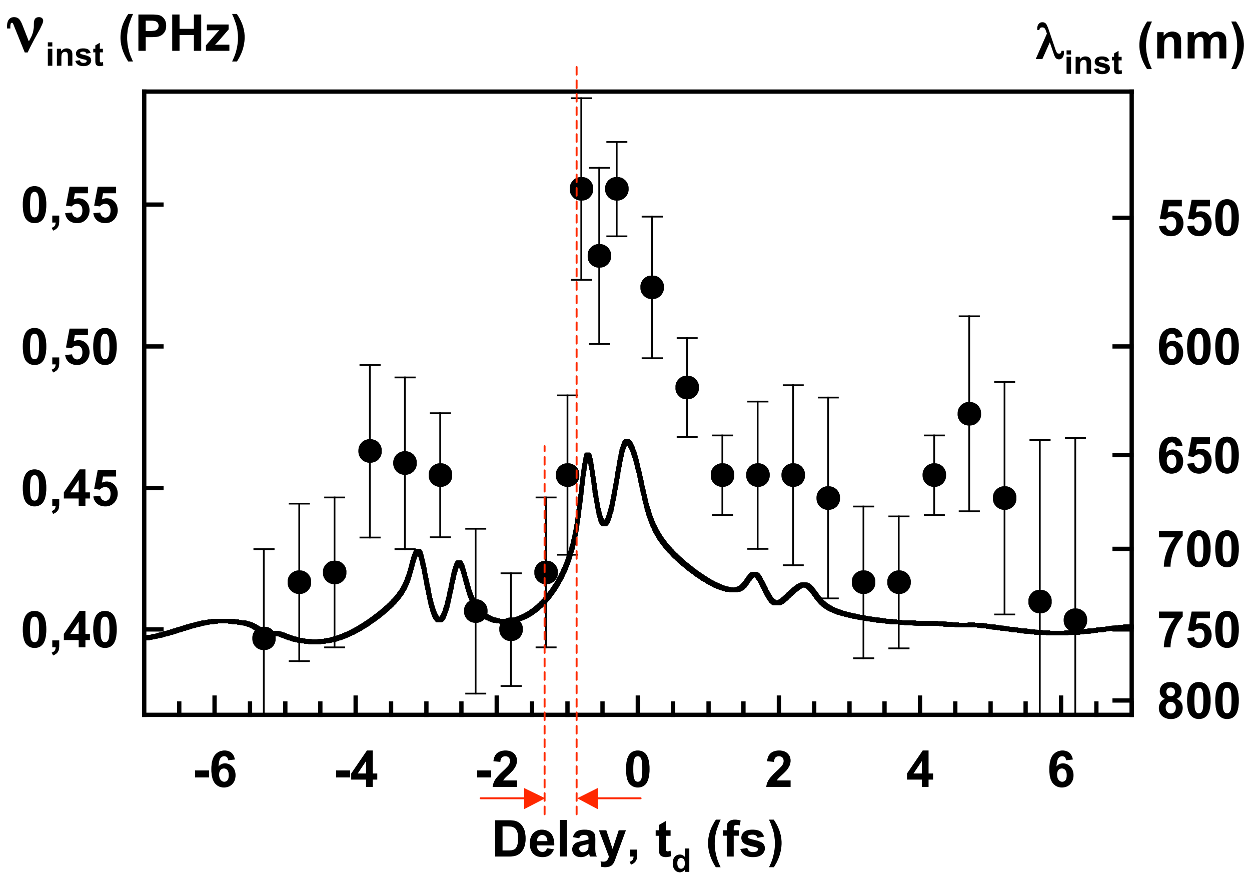


Hentschel *et al.*,
Nature **414**, 509 (2001).



- τ_x = 650as ± 150as
- Attosecond synchronism of XUV pulse to light wave

Attosecond Metrology: Direct Measurement of Dynamic Frequency Shifts of Light



$t_{rise} < 1$ fs isolated sampling x-ray pulse

Measurement of the sub-fs temporal evolution of the instantaneous energy (chirp) and intensity of electrons emitted from atoms or molecules

delay!!