Attosecond Electron Dynamics on Surfaces and Layered Systems

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ICPEAC
Cairns, Australia
26.07.2017

Scope of ICPEAC

The ICPEAC conference attracts several hundred scientists from all over the globe who work actively in the fields **of collisions involving photons, electrons, ions, atoms, molecules, clusters, surfaces, and exotic particles**. Recent expansions of the conference scope include **collision with cold targets and attosecond science**.

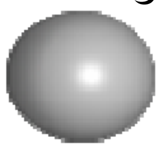
electrons – atoms (ions): High-order Harmonic Generation

photons – surfaces: photoelectroc effect

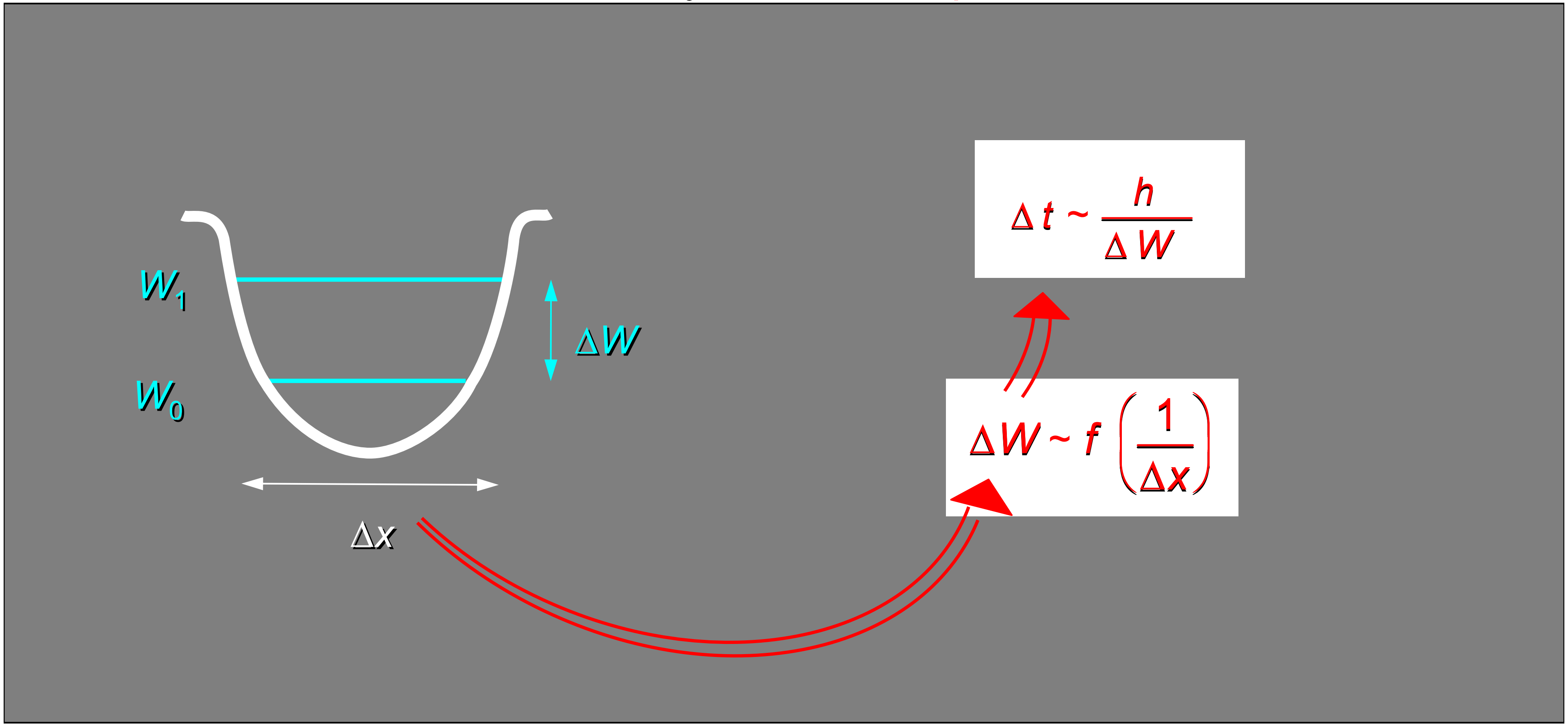
Attosecond science

Observation and steering of electronic processes

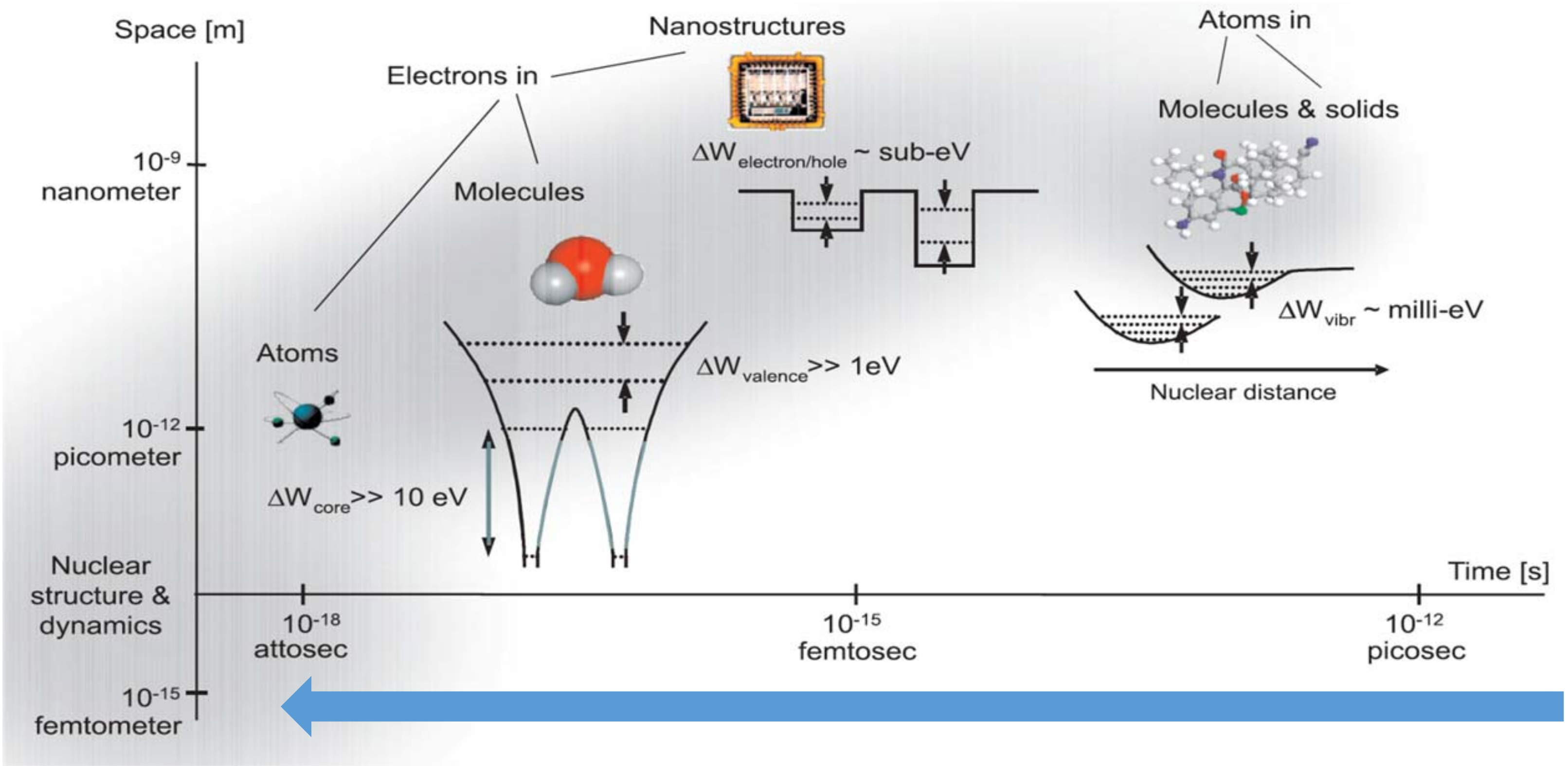
e^-



the time & length scales of microscopic motion
are connected by the laws of quantum mechanics



Timescales of electronic dynamics in ever smaller systems



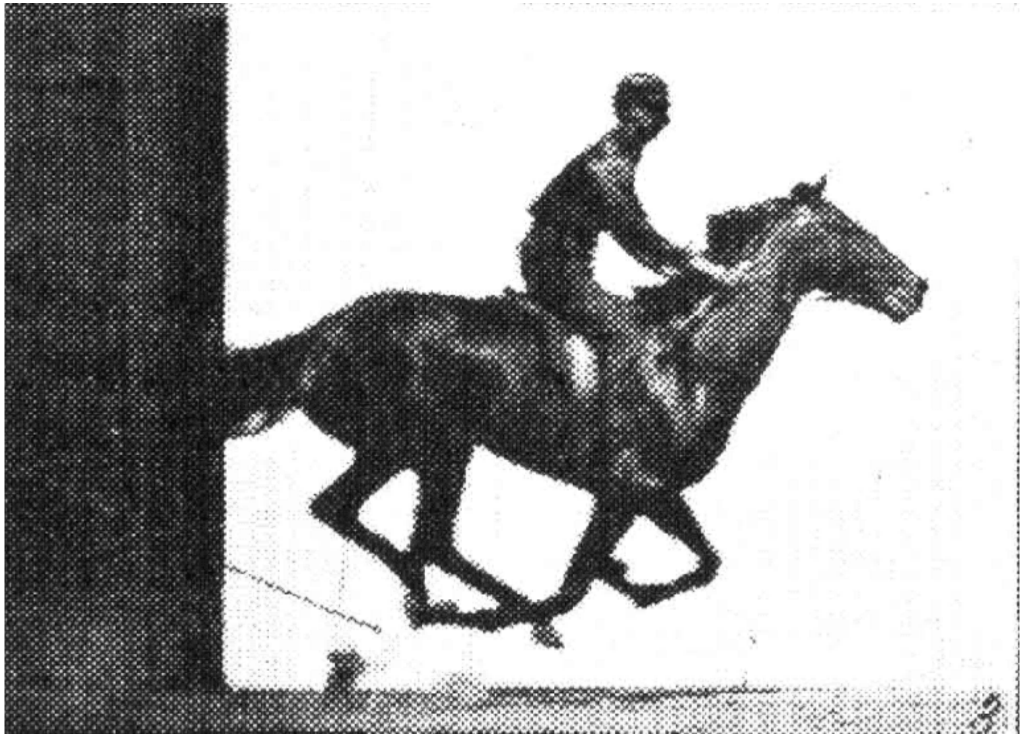
Limit in resolution: exposure time

microsecond - photography

bullet in a glass block

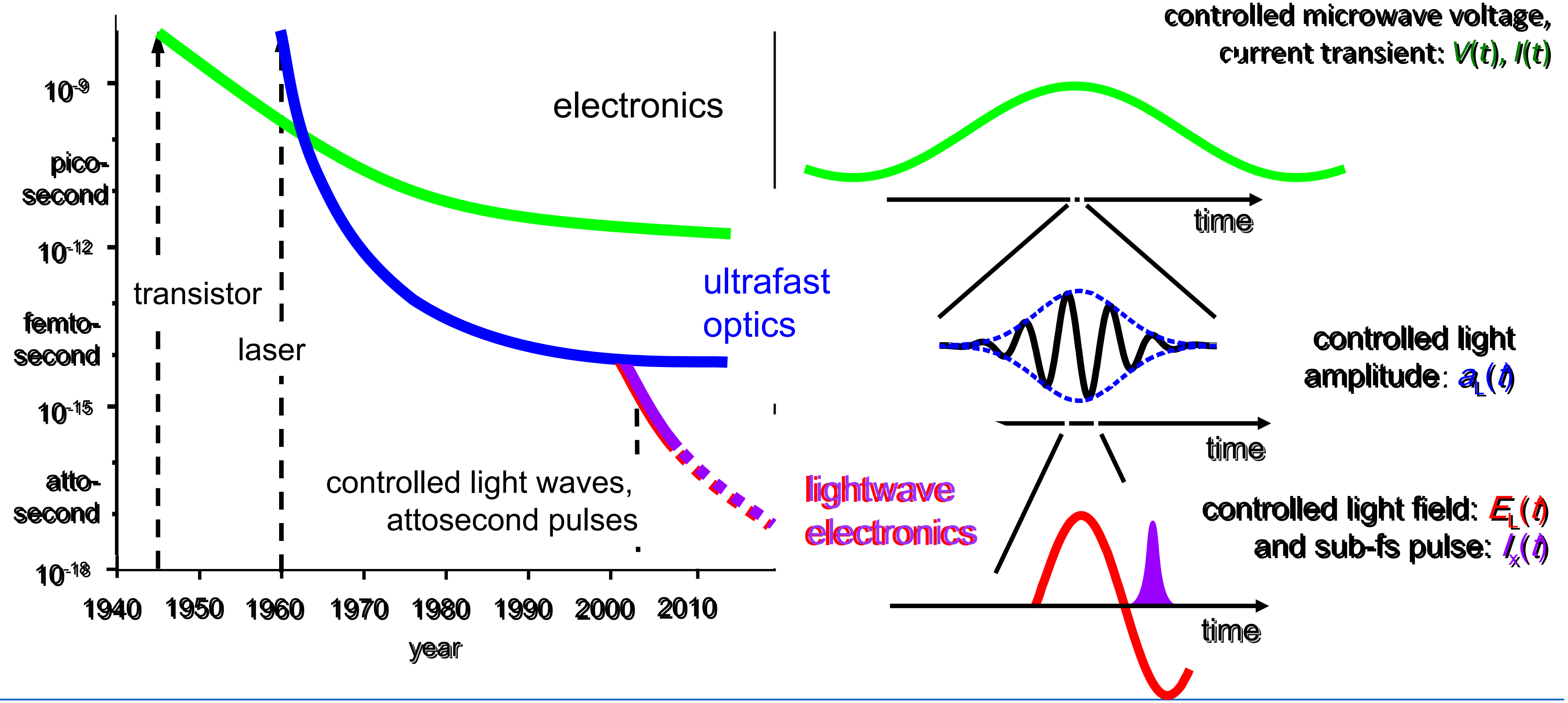
1878: E. Muybridge, Stanford

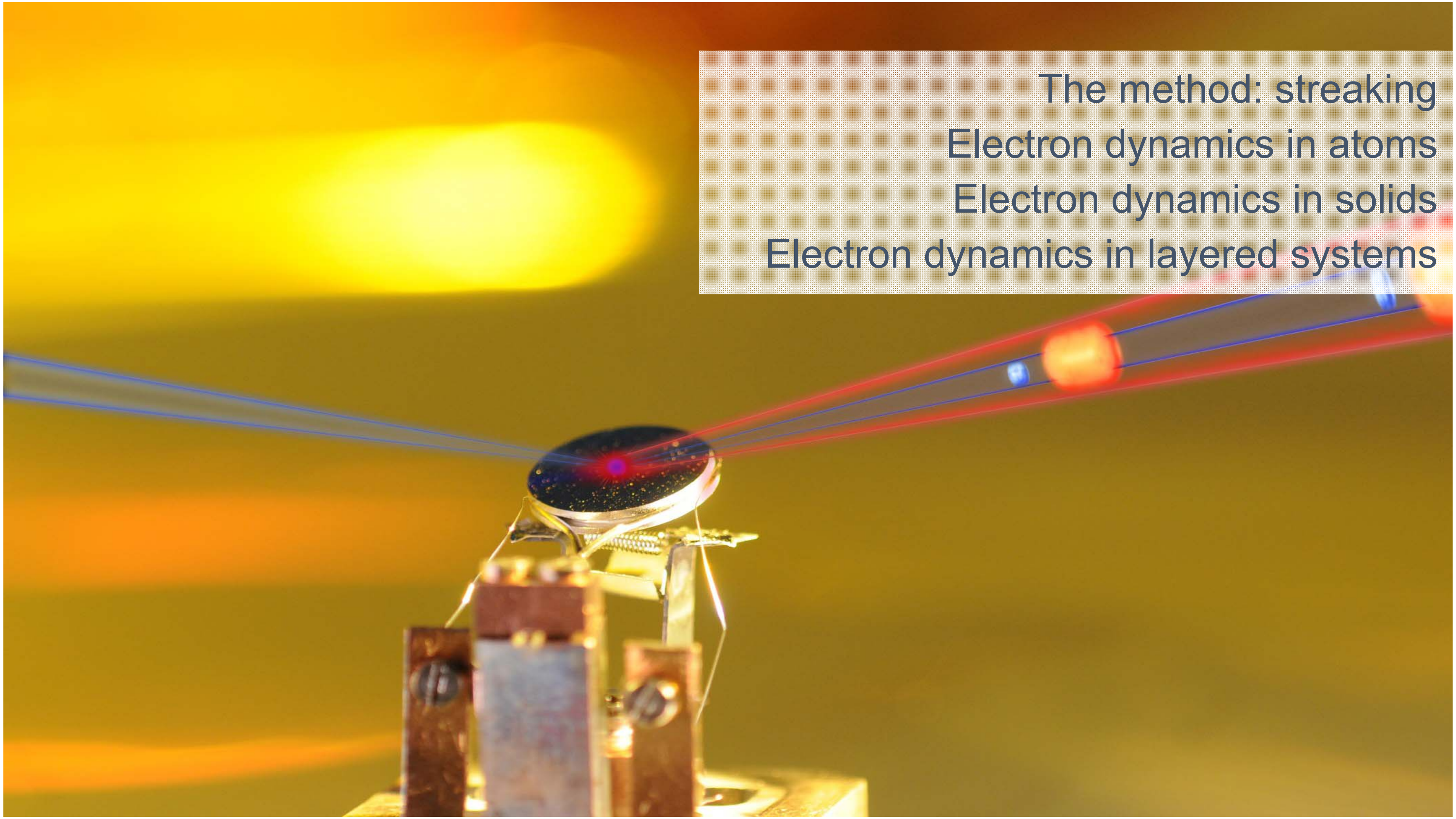
Reconstruction of the process (“slow-motion”)
by stringing together snapshots at different phases of the process

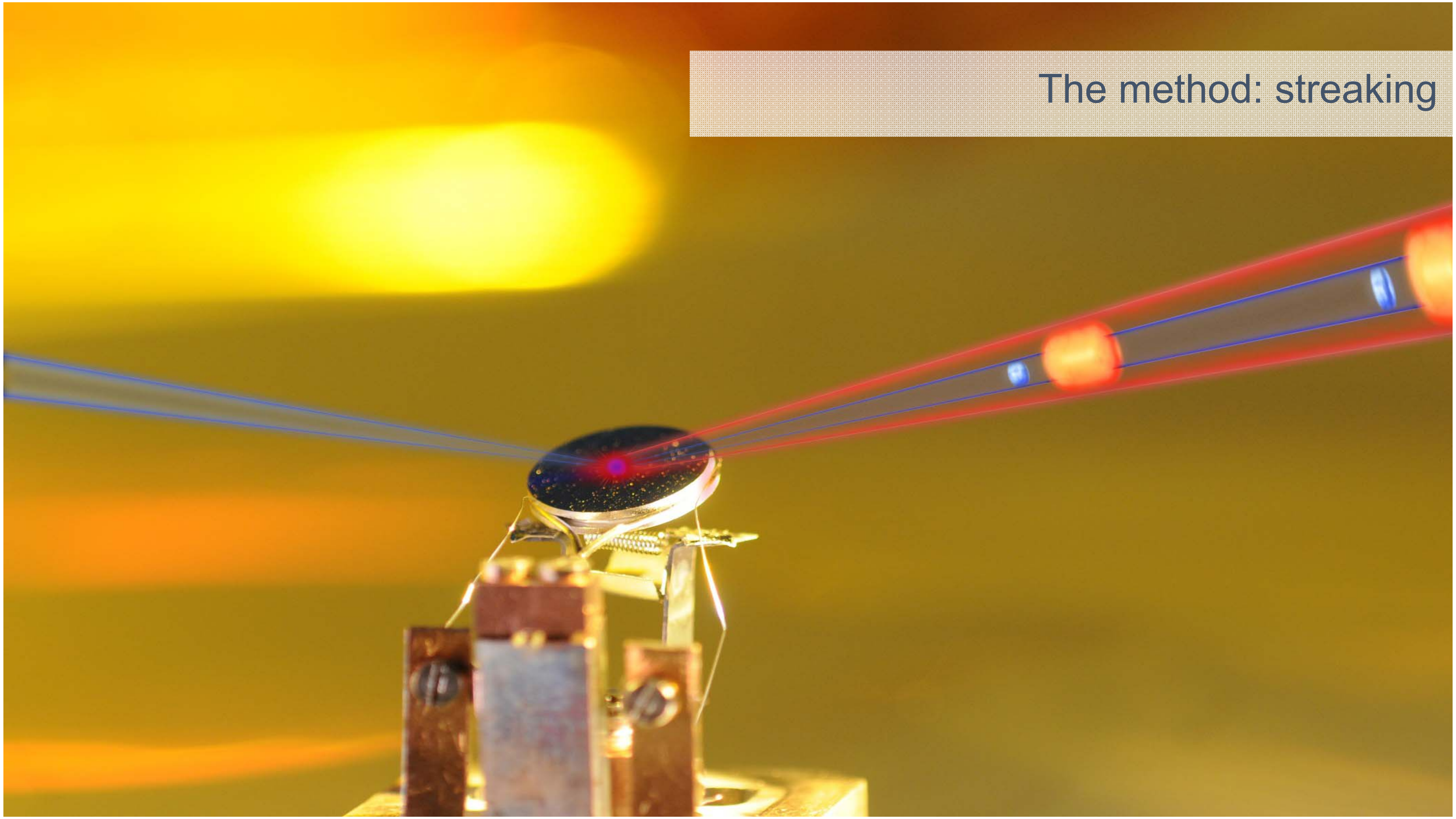


E. Muybridge, *Animals in Motion*, ed. by L. S. Brown (Dover Publ. Co., New York 1957)

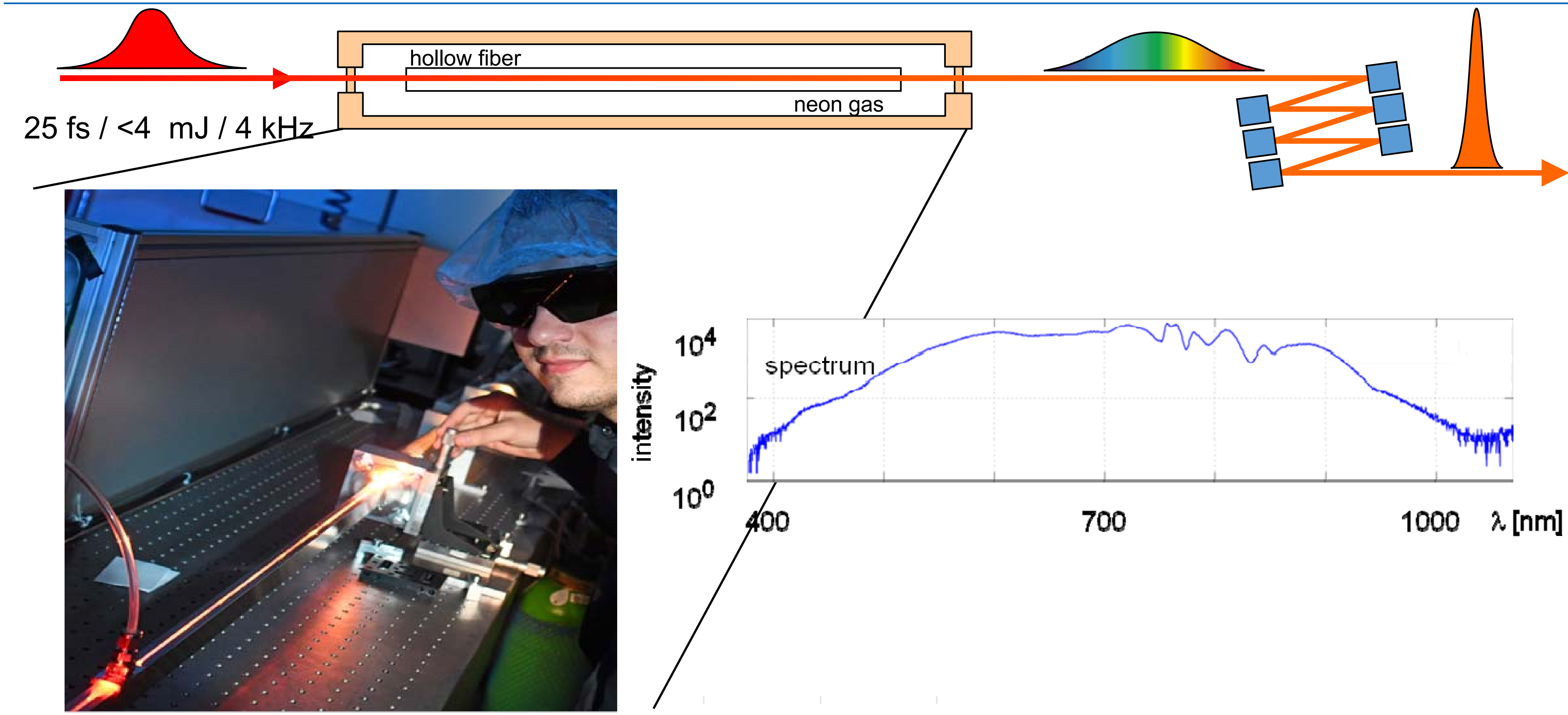
from superfast electronics to lightwave driven electronics




The method: streaking
Electron dynamics in atoms
Electron dynamics in solids
Electron dynamics in layered systems

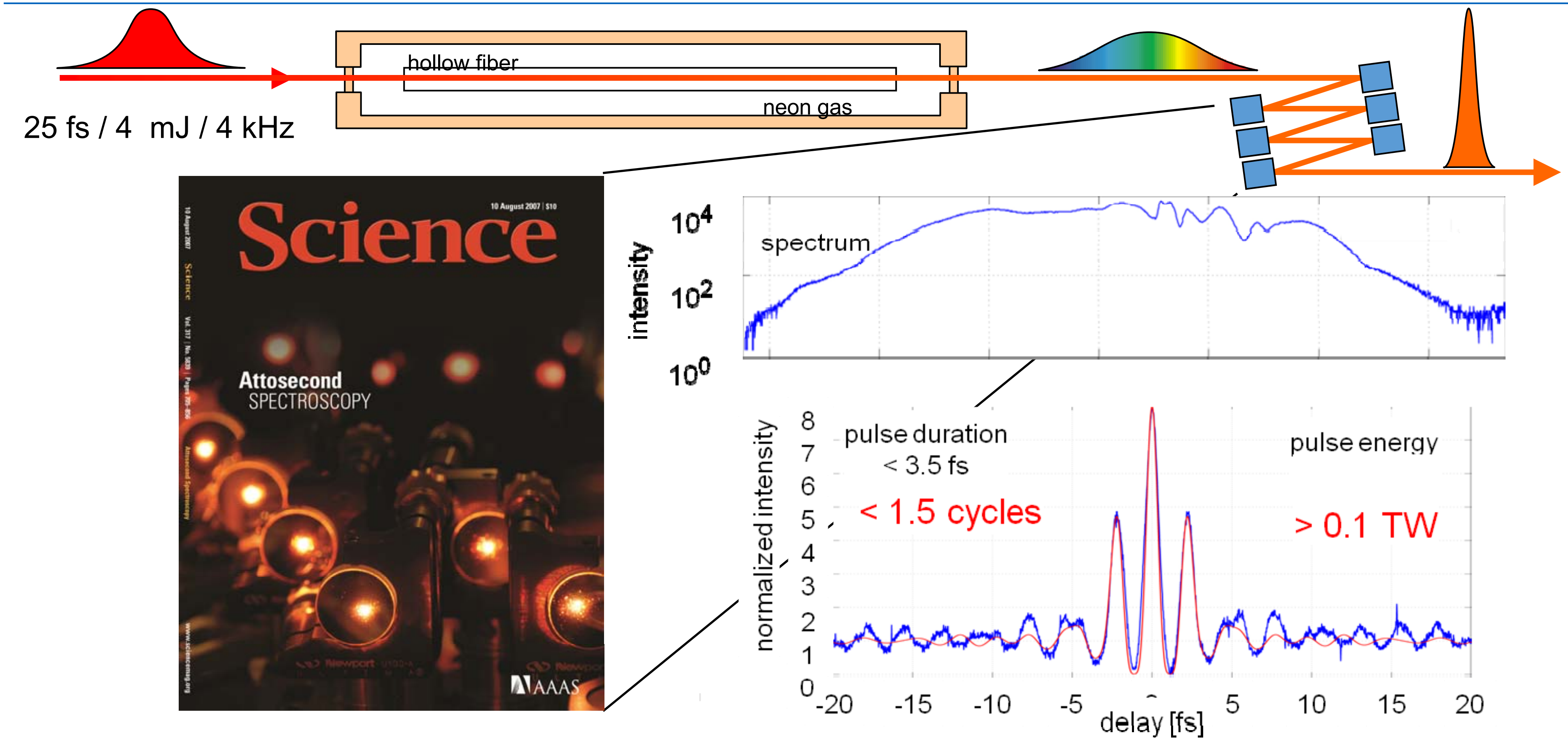

The method: streaking

few-cycle pulses

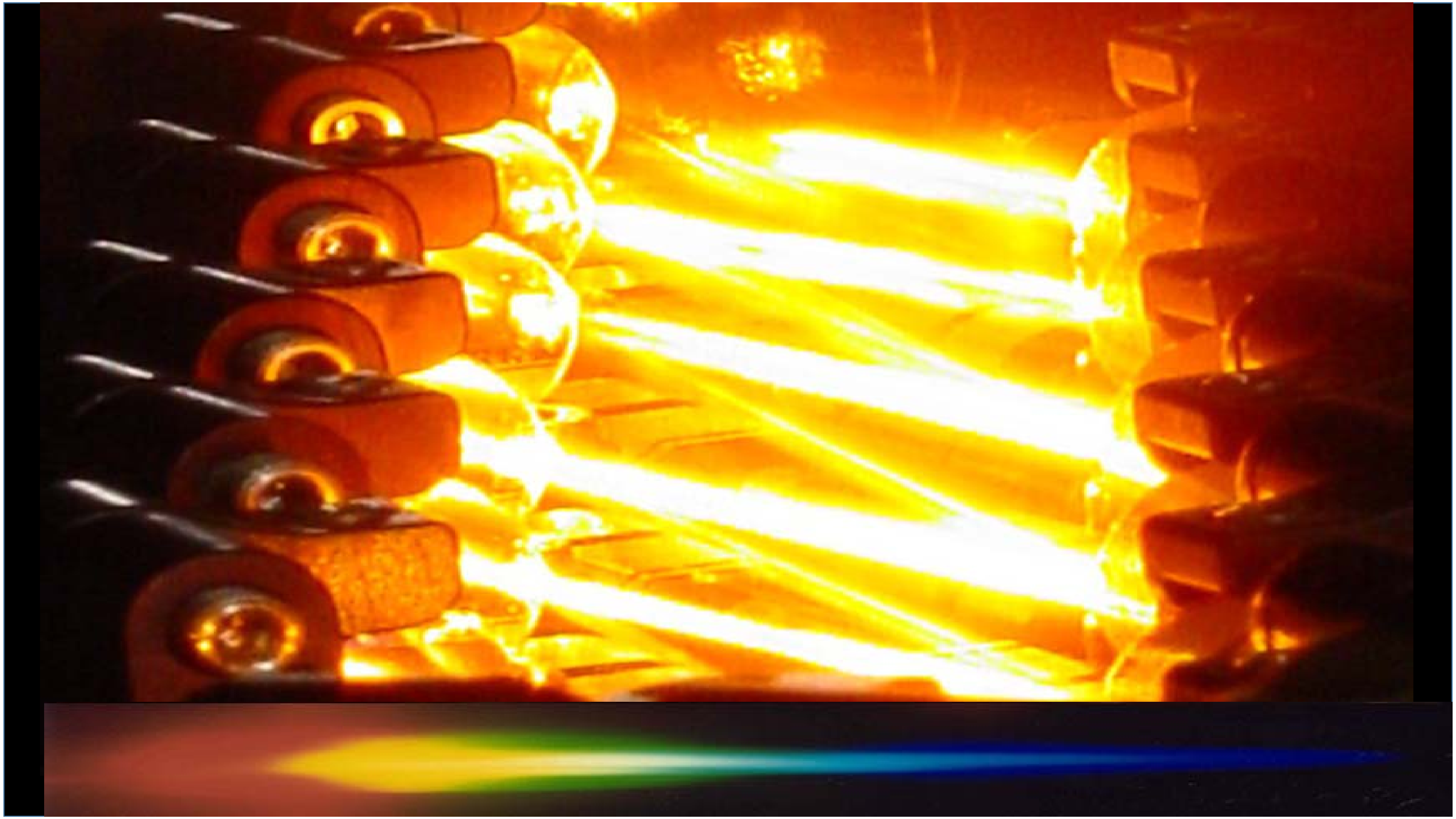


A. L. Cavalieri *et al*, *New J. Phys.* 9, 242 (2007); E. Goulielmakis *et al*, *Science* 317, 769 (2007)

few-cycle pulses

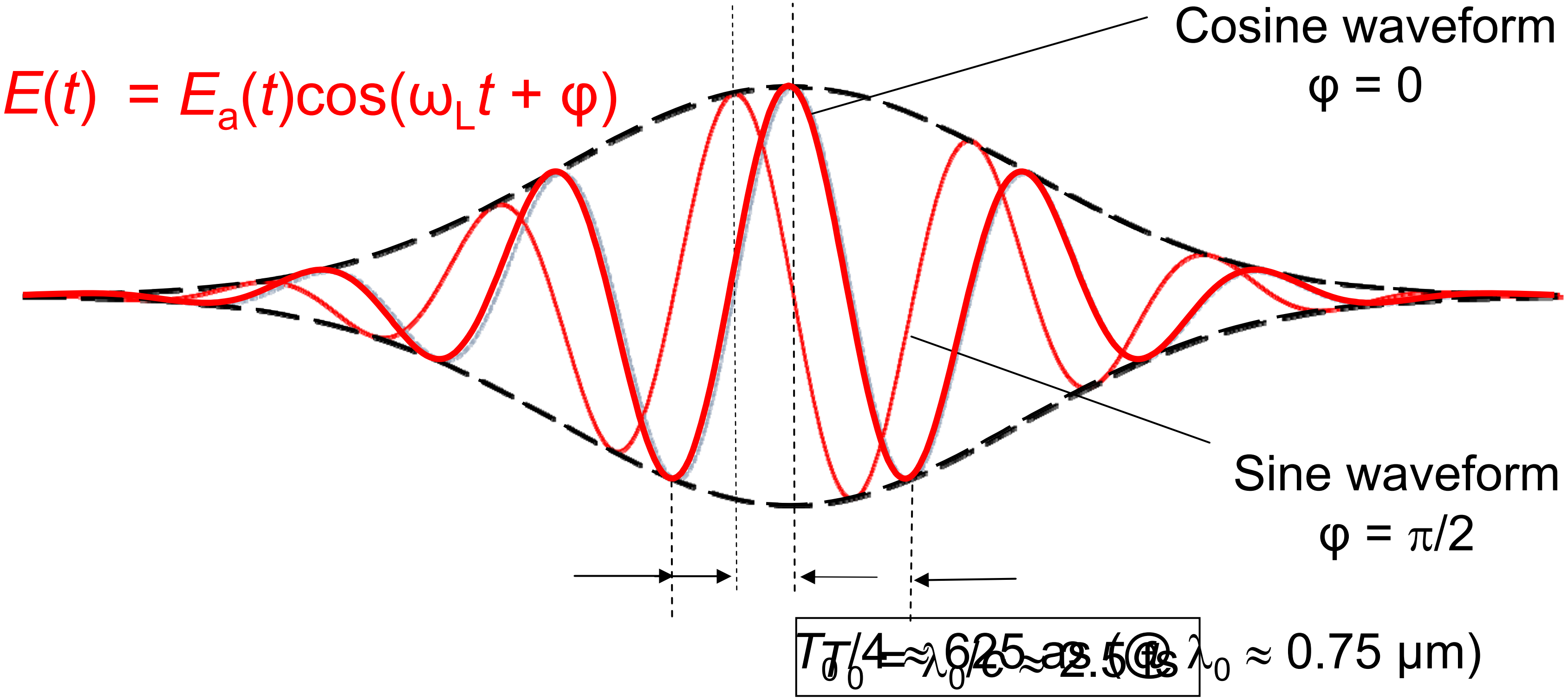


A. L. Cavalieri *et al*, *New J. Phys.* 9, 242 (2007); E. Goulielmakis *et al*, *Science* 317, 769 (2007)



the carrier-envelope phase CEP

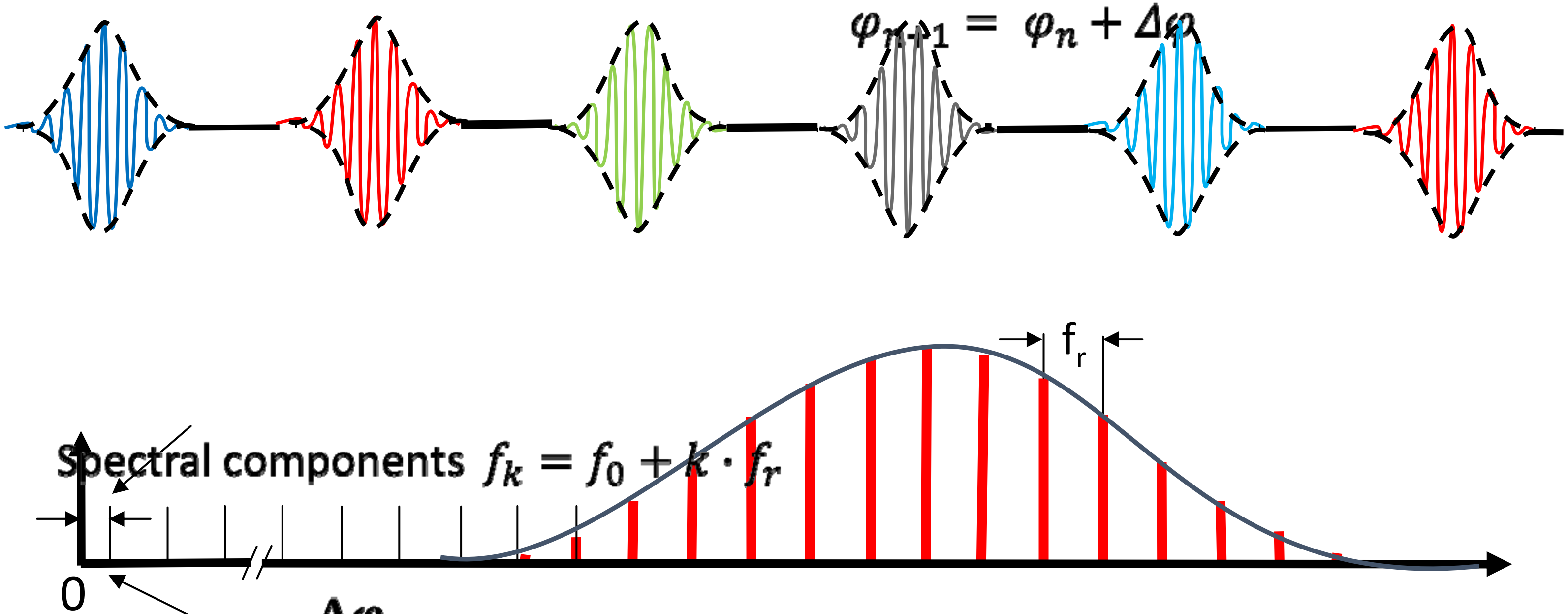
$$E(t) = E_a(t)\cos(\omega_L t + \varphi)$$



pulse train from a mode-locked laser

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

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

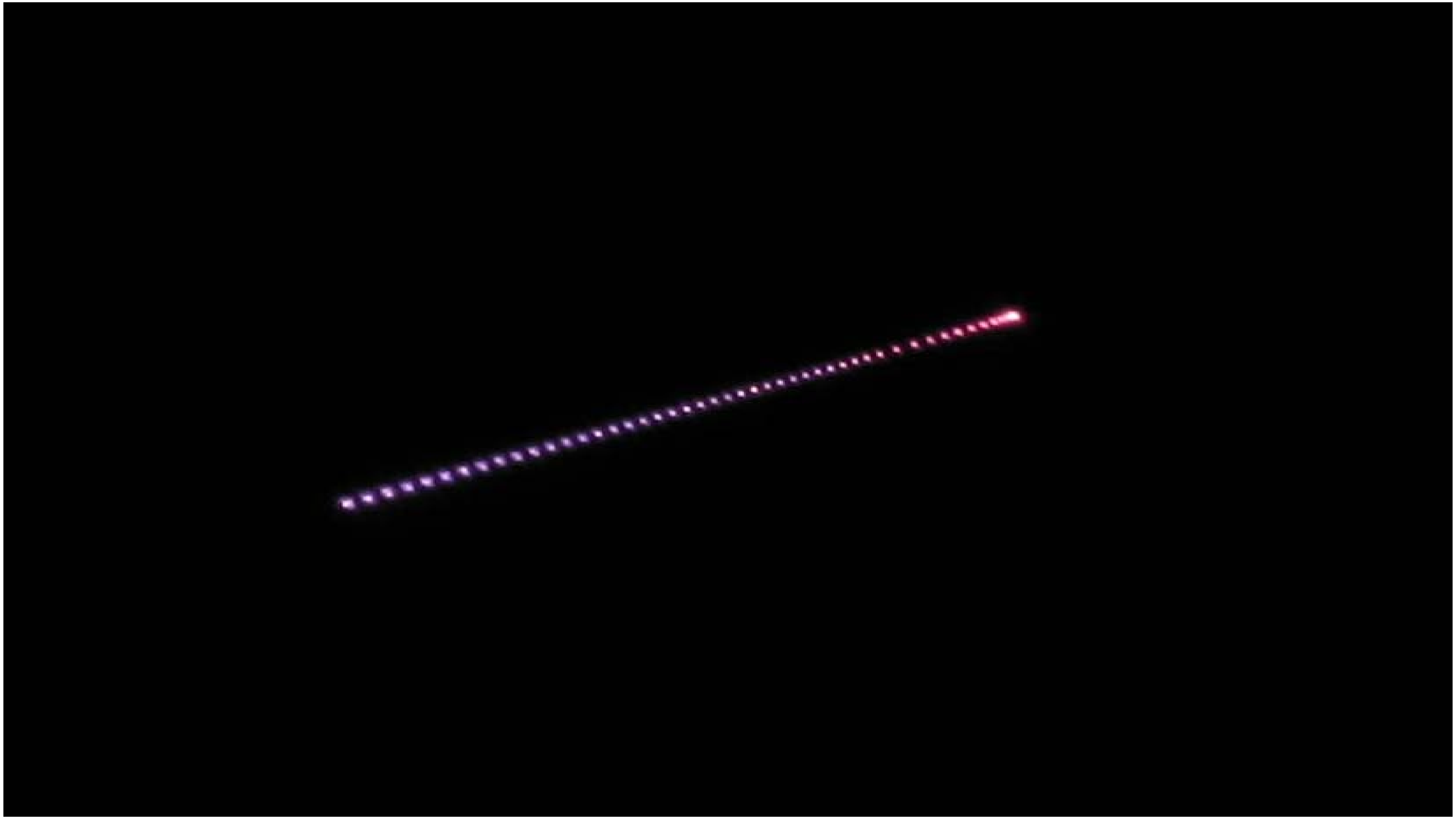


Spectral components $f_k = f_0 + k \cdot f_r$

$$f_0 = \frac{\Delta\varphi}{2\pi} f_r$$

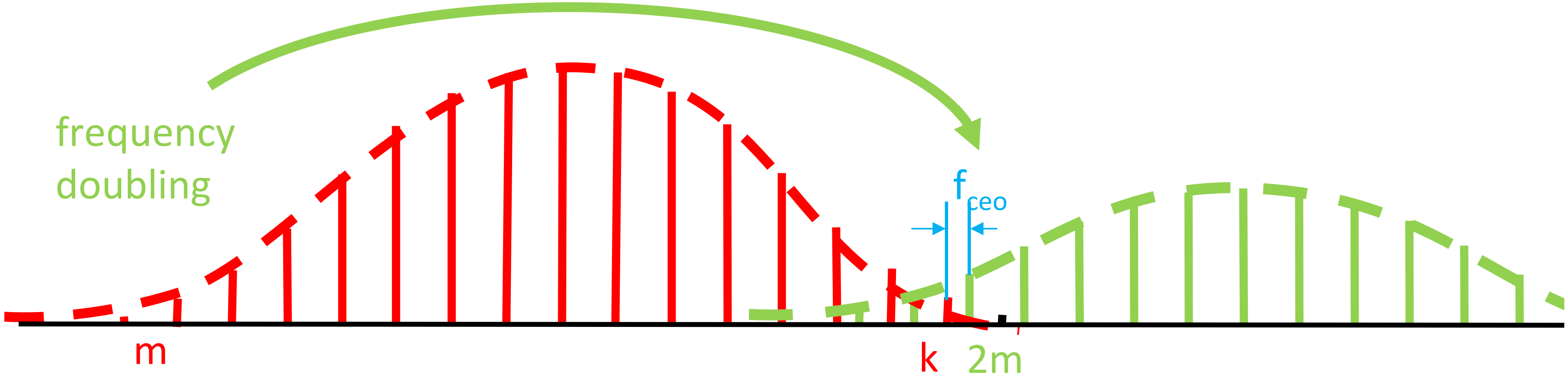
Carrier-envelope phase slip signal

$F_0 \equiv f_{ceo}$: carrier-envelope-offset frequency



frequency-domain control of the CEP

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



Beating of the fundamental $f_k = f_0 + k \cdot f_r$

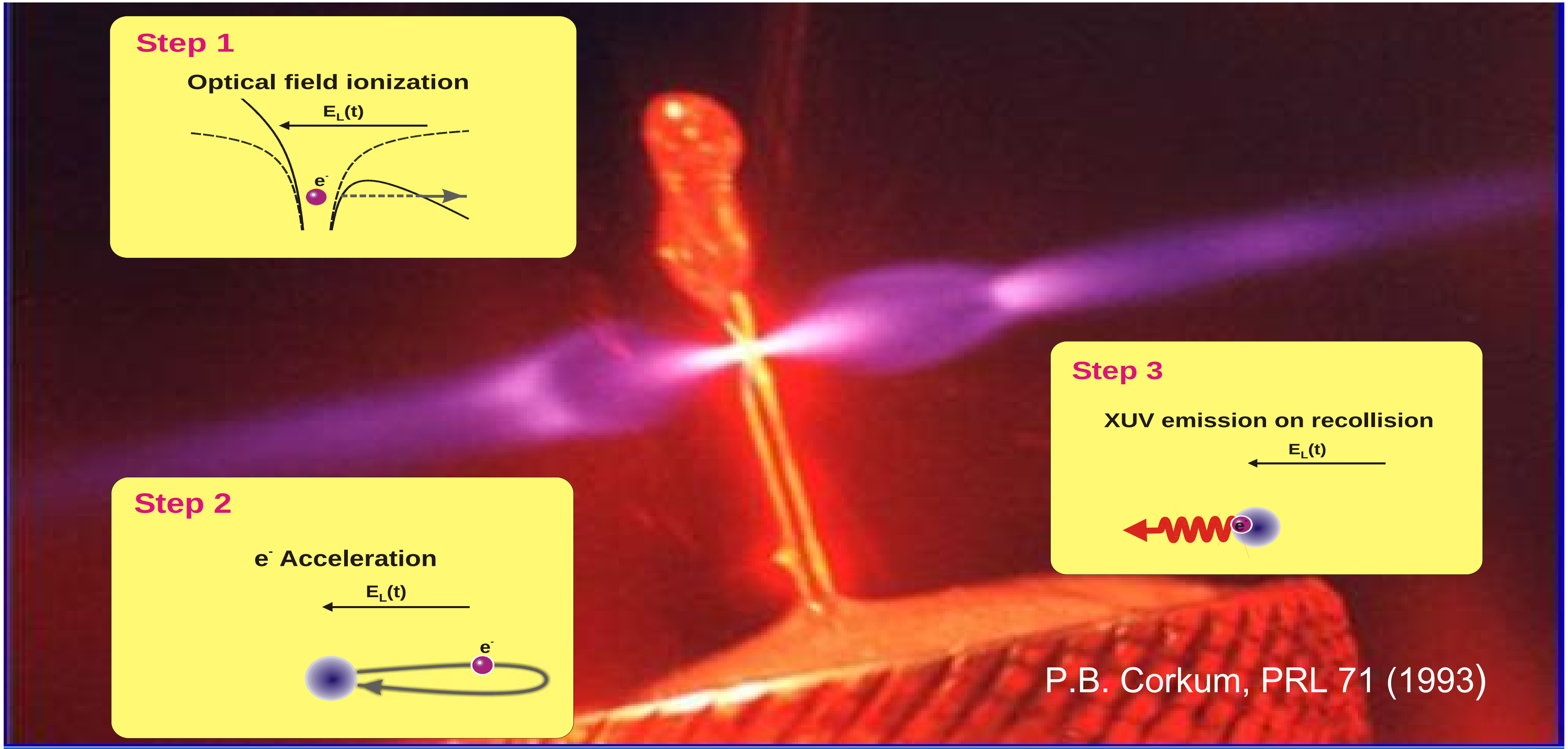
and SH $2f_m = 2f_0 + 2m \cdot f_r$

for k = 2m $2f_m - f_k = 2f_0 - f_0 + (2m - k) \cdot f_r = f_0 \equiv f_{ceo}$

Beat signal yields temporal evolution of φ_n

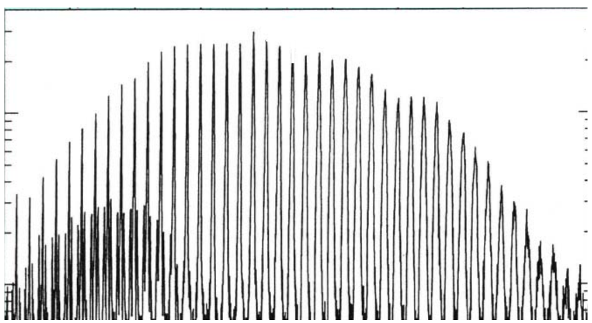
First implementation: D. Jones *et al.*, Science 288, 635 (2000); A. Apolonski *et al.*, PRL 85, 740 (2000)

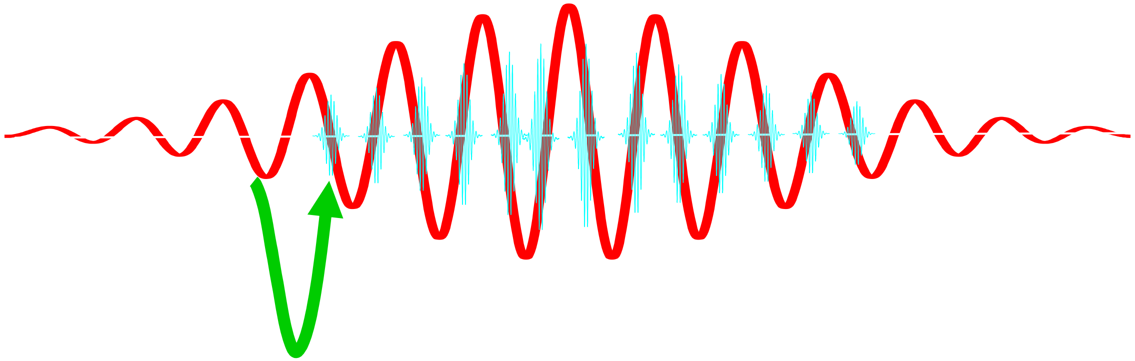
high-order harmonic generation in the gas phase



recombination emission from strongly-driven atoms

Multi-cycle driver pulse : $\tau_p \gg T_o$
High-order odd harmonics of the driver laser



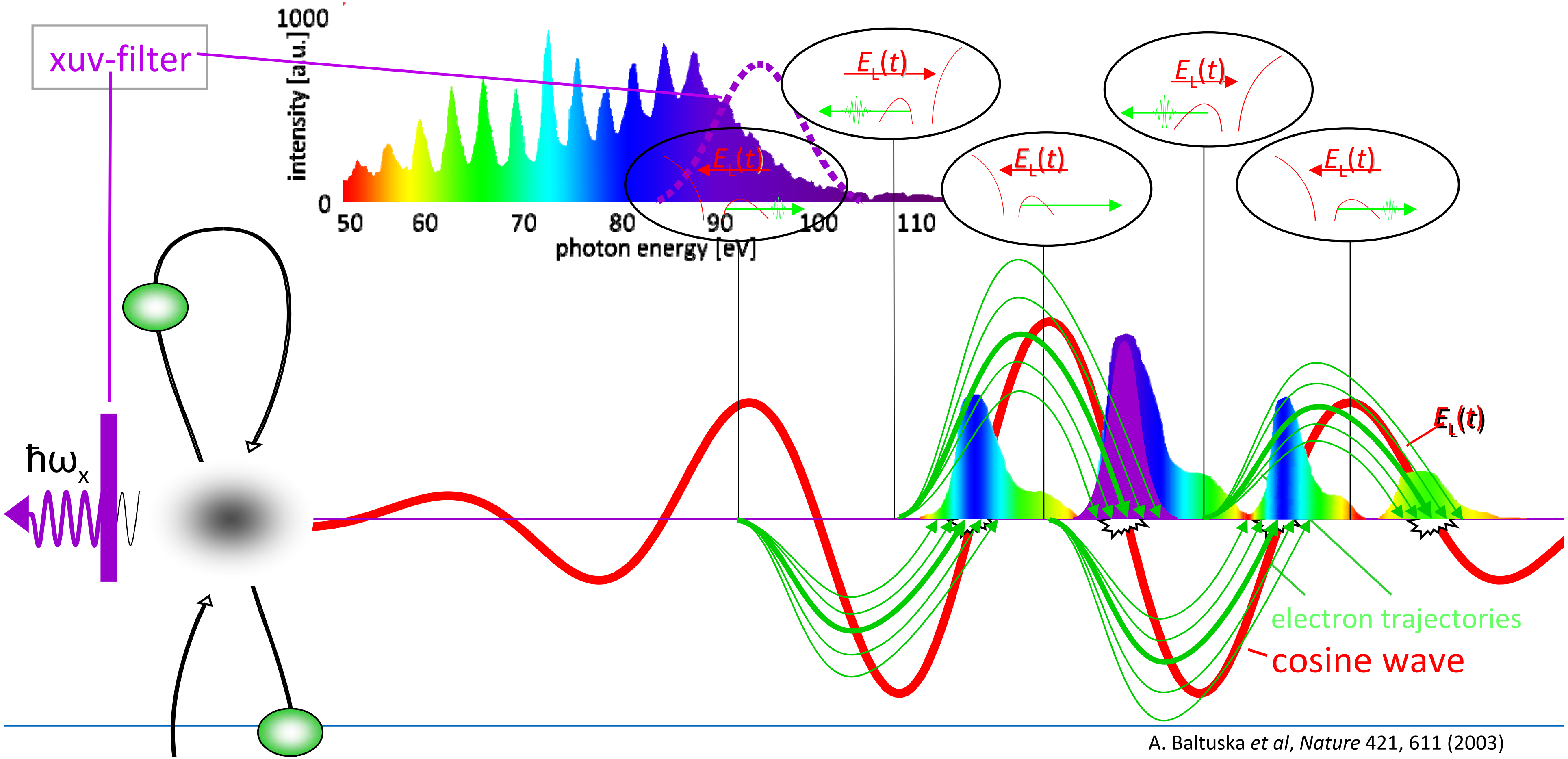


Cut-off harmonics: train of attosecond bursts

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

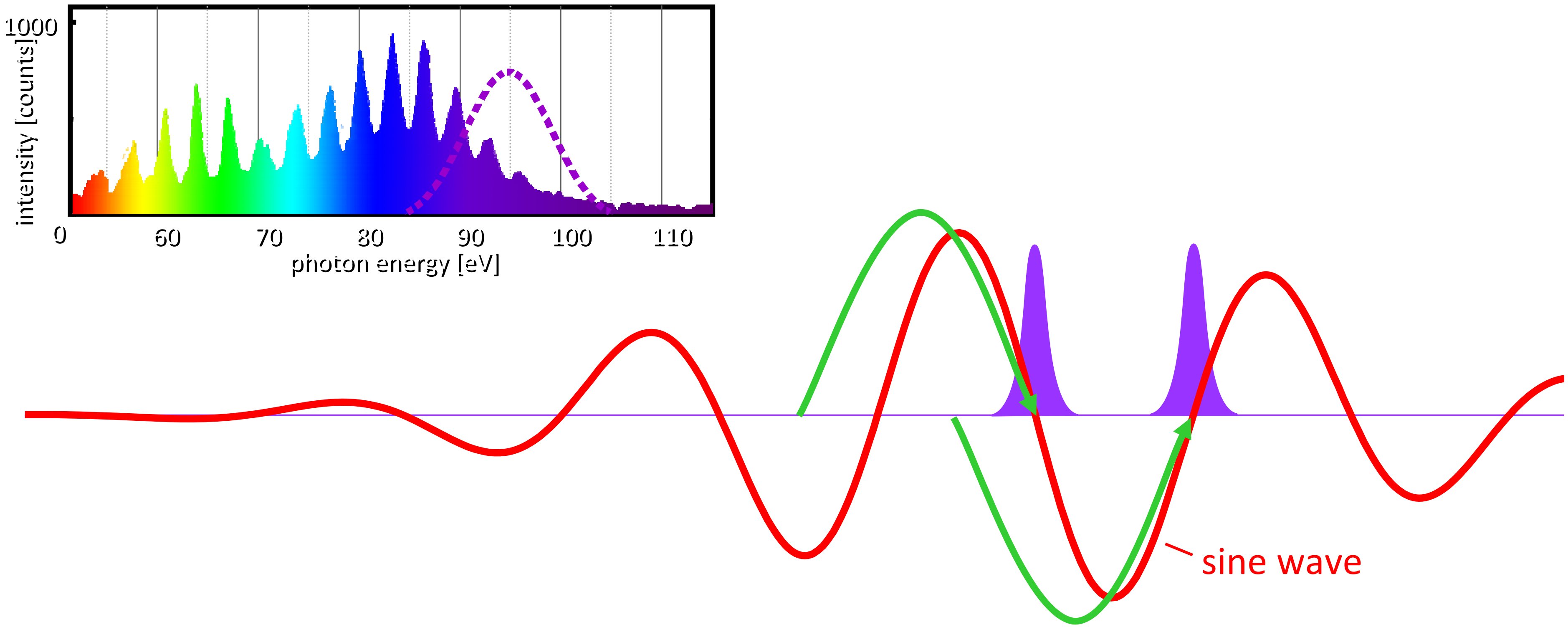
Paul *et al,* Science 292, 1689 (2001)
Tsakiris, Charalambidis *et al,* 2003

steering bound electrons with controlled light fields: the birth of an attosecond pulse



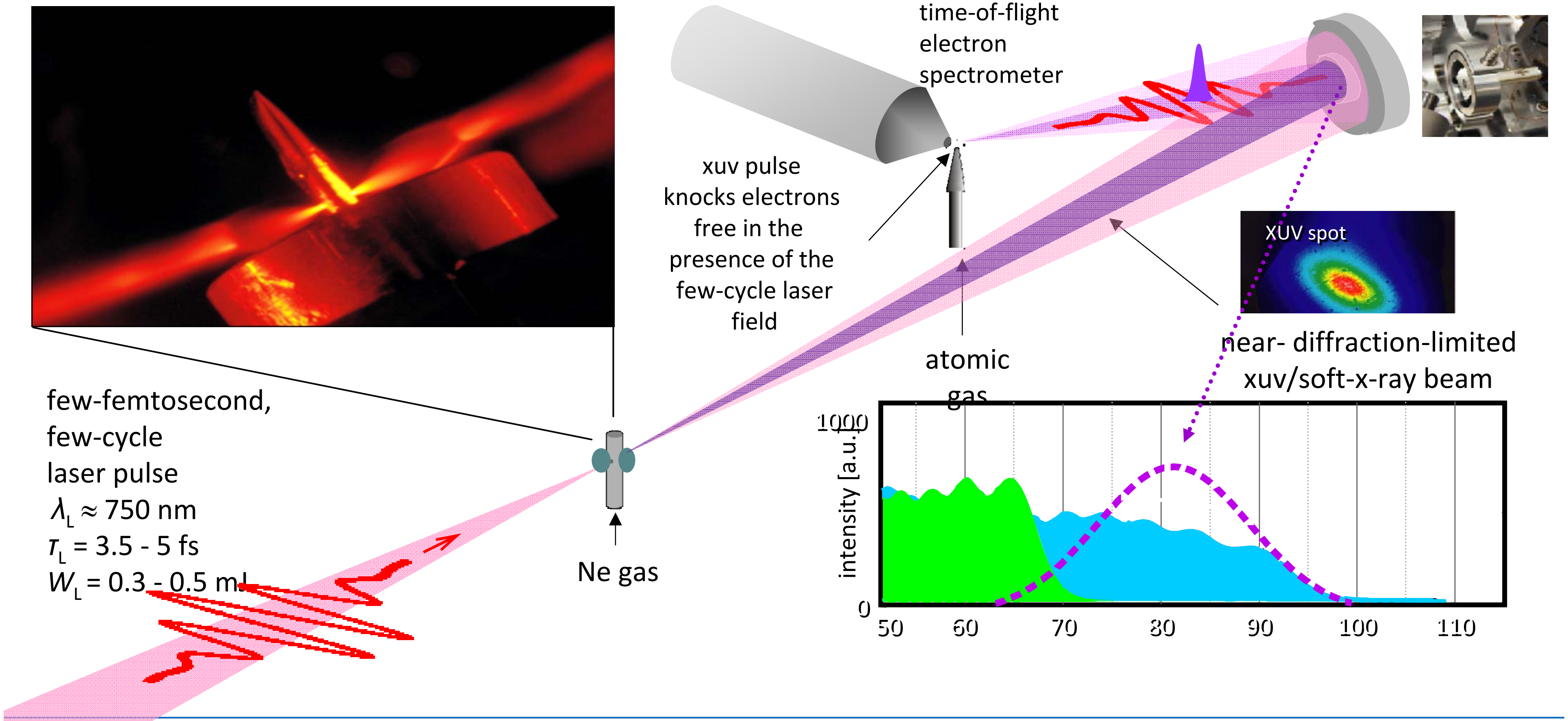
A. Baltuska *et al*, *Nature* 421, 611 (2003)

attosecond xuv/soft x-ray pulse generation depending on the CEP



A. Baltuska *et al.*, *Nature* 421, 611 (2003)

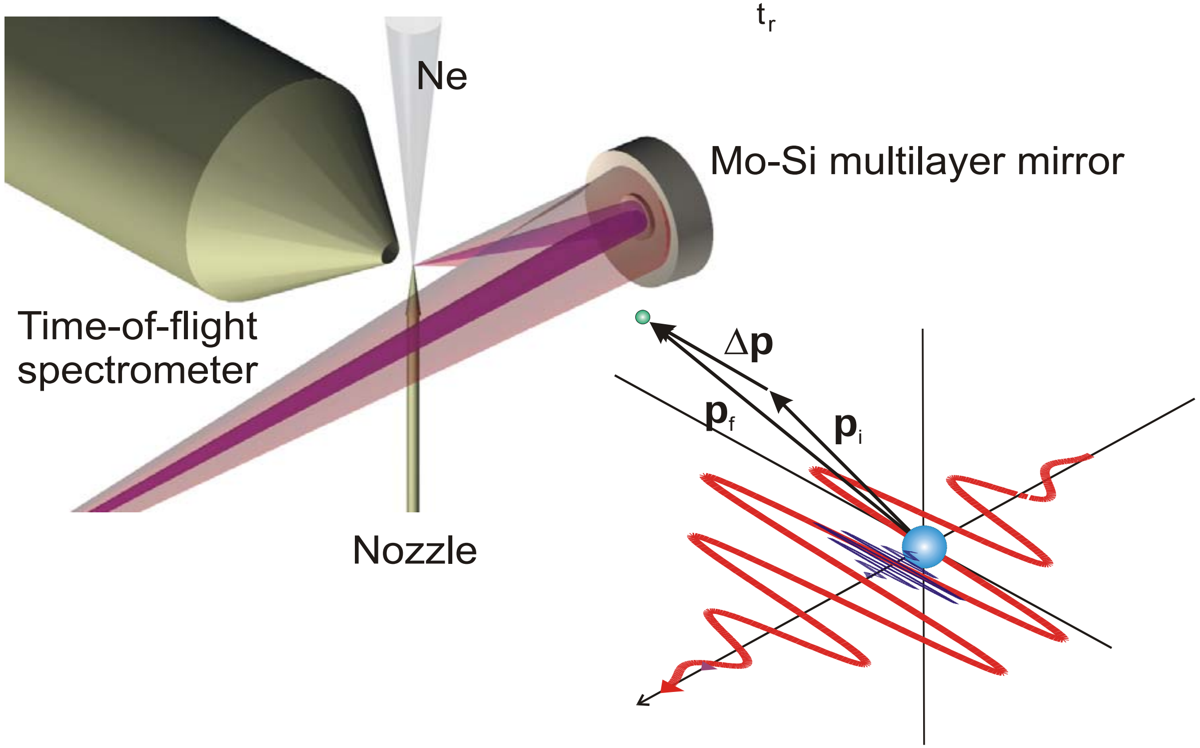
attosecond pulse generation and measurement



Drescher *et al*, *Science* 291, 1923 (2001); Hentschel *et al*, *Nature* 414, 509 (2001); Kienberger *et al*, *Science* 297, 1144 (2002)

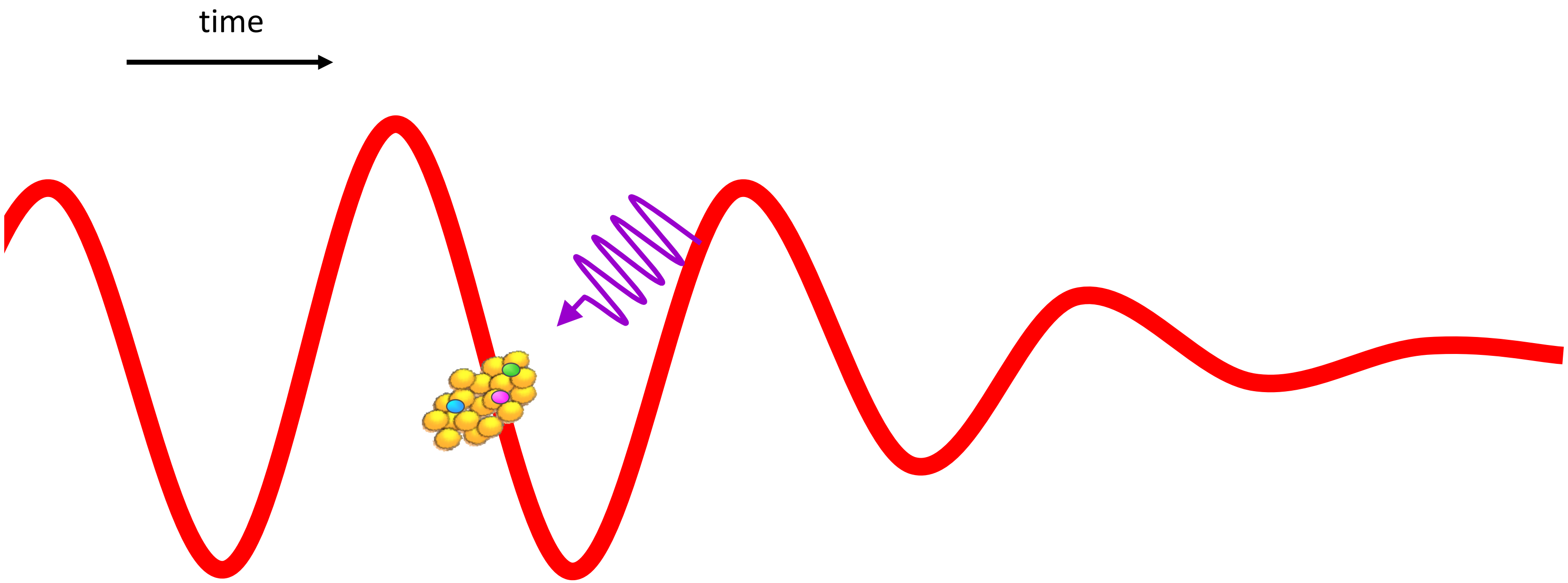
photoelectrons generated by an as-pulse

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

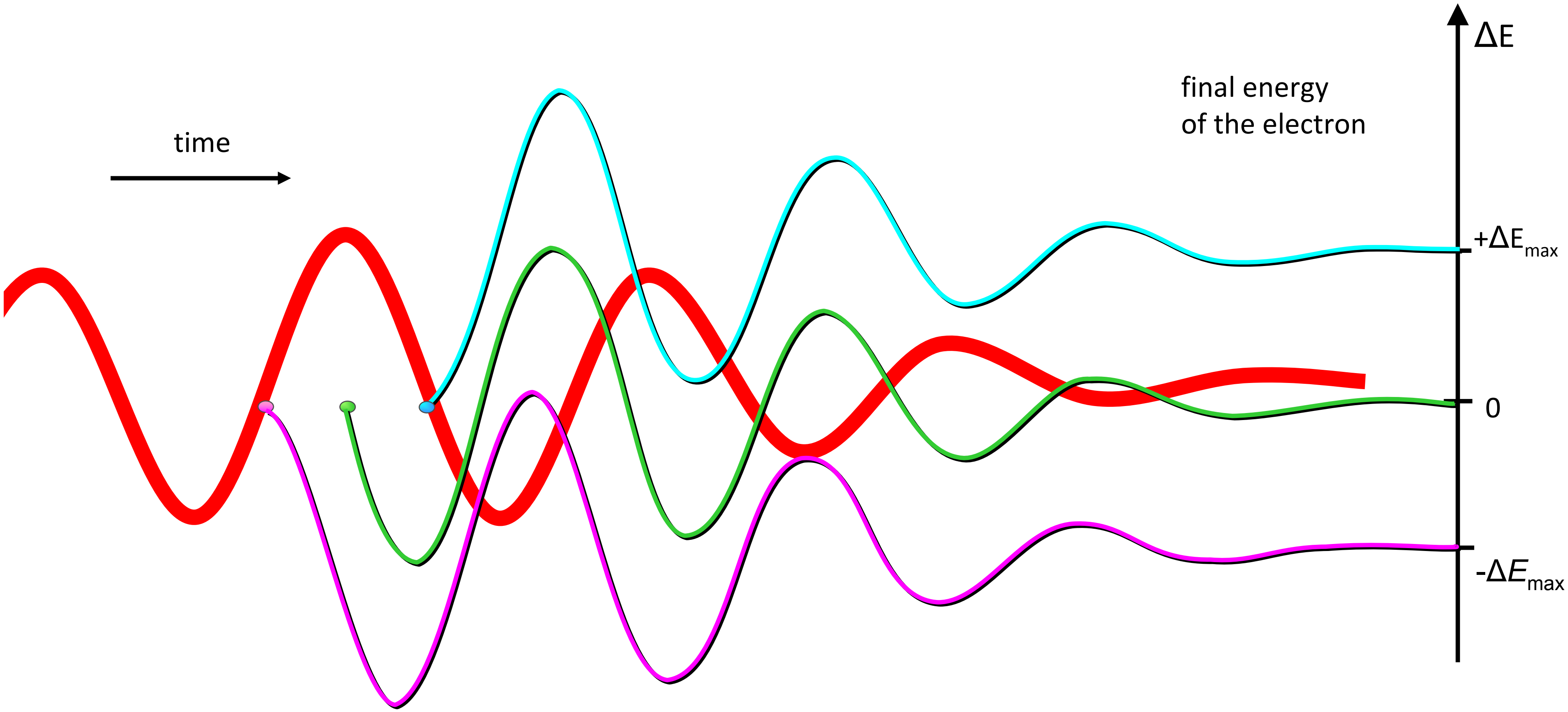


Kienberger *et al.*, Science 297, 1144 (2002)

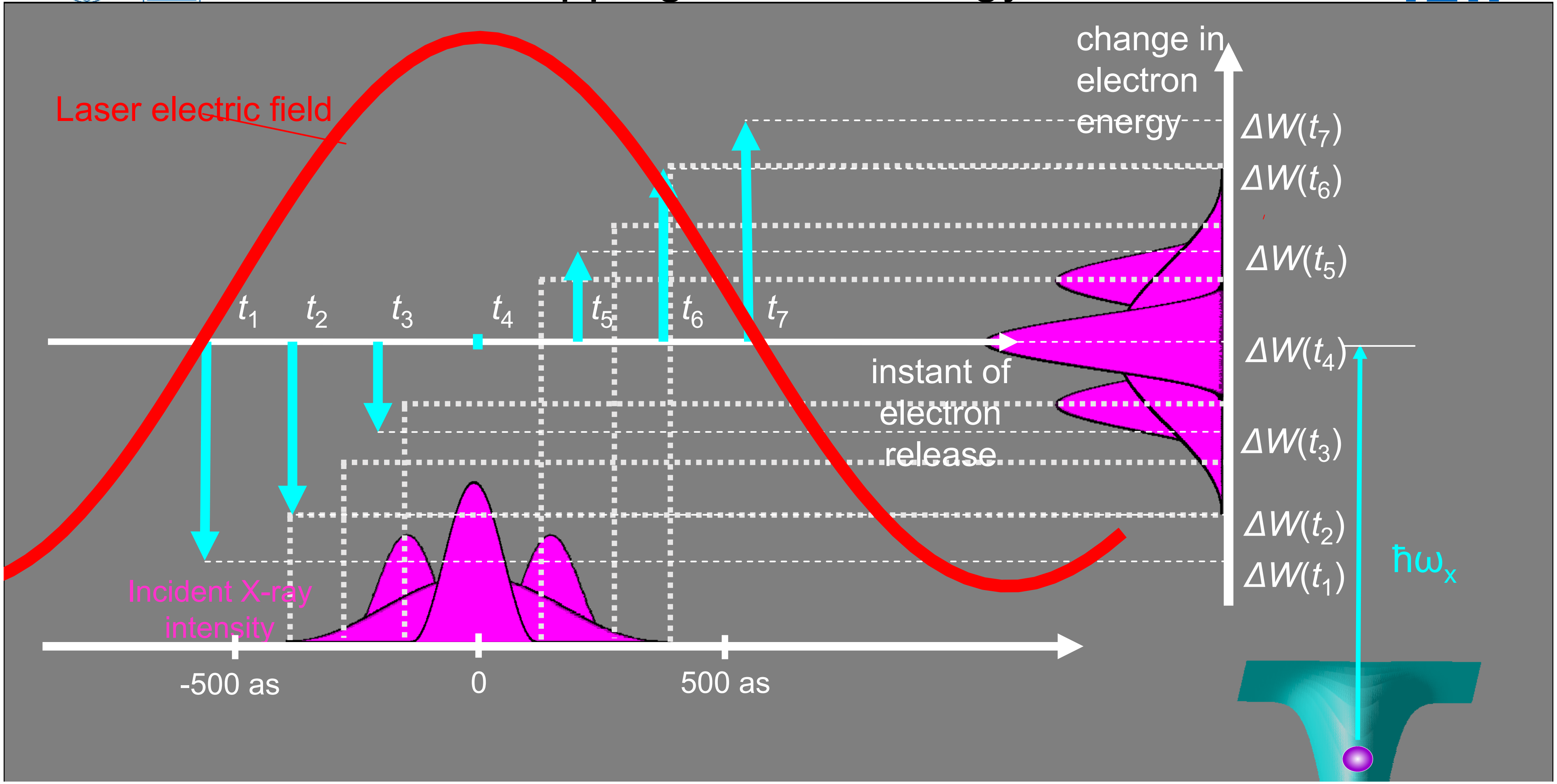
ionization at different instants of time



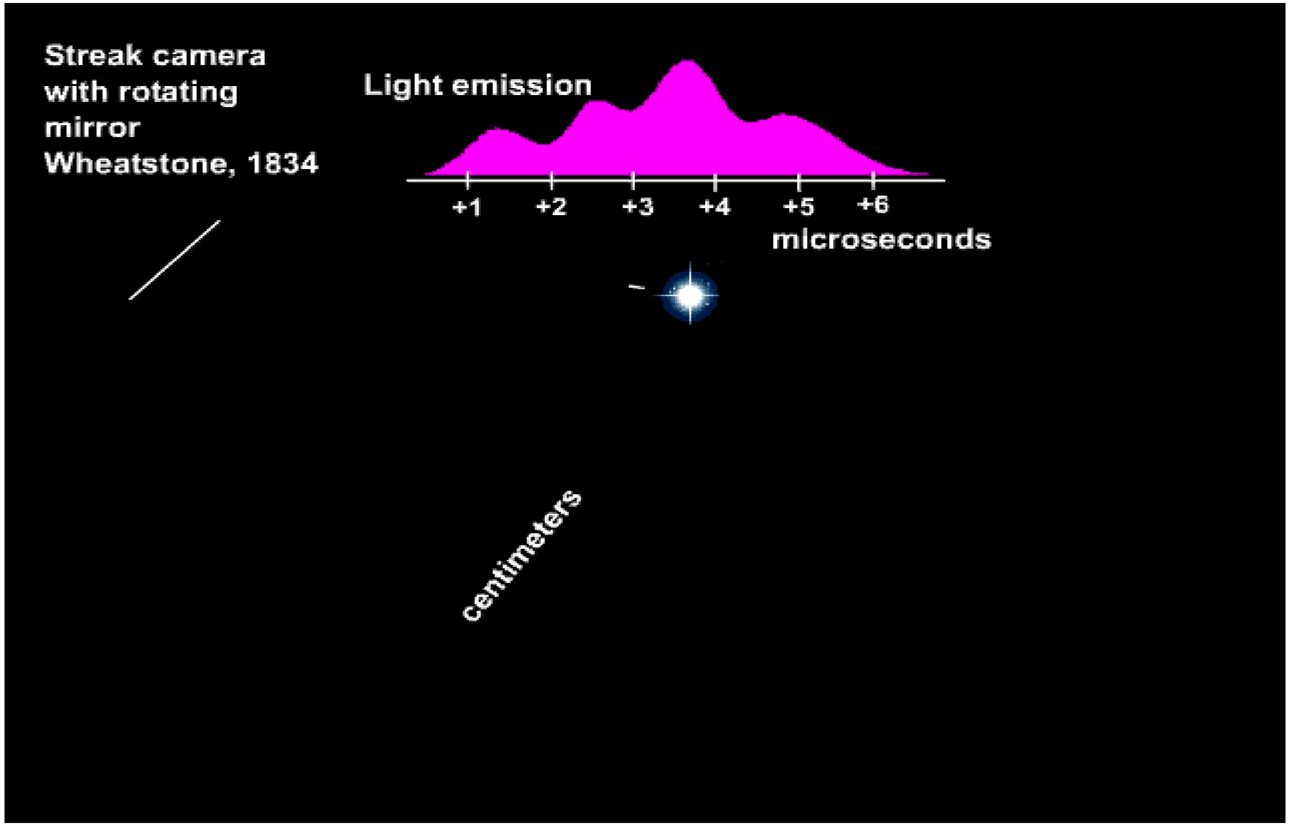
final energy of photoelectrons depends on the release time



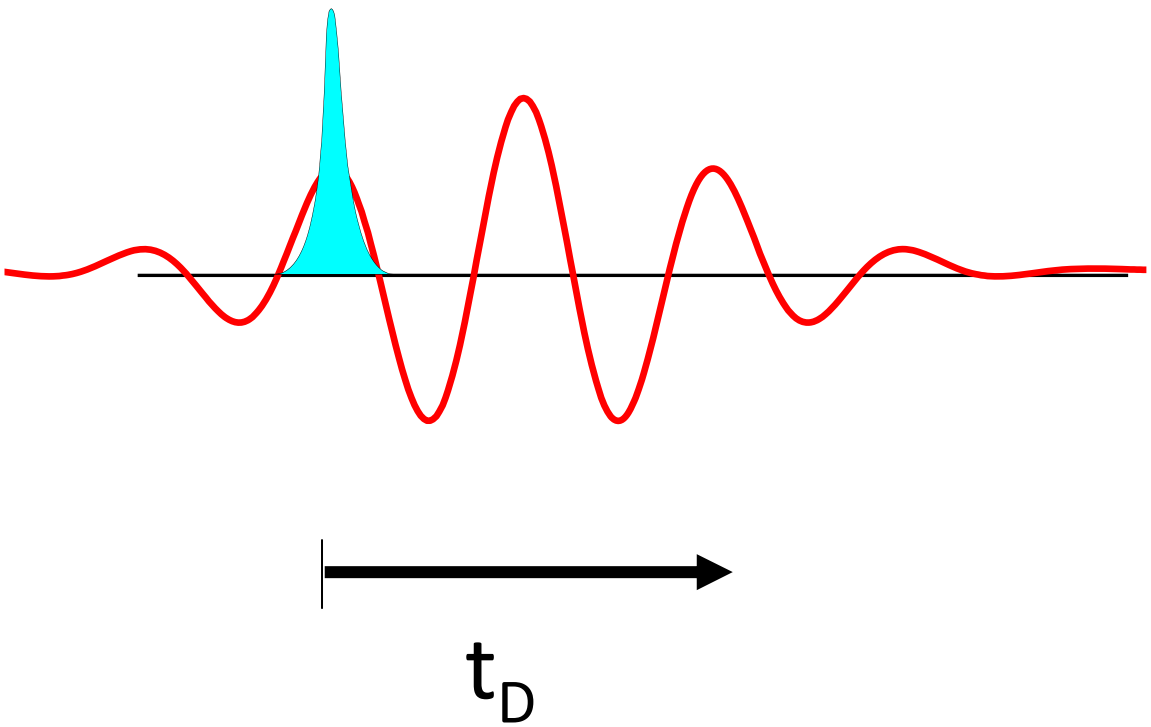
mapping time to energy

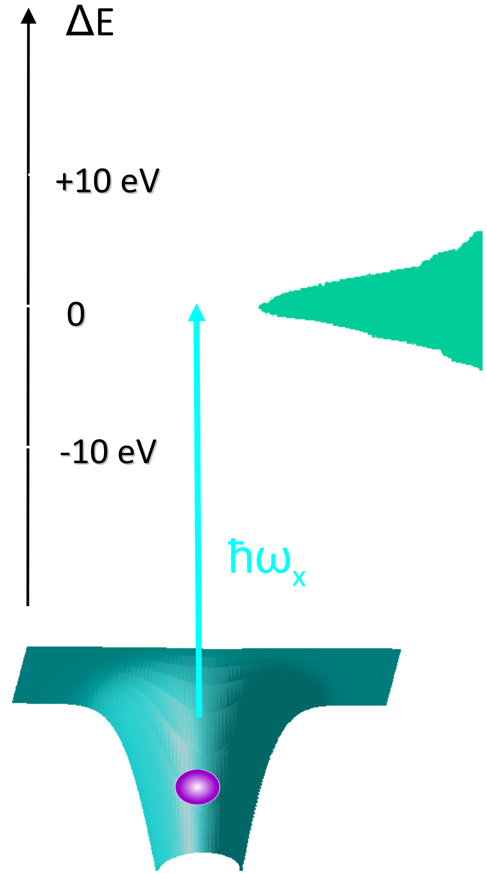


optical streak camera, 1834



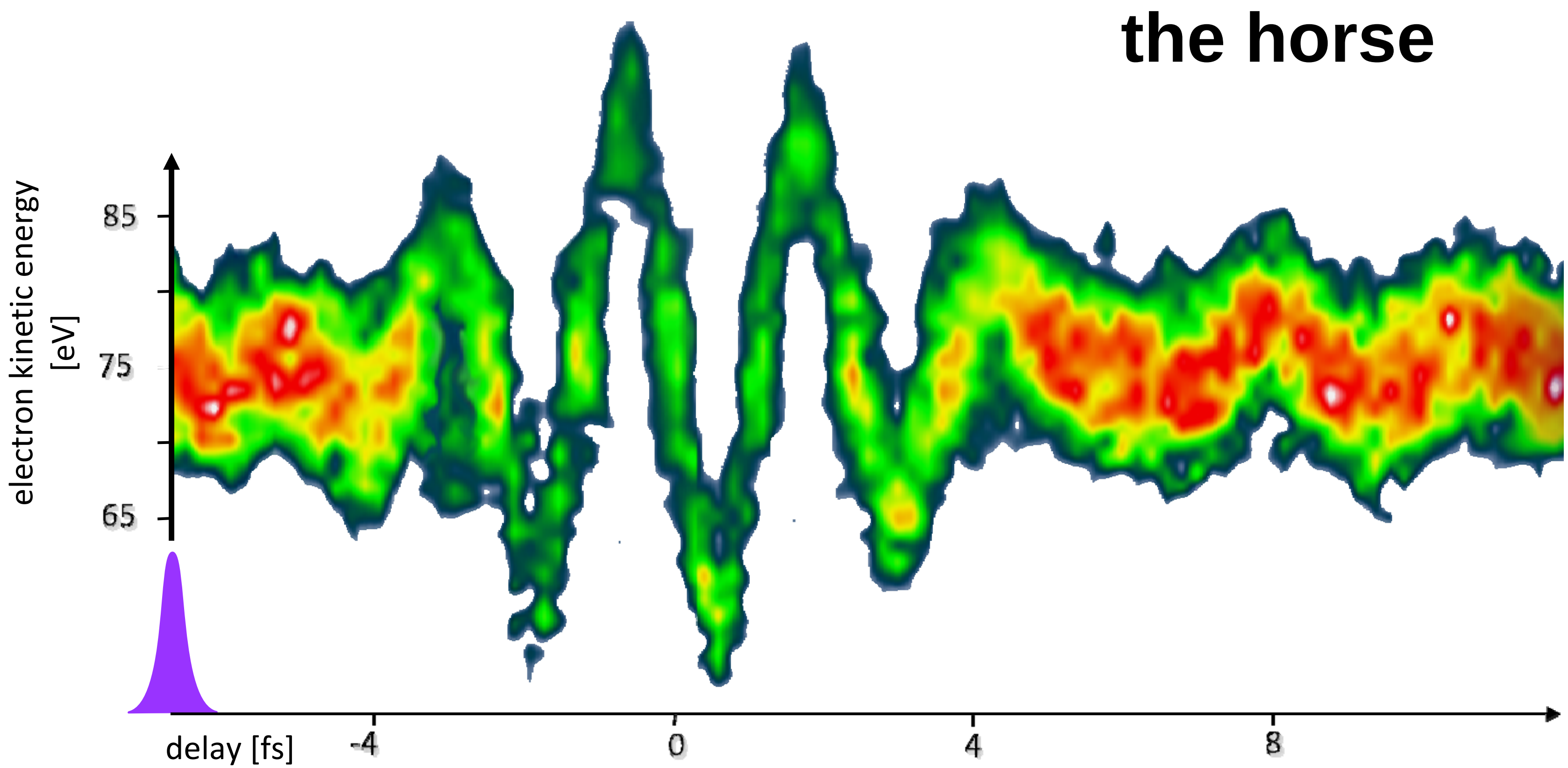
sampling field oscillations



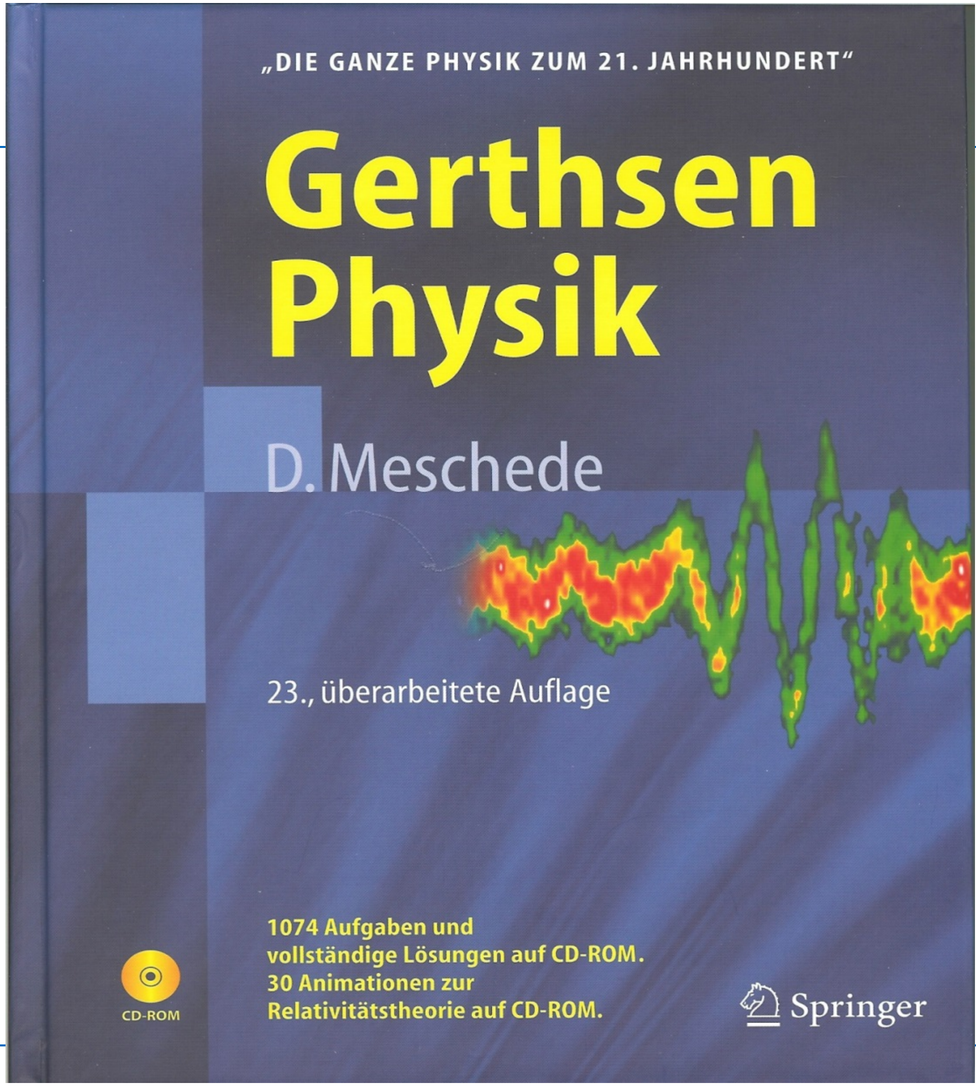


R. Kienberger et al, Nature 294 (2004)

the spectrogram: “a streaking trace”



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


„DIE GANZE PHYSIK ZUM 21. JAHRHUNDERT"
Gerthsen Physik
D. Meschede
23., überarbeitete Auflage
1074 Aufgaben und vollständige Lösungen auf CD-ROM.
30 Animationen zur Relativitätstheorie auf CD-ROM.
CD-ROM
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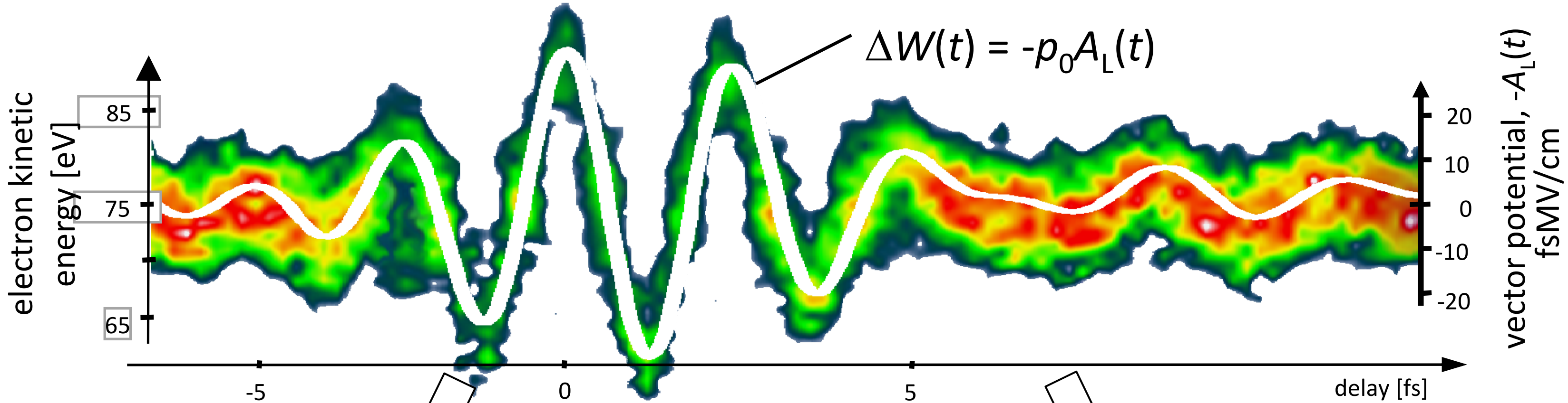
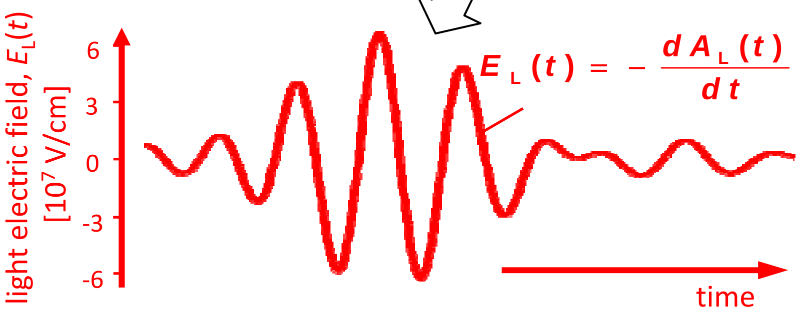
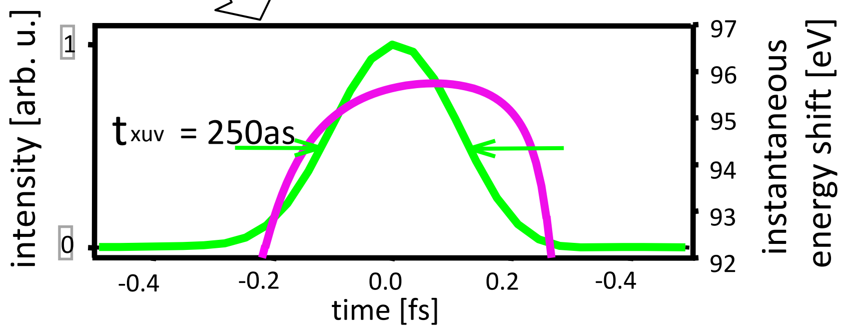
attosecond streak camera: complete measurement of a few-cycle light wave *and* a sub-fs xuv pulse





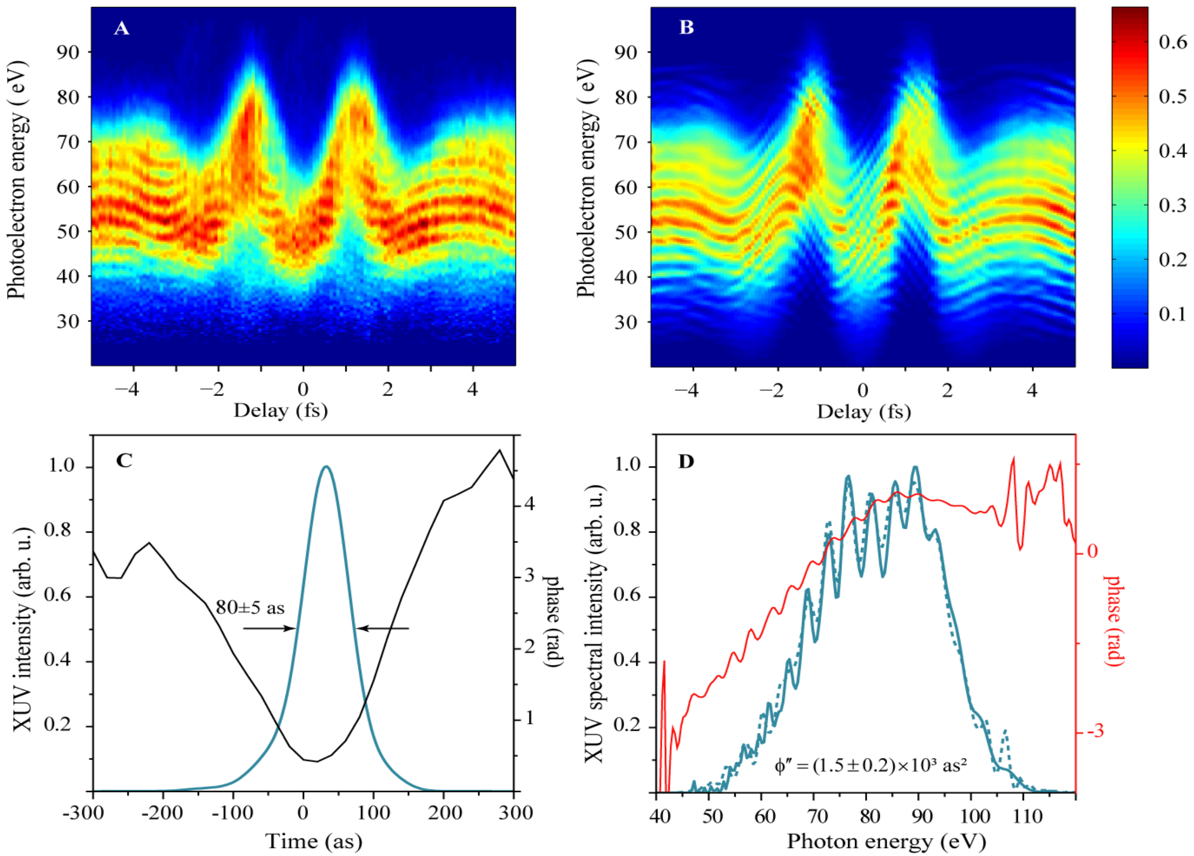
electric field of few-cycle light wave



amplitude envelope & frequency chirp of sub-fs XUV pulse

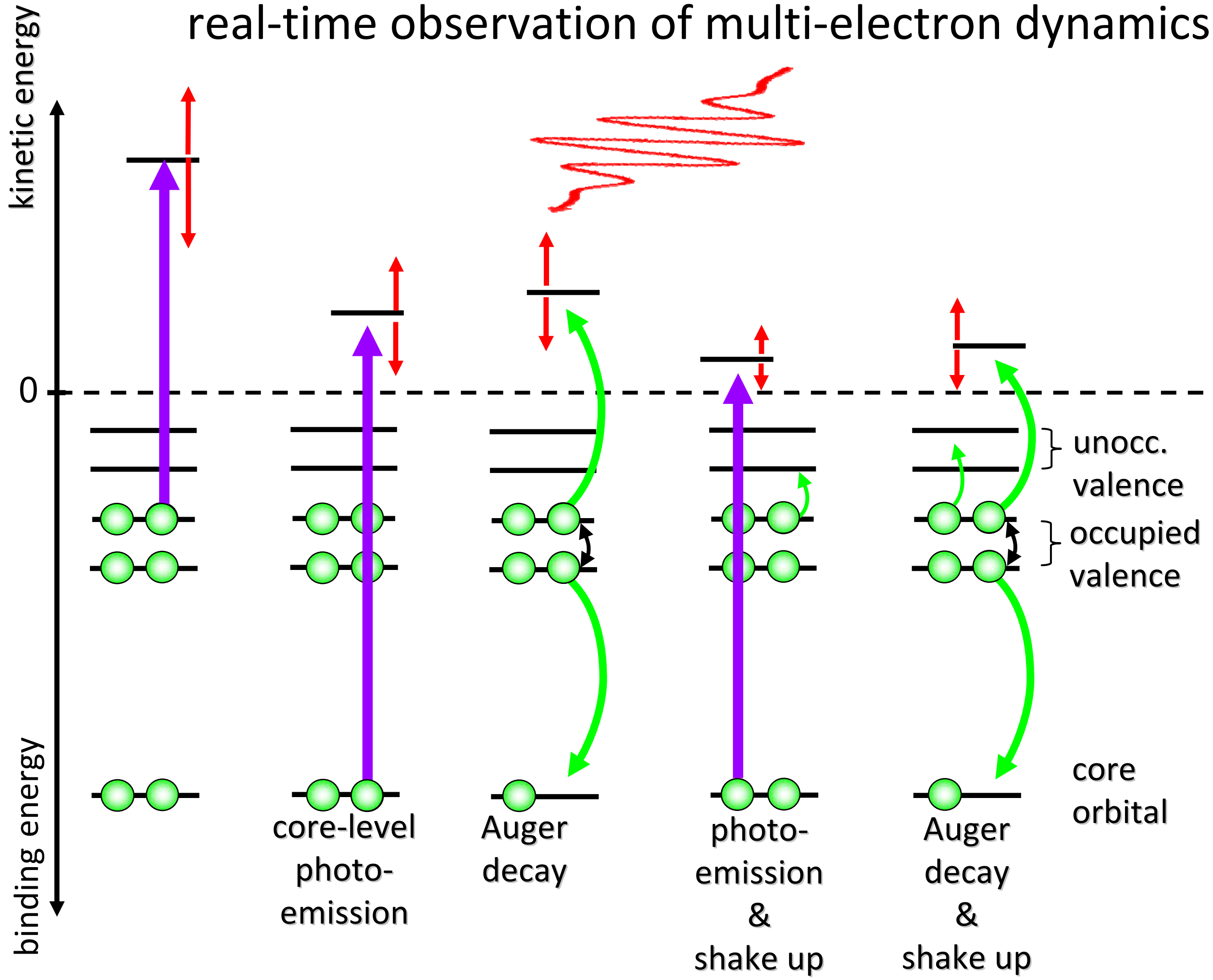
J. Itatani et al., PRL 88 (2002); Goulielmakis *et al*, *Science* 305, 1267 (2004)

FROG-like spectrograms of sub-100-as pulses

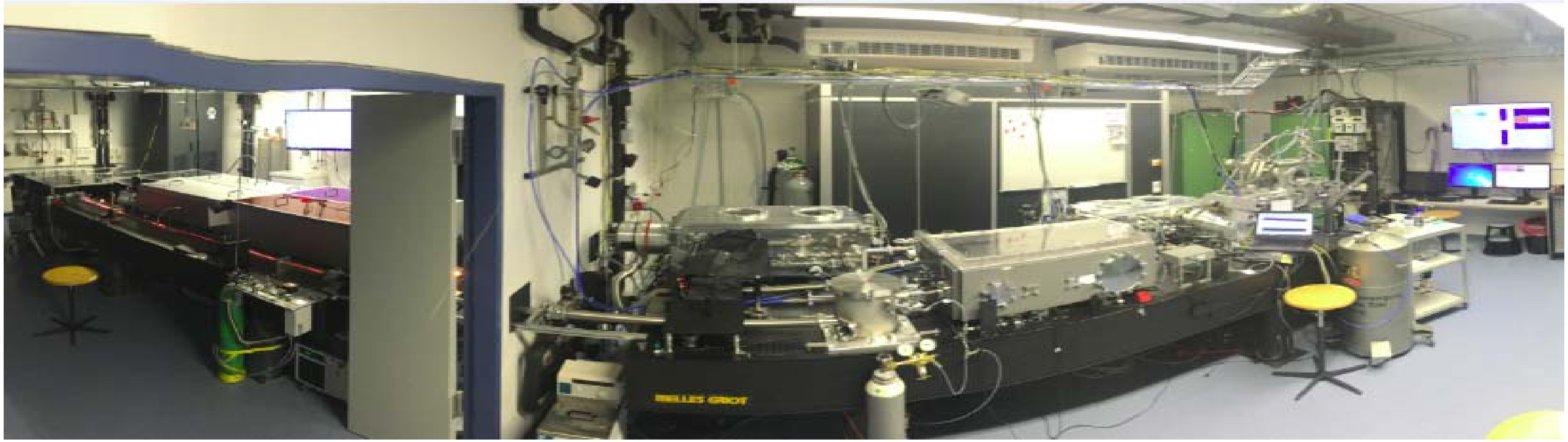


Goulielmakis *et al*, *Science* 320, 1614 (2008)

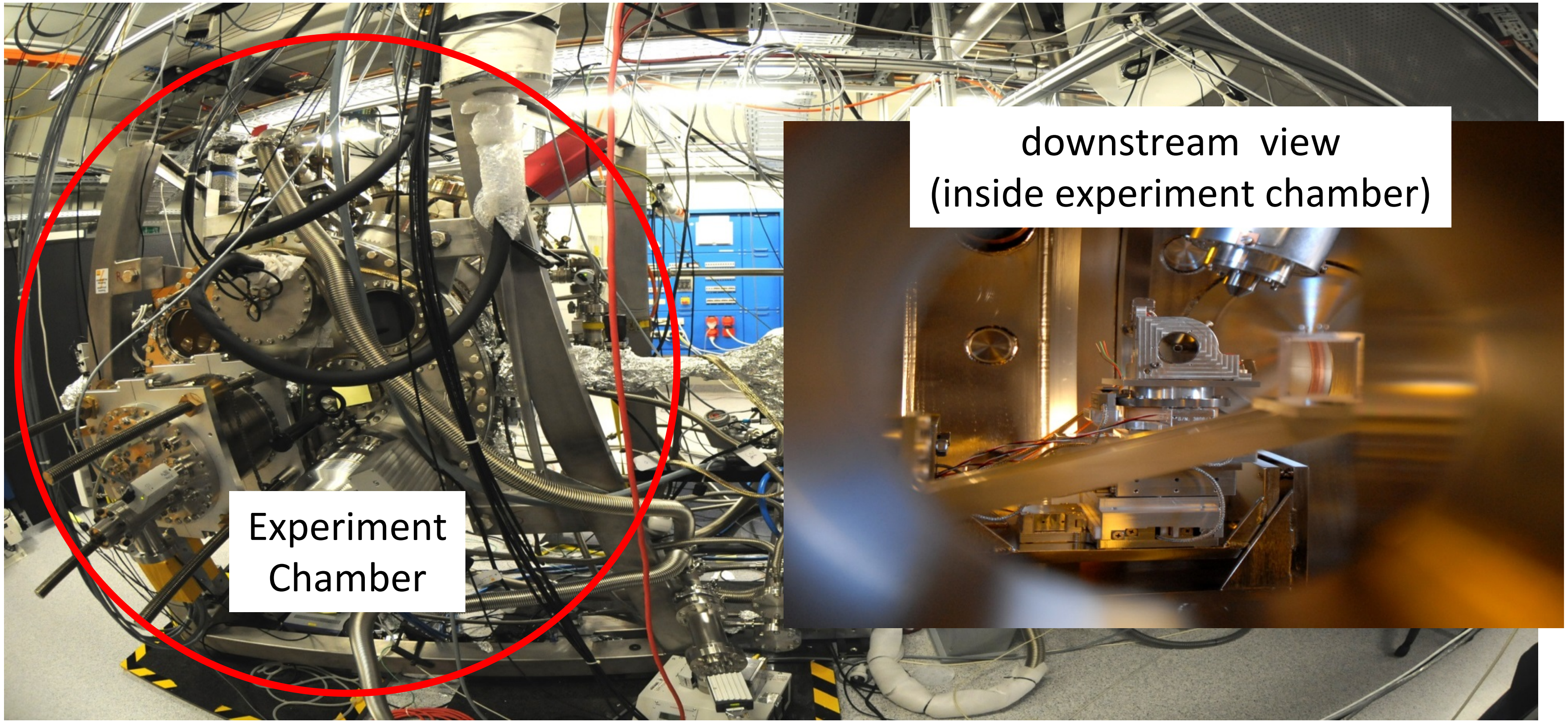
real-time observation of multi-electron dynamics



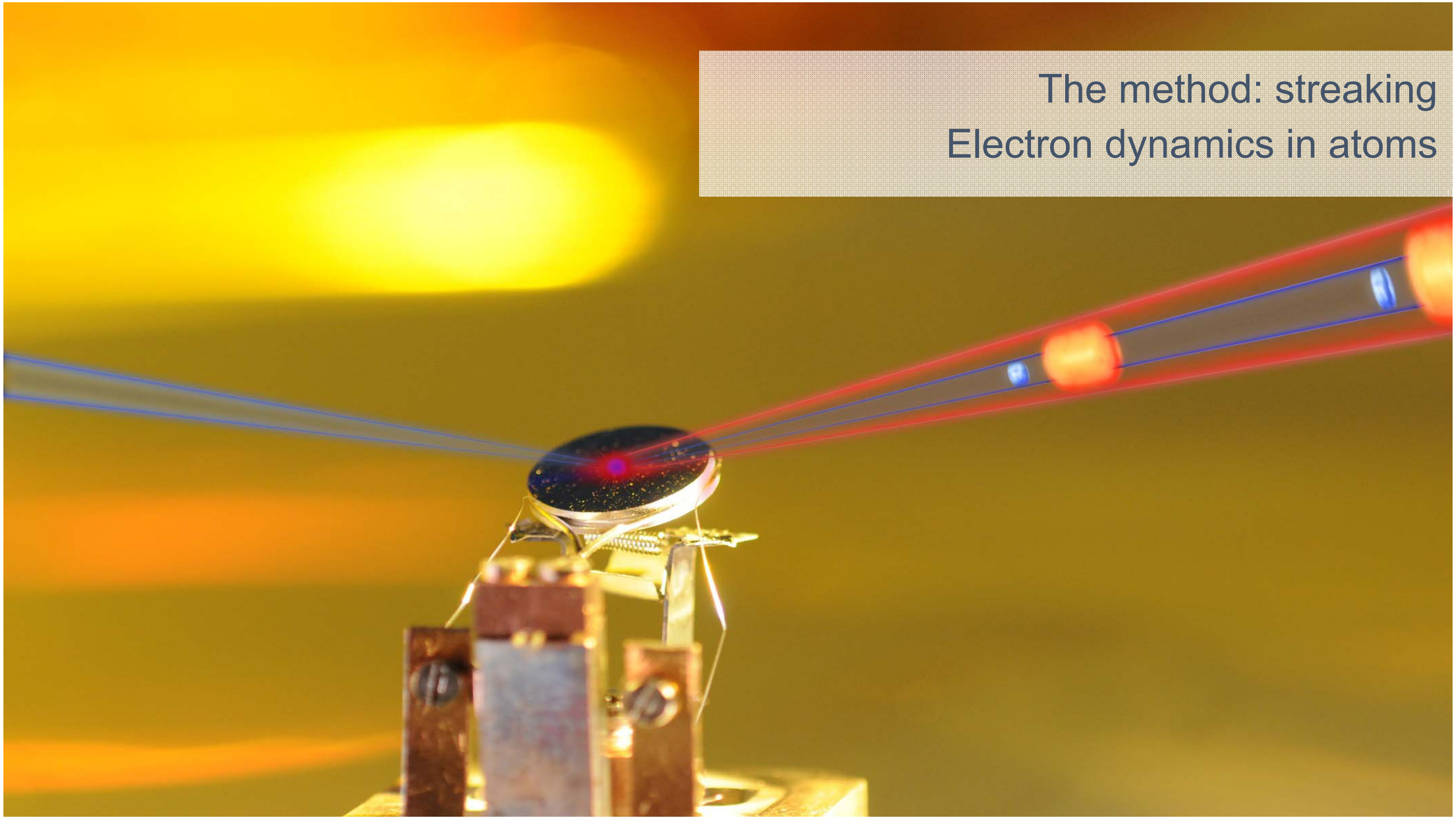
AS-beamline at TUM



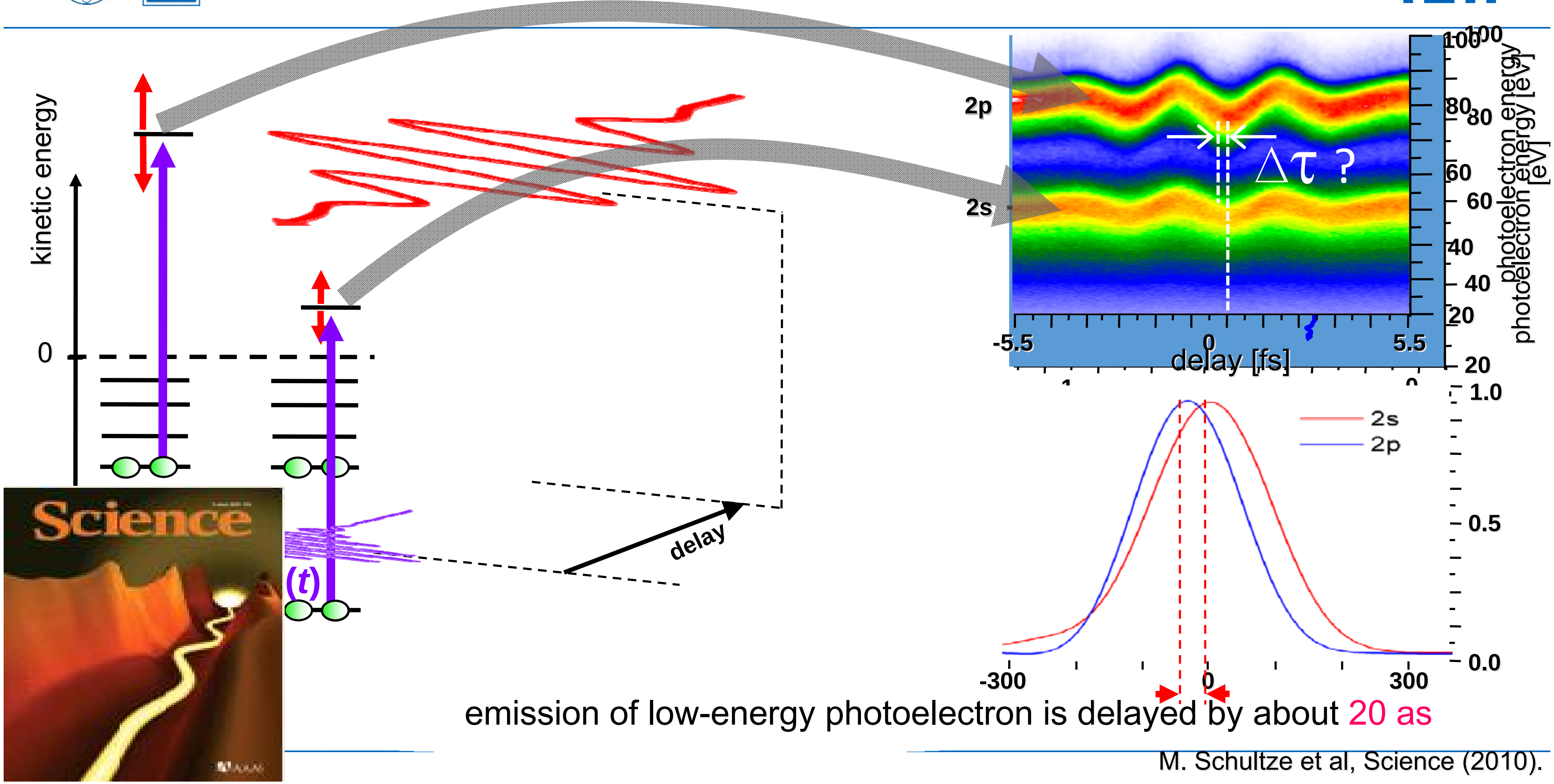
AS3 beamline



3x10^{-11} mbar 5x10^{-9} mbar 5x10^{-3} mbar

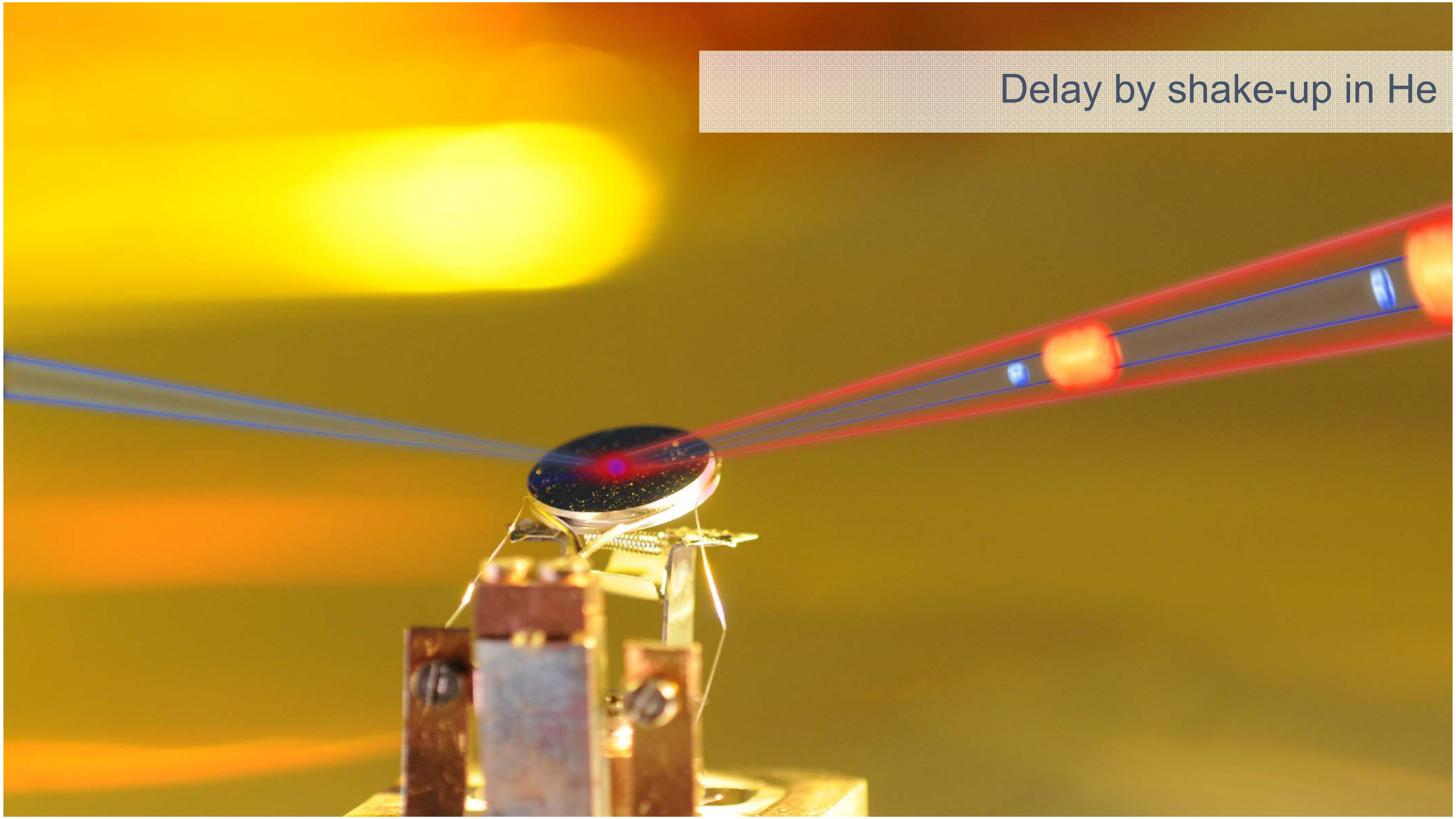

The method: streaking
Electron dynamics in atoms

What about single photon ionization? Ne 2s / 2p

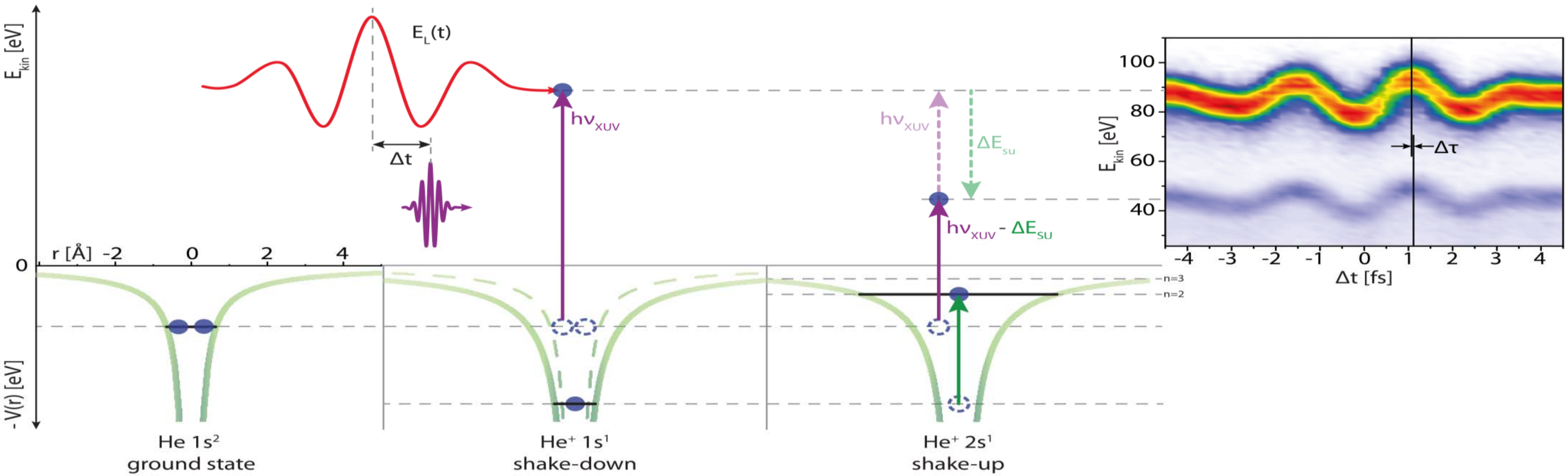


emission of low-energy photoelectron is delayed by about 20 as

M. Schultze et al, Science (2010).

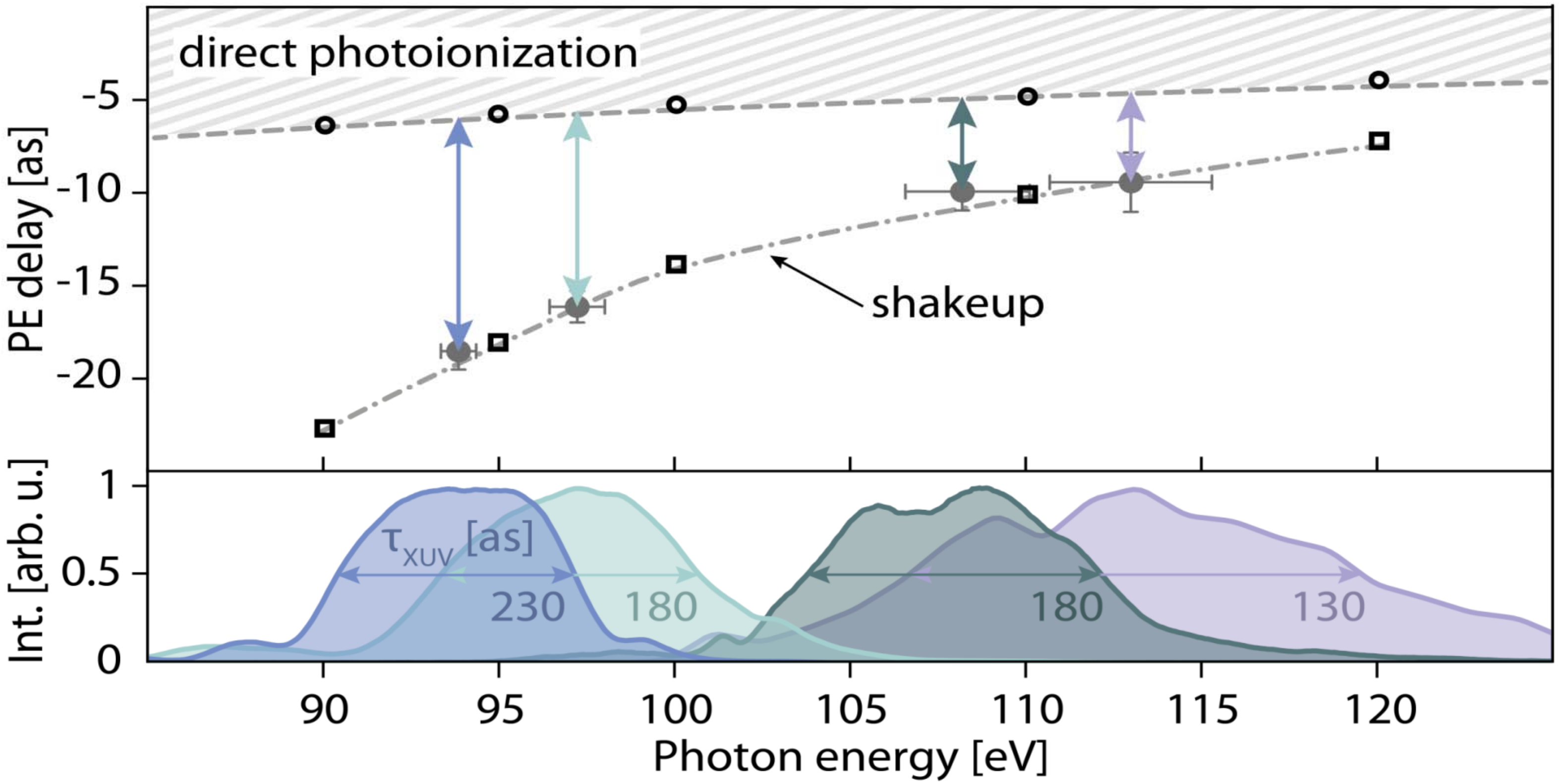

Delay by shake-up in He

photoemission in Helium direct PE vs. shake-up



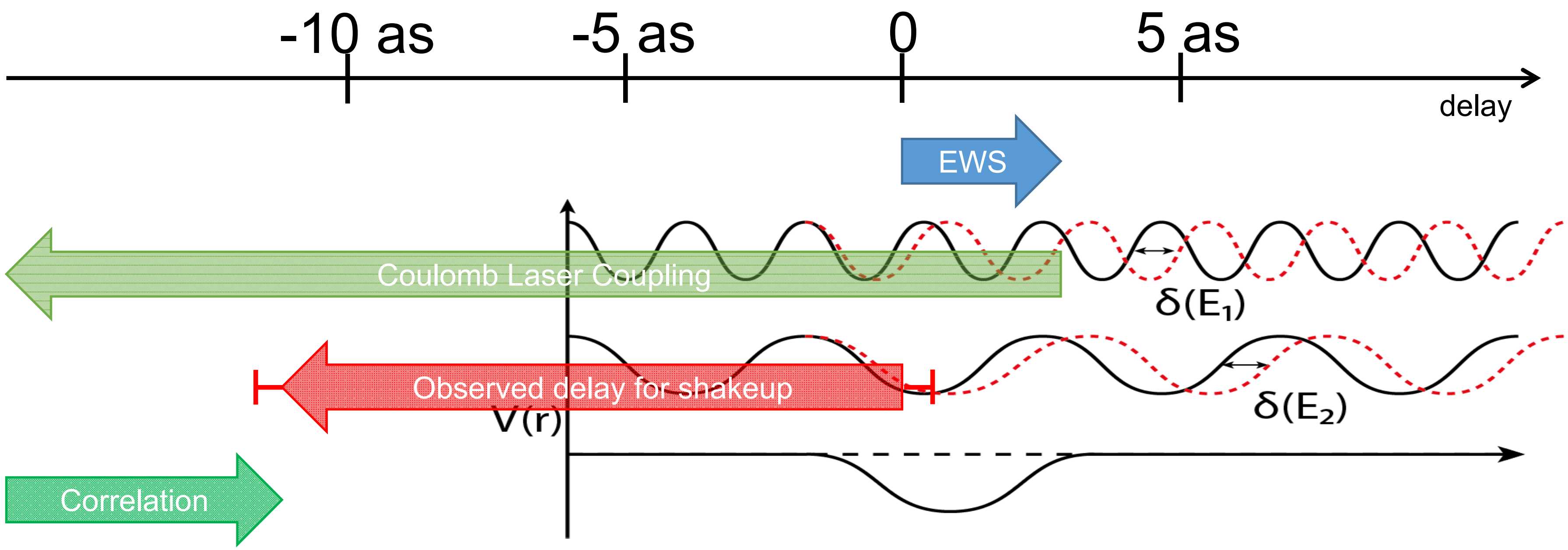
Ossiander et al., Science 2016

timing of photoemission in Helium



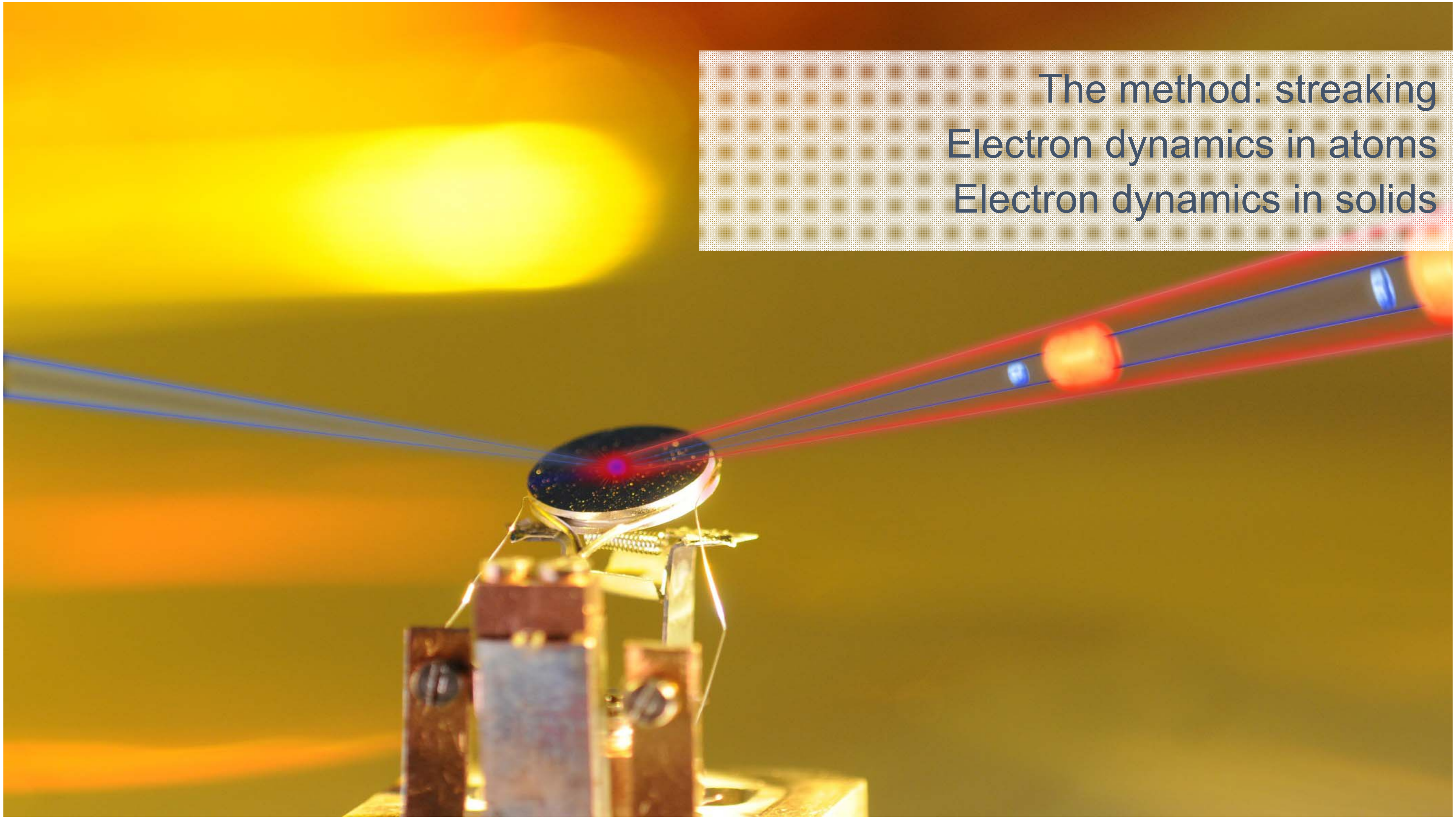
Ossiander et al., Science 2016

delay contributions

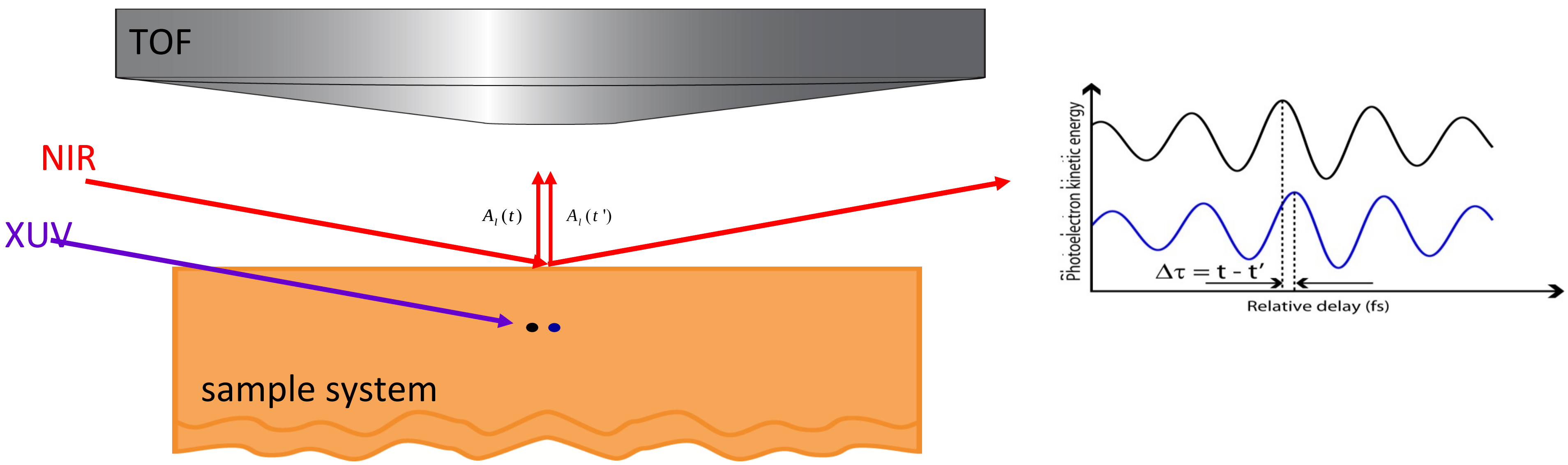


depending on energy: between 5as and 12 as

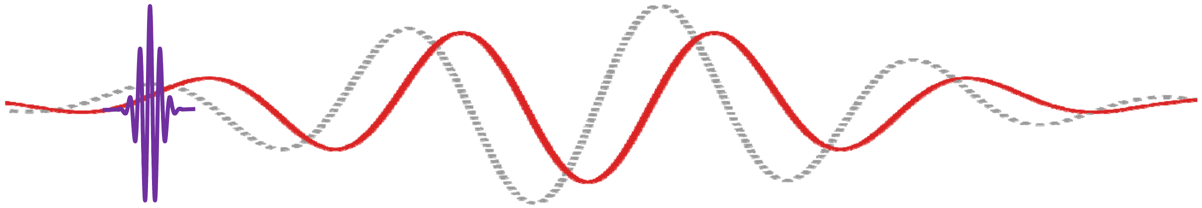
Pazourek, R., Time-resolved photoemission in one- and two-electron atoms. (TU Vienna, 2013). at <http://katalog.ub.tuwien.ac.at/AC10774478>

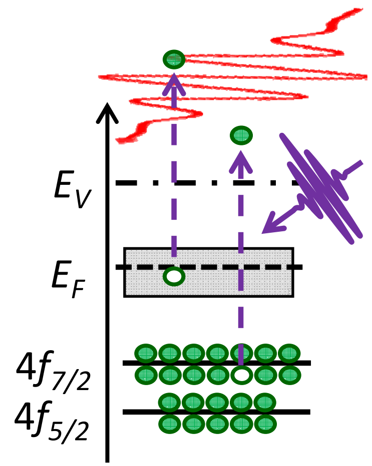

The method: streaking
Electron dynamics in atoms
Electron dynamics in solids

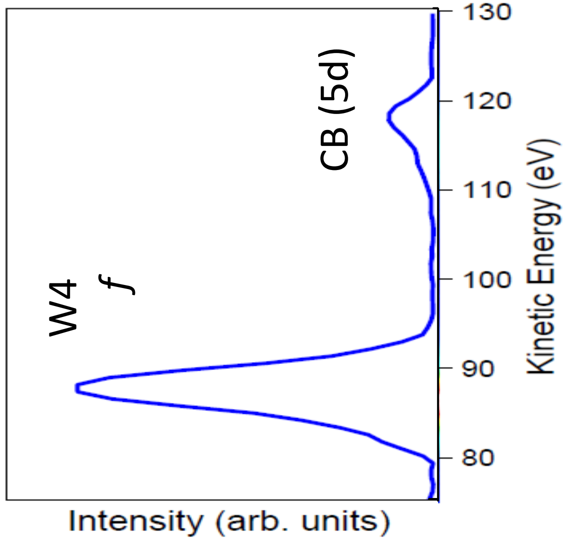
ionization of electrons in a solid



streaked photoemission from W(110)







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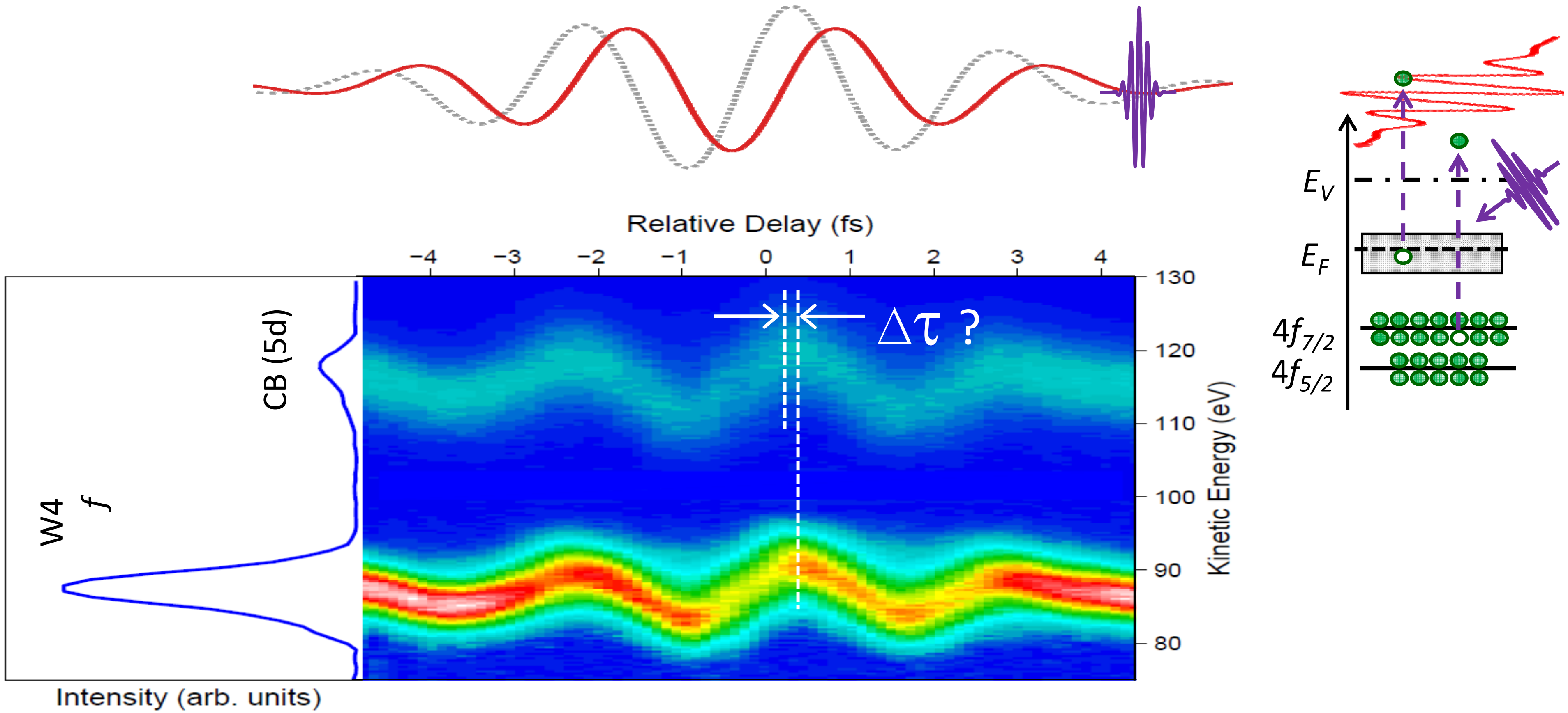
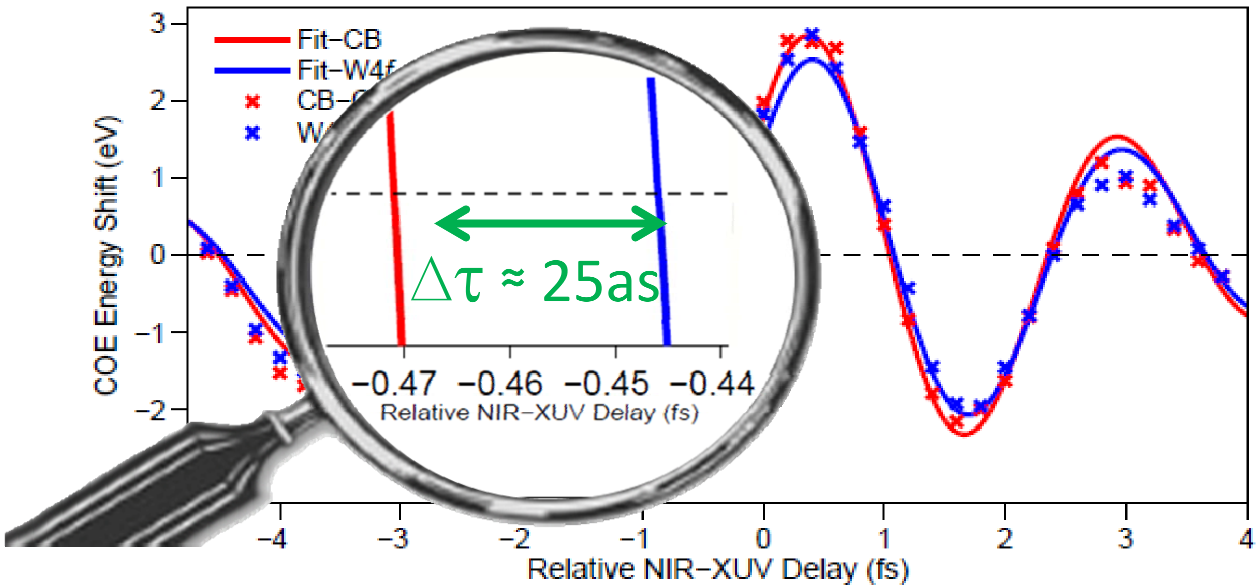
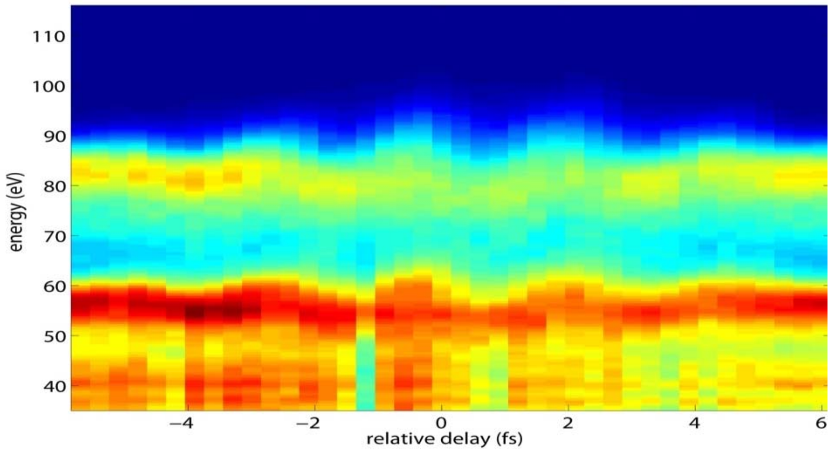
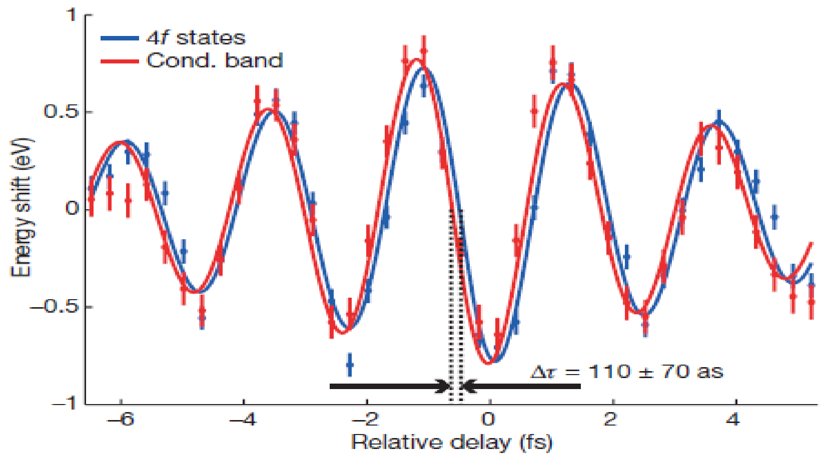




- Emission of the slower W4*f* core electrons is delayed

experiment from 2007





Sample: W(110) single crystal
Excitation: 300as XUV pulses centered at ~91eV

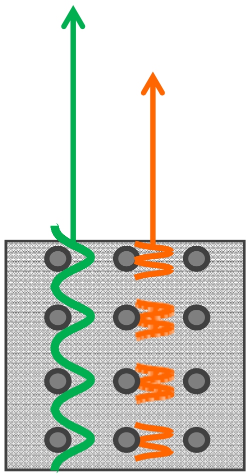
- ➢ Emission of W4*f* electrons delayed by 110 ± 70as with respect to emission of CB electrons (derived from a *single* measurement)

Cavalieri *et al*, Nature 449, 1029 (2007)

different ways of interpretation

Time delay in photoemission form solids is mainly due to:

…the difference in the spatial
 localization of the initial-state
wave functions of CB and
core-level electrons.
Surface states?

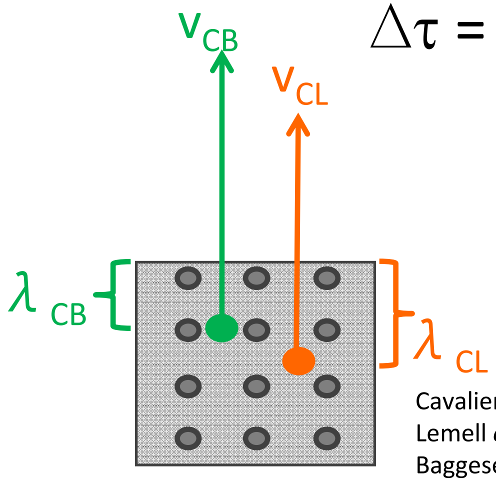


Zhang *et al*, PRA 84, 065403 (2011)
Zhang *et al*, PRL 102, 123601 (2009)
Kazansky *et al*, PRL 102, 177401 (2009)

…a propagation effect of the
escaping electrons:

$$\Delta\tau = \lambda_{CL}/v_{CL} - \lambda_{CB}/v_{CB}$$

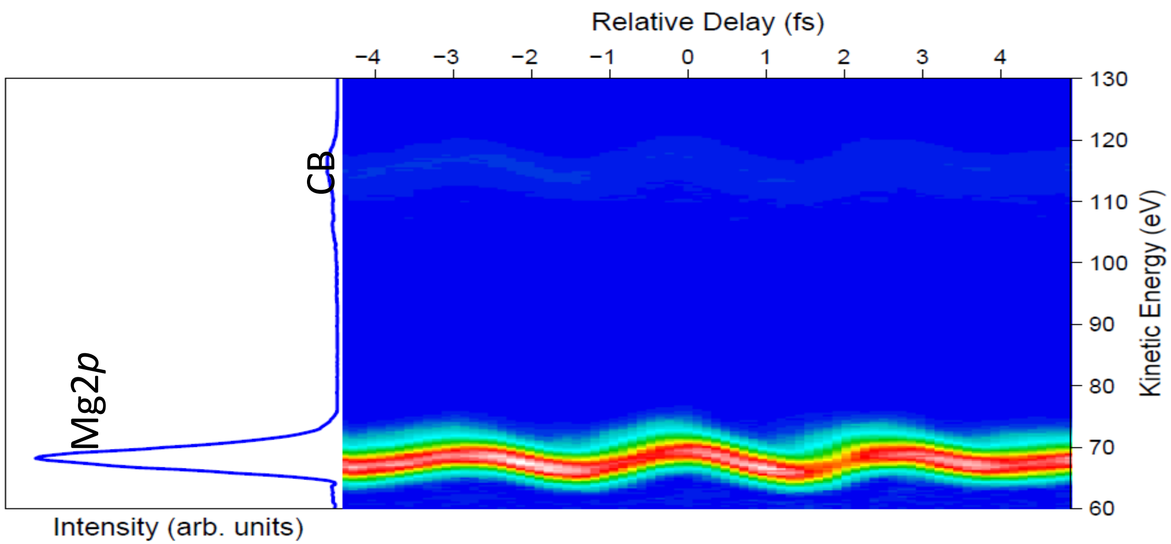
Impact of final-state
band-structure?

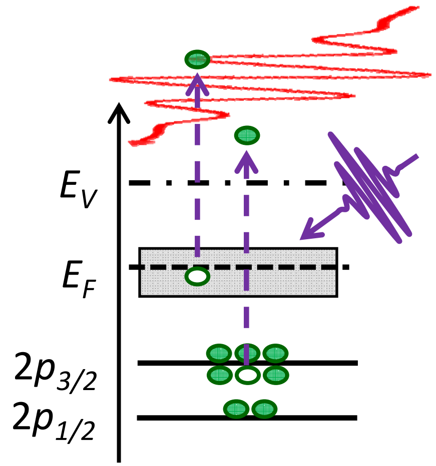


Cavalieri *et al*, Nature 449, 1029 (2007)
Lemell *et al*., PRA 79, 062901 (2009)
Baggesen *et al*., PRA 80, 030901 (2009)

attosecond streaking of Mg(0001)

- Ideal test case: truly delocalized CB electrons & localized 2*p* core-level electrons
- Free-electron-like final-state band structure
- Challenging: reactive material & low CB density of states





Neppl *et al*, PRL 109, 087401 (2012)

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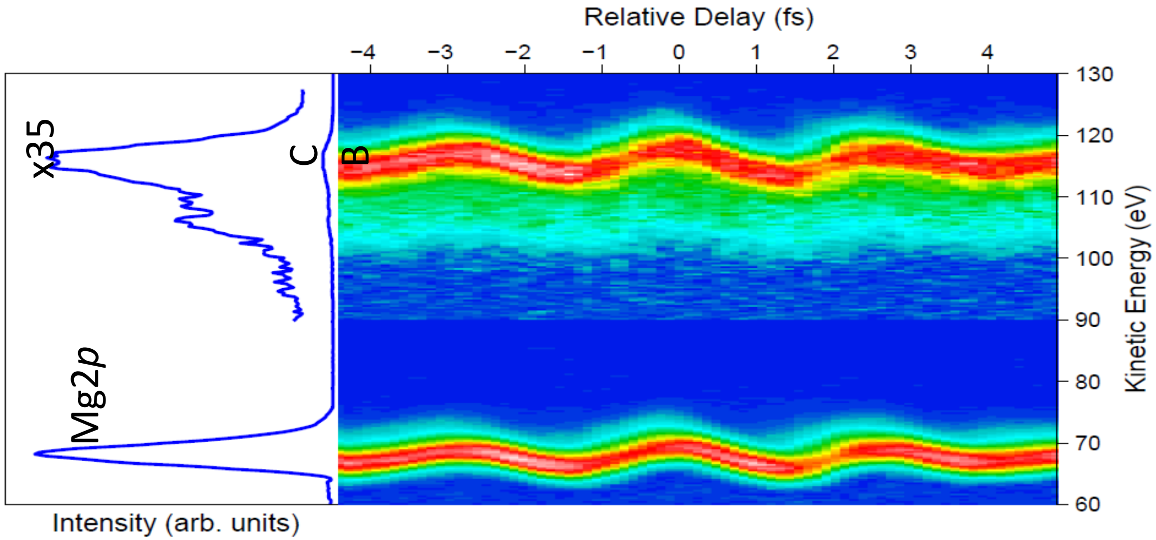
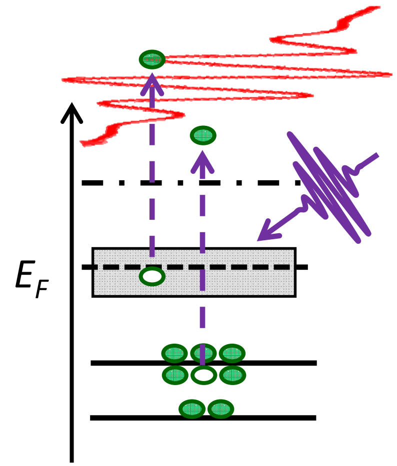
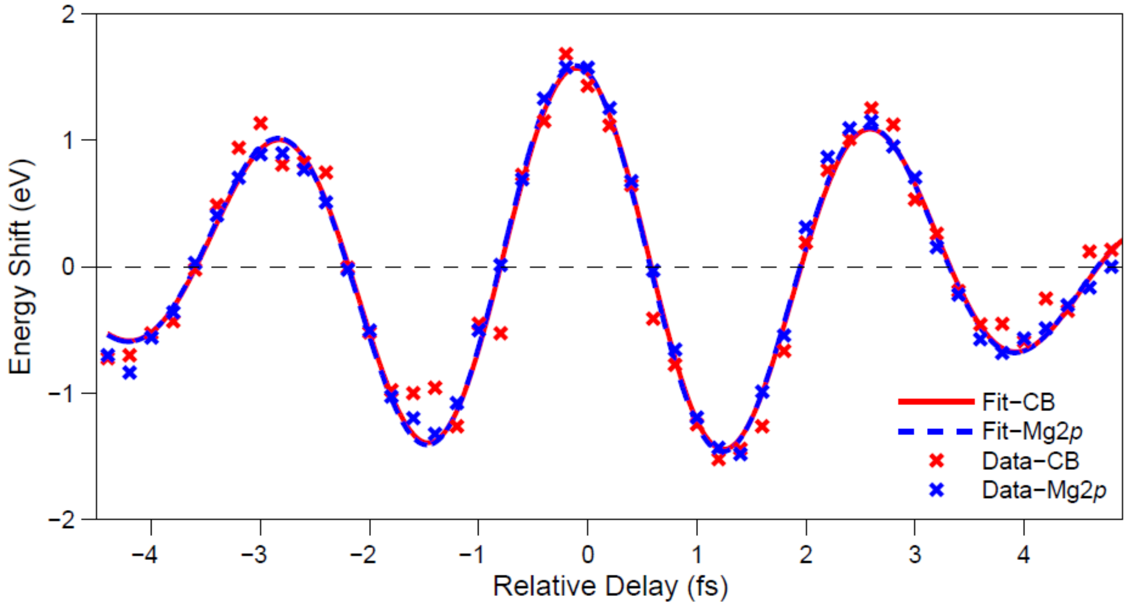




Neppl *et al*, PRL 109, 087401 (2012)

attosecond streaking of Mg(0001)



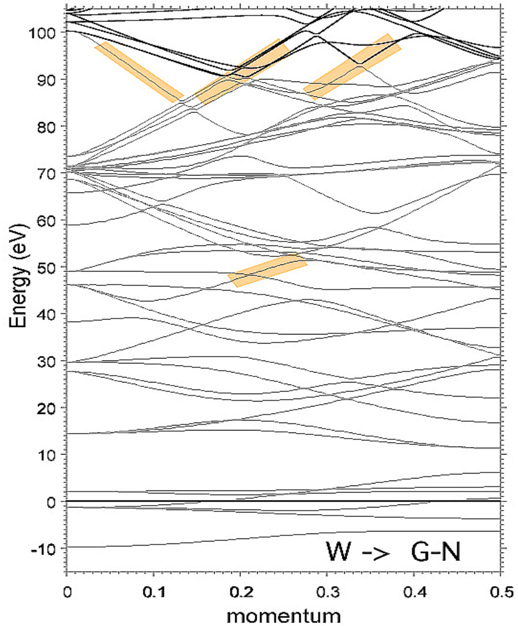
➢Strong discrepancy with theories that emphasize the role of the initial-state localization ($\Delta\tau \rightarrow$ 100as !)

➢ Perfect agreement with the free-electron propagation limit: $\Delta\tau = \lambda_{CB}/v_{CB} - \lambda_{2p}/v_{2p} =$ 0as

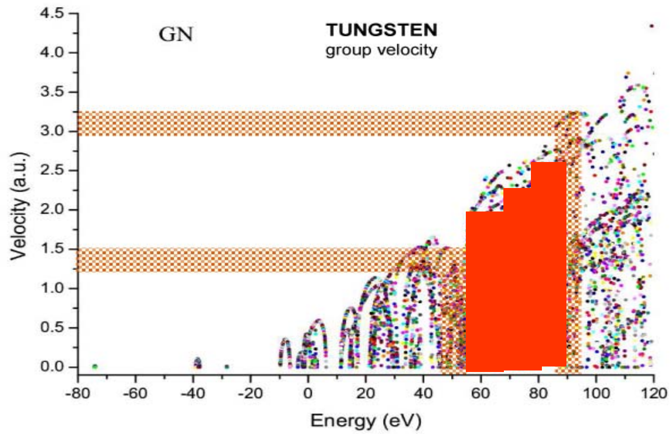
Neppl *et al*, PRL 109, 087401 (2012)

final-state bandstructure in the non-free-electron-like case

calculated W bandstructure

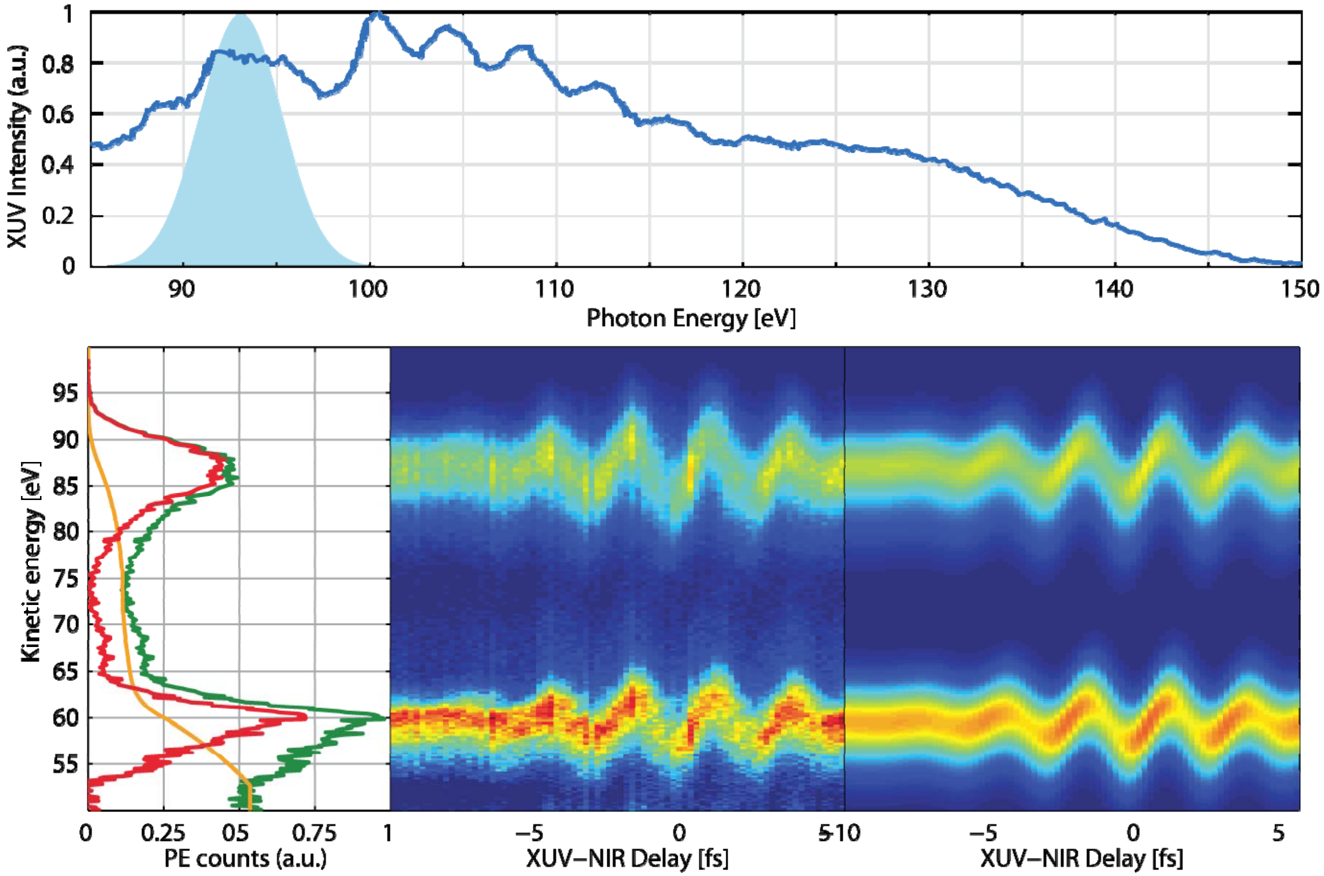


calculated group velocities for hν = 95 eV

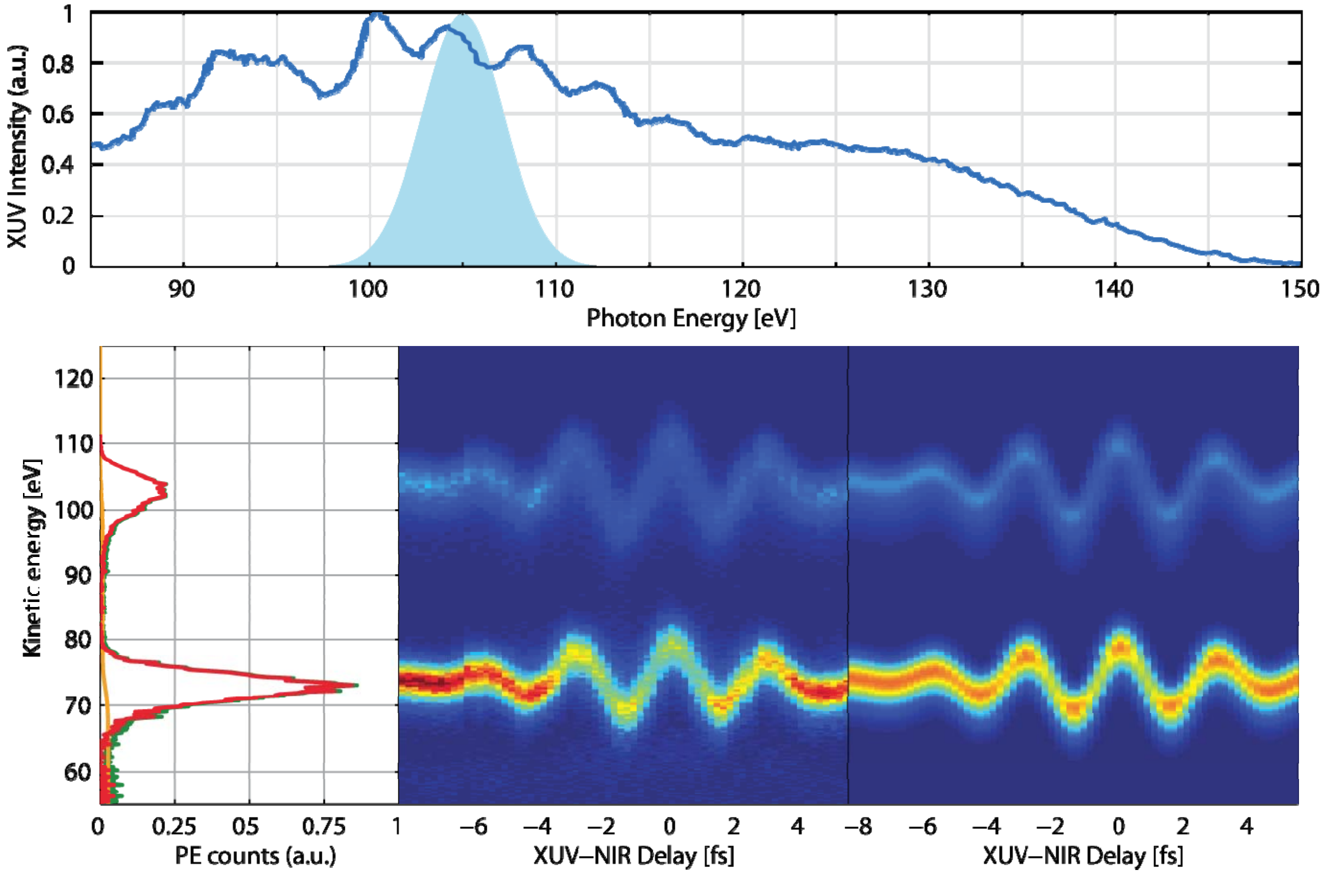


- Conduction-band electrons (from near the Fermi energy) are excited into upper-conduction band states where their group velocity is ~twice that of the excited 4*f*-state electrons

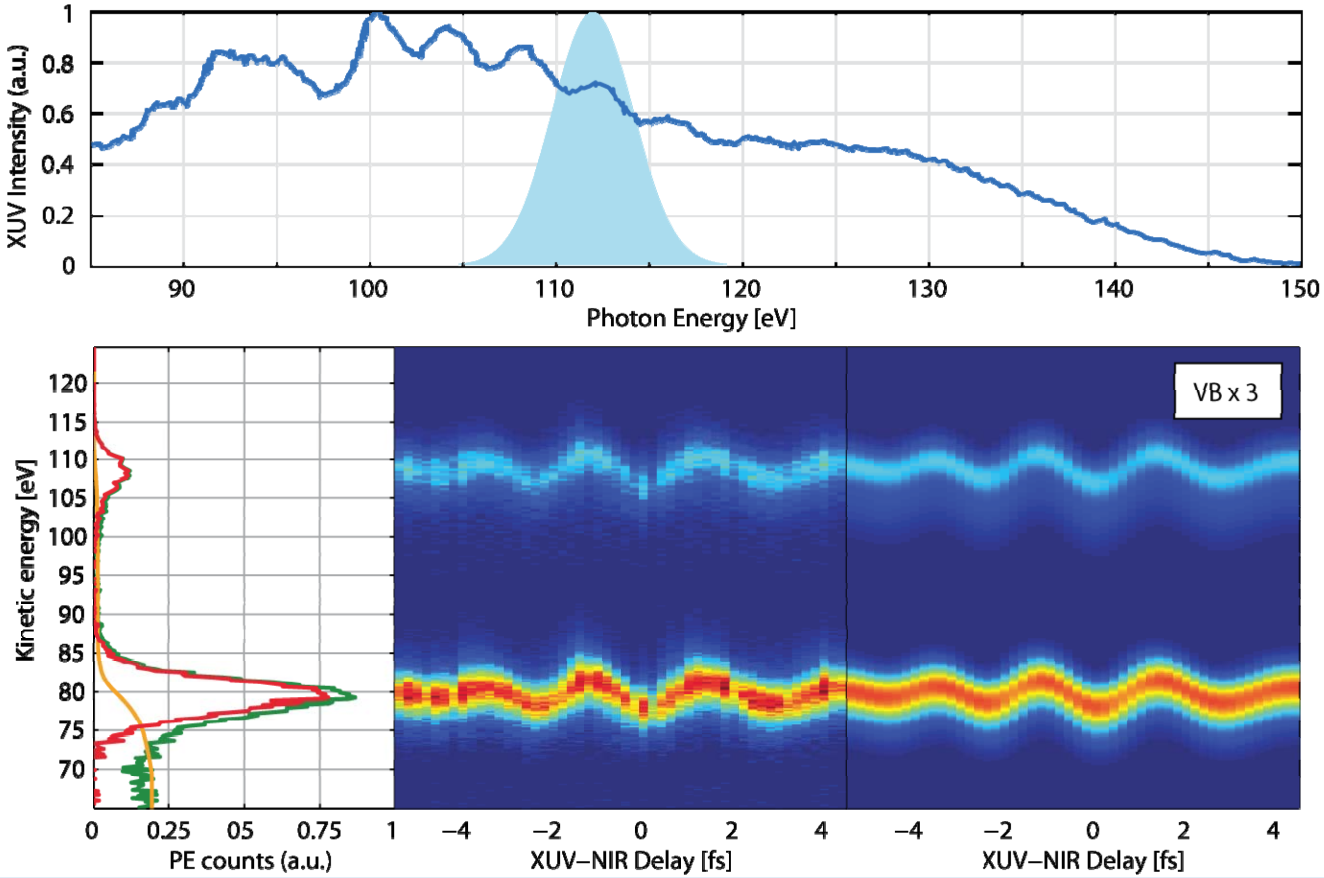
energy dependent *as* streaking spectroscopy



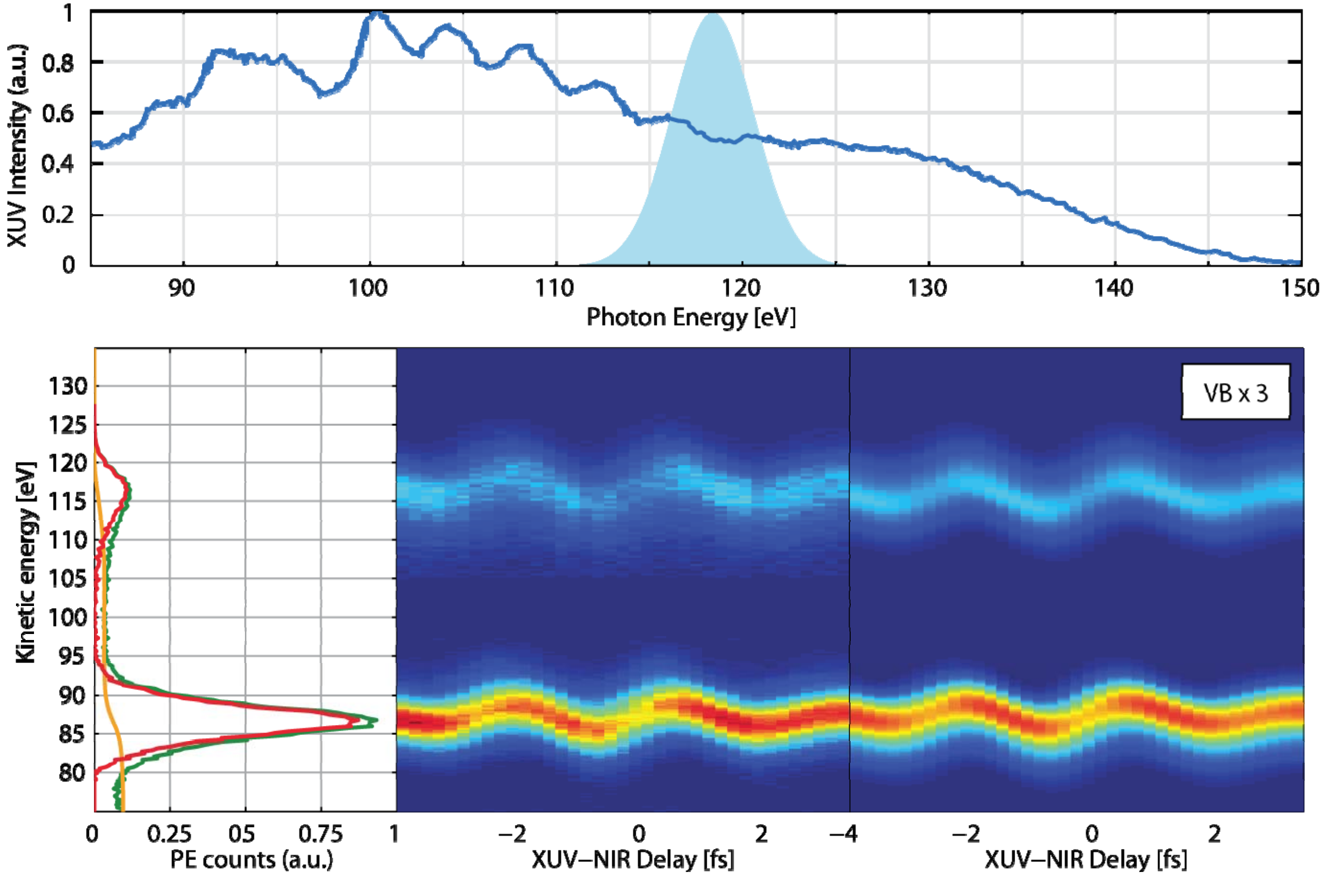
energy dependent *as* streaking spectroscopy



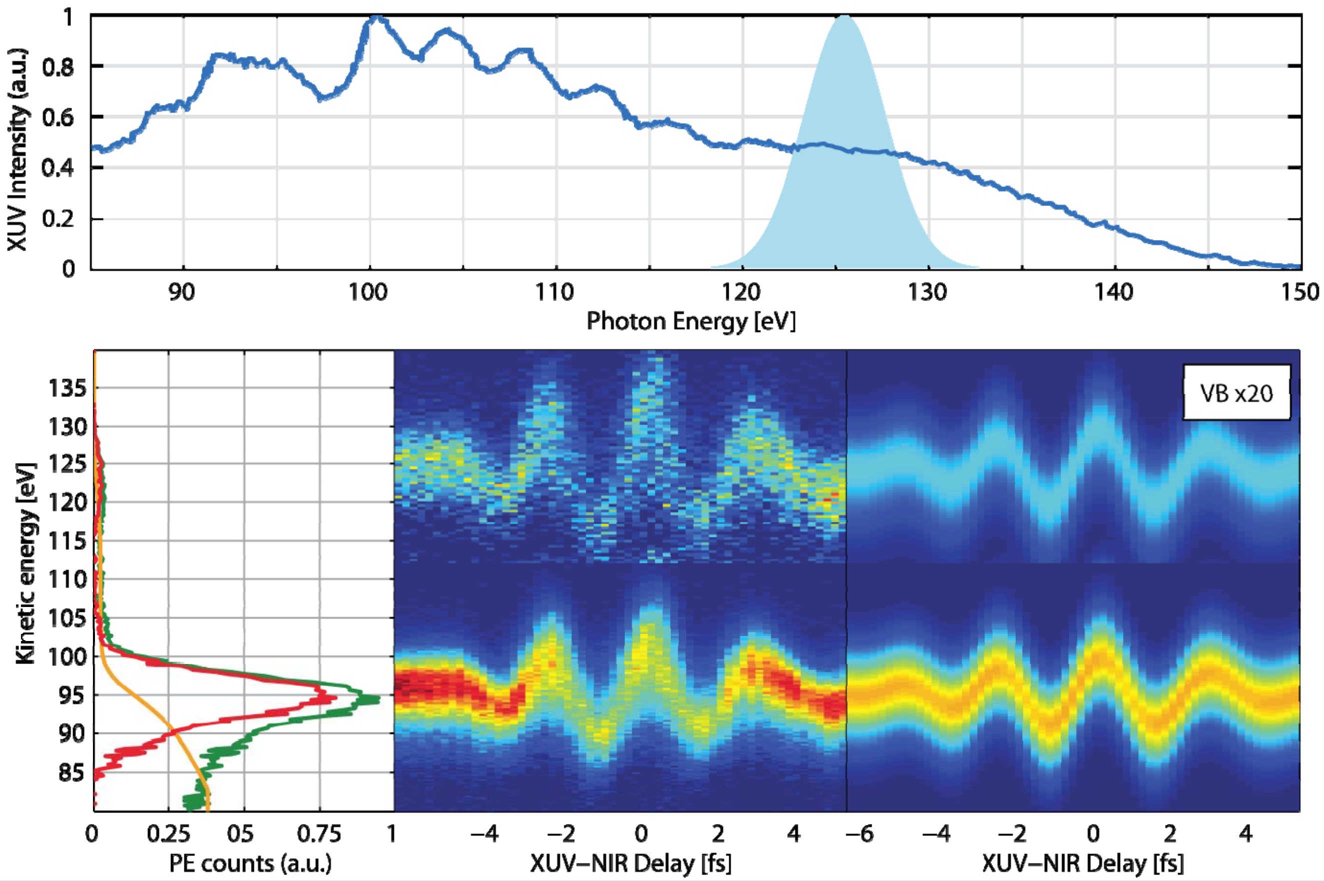
energy dependent *as* streaking spectroscopy



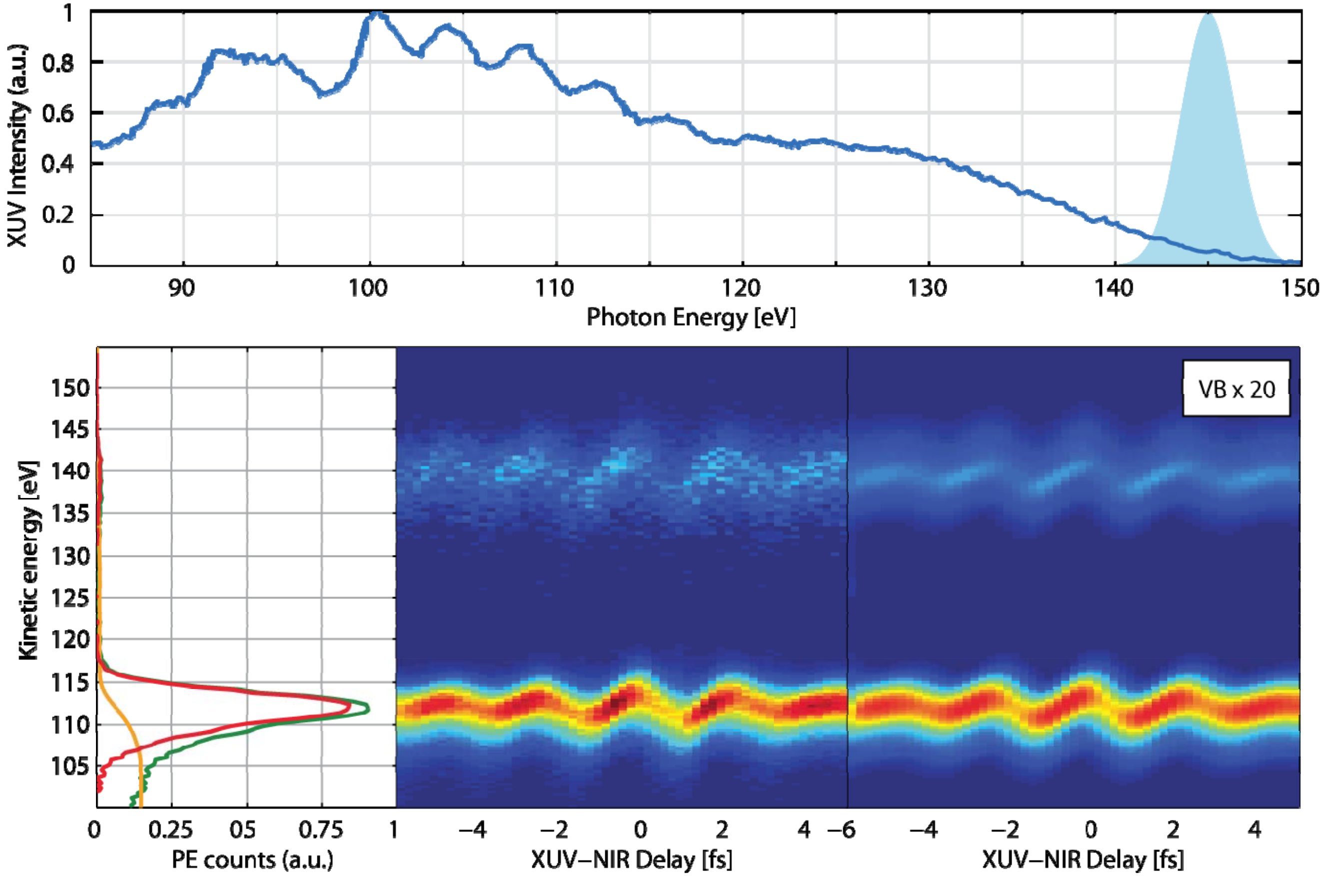
energy dependent *as* streaking spectroscopy



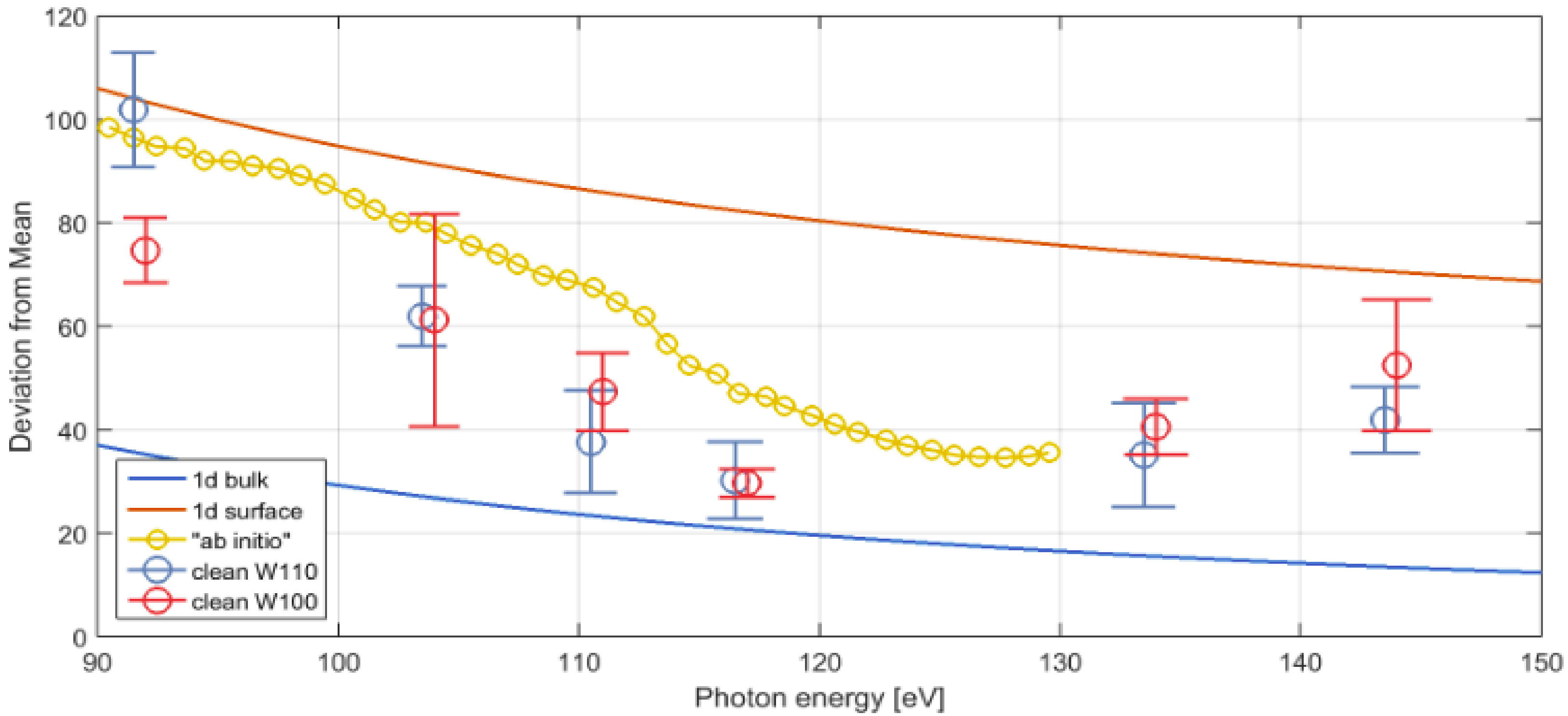
energy dependent *as* streaking spectroscopy

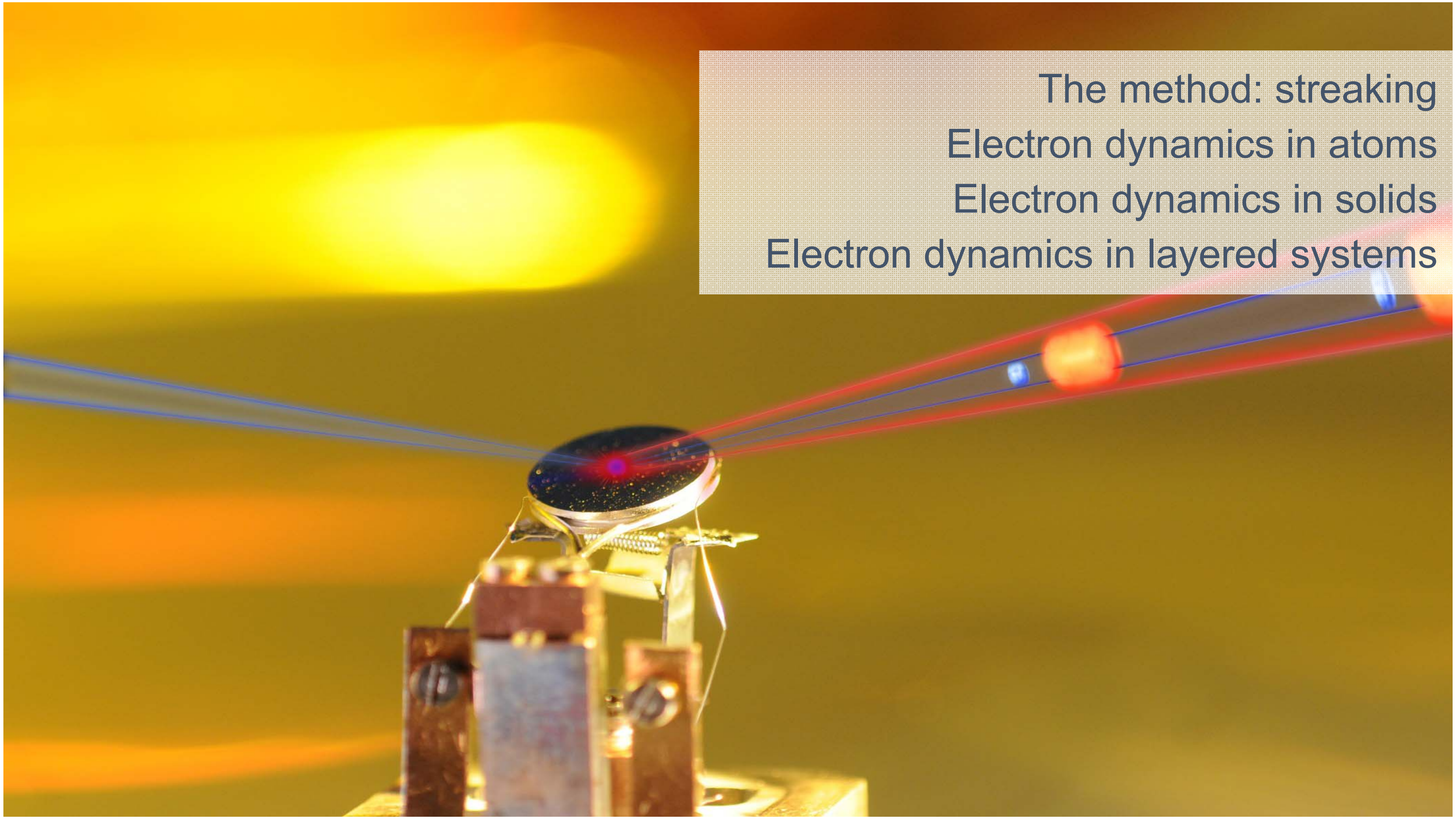


energy dependent *as* streaking spectroscopy



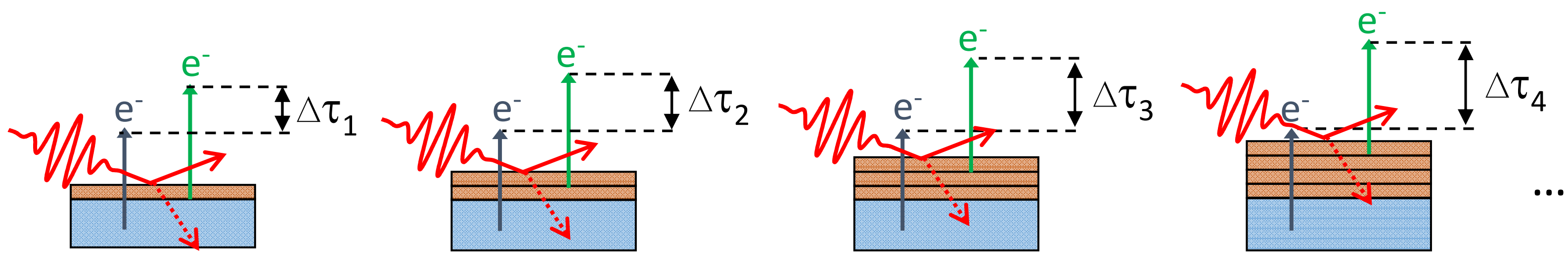
delay: simulation (San Sebastian) vs. experiment




The method: streaking
Electron dynamics in atoms
Electron dynamics in solids
Electron dynamics in layered systems

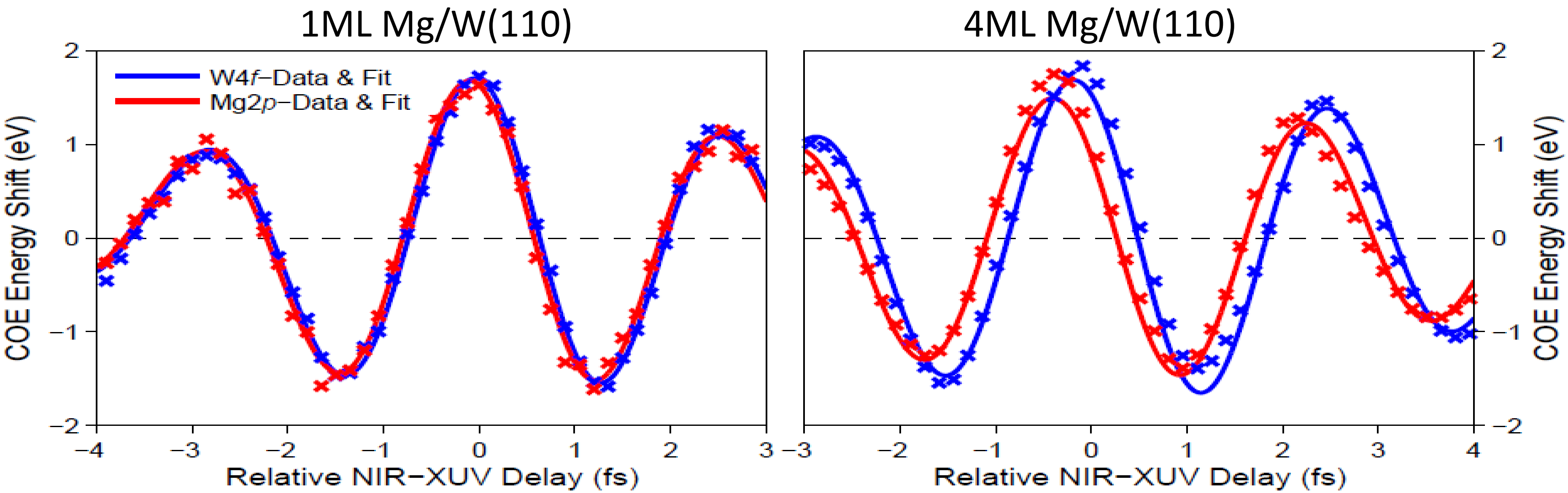
real-time observation of electron propagation

- Tuning the transport experienced by substrate electrons by introducing additional metal overlayers: $\Delta\tau_1 < \Delta\tau_2 < \Delta\tau_3 < \Delta\tau_4$ … ?



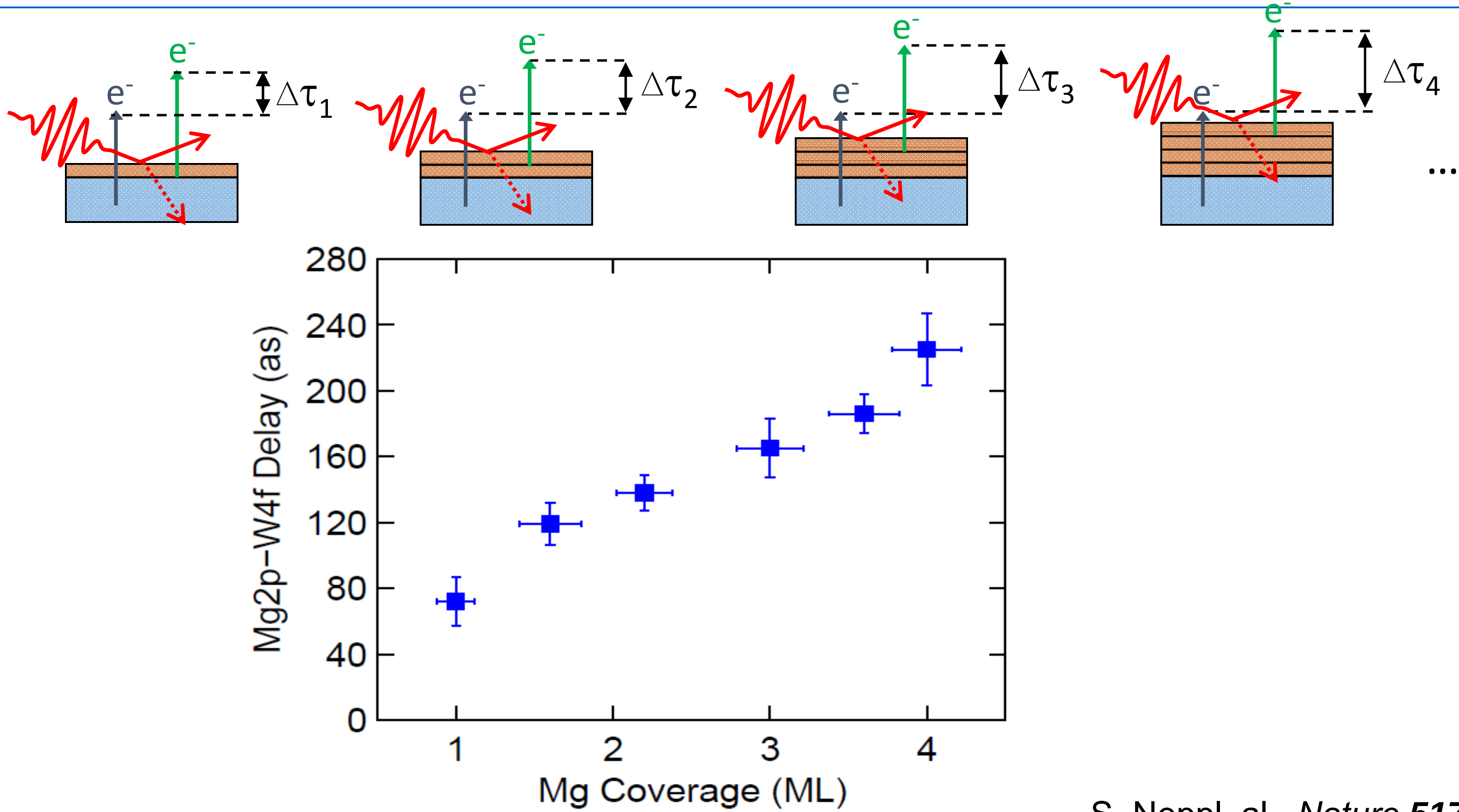
- Investigated system: magnesium layers epitaxially grown on W(110)
- Simultaneous observation of W4*f*, Mg2*p* and CB photoelectrons possible

delay of W4*f* electrons in Mg/W(110)



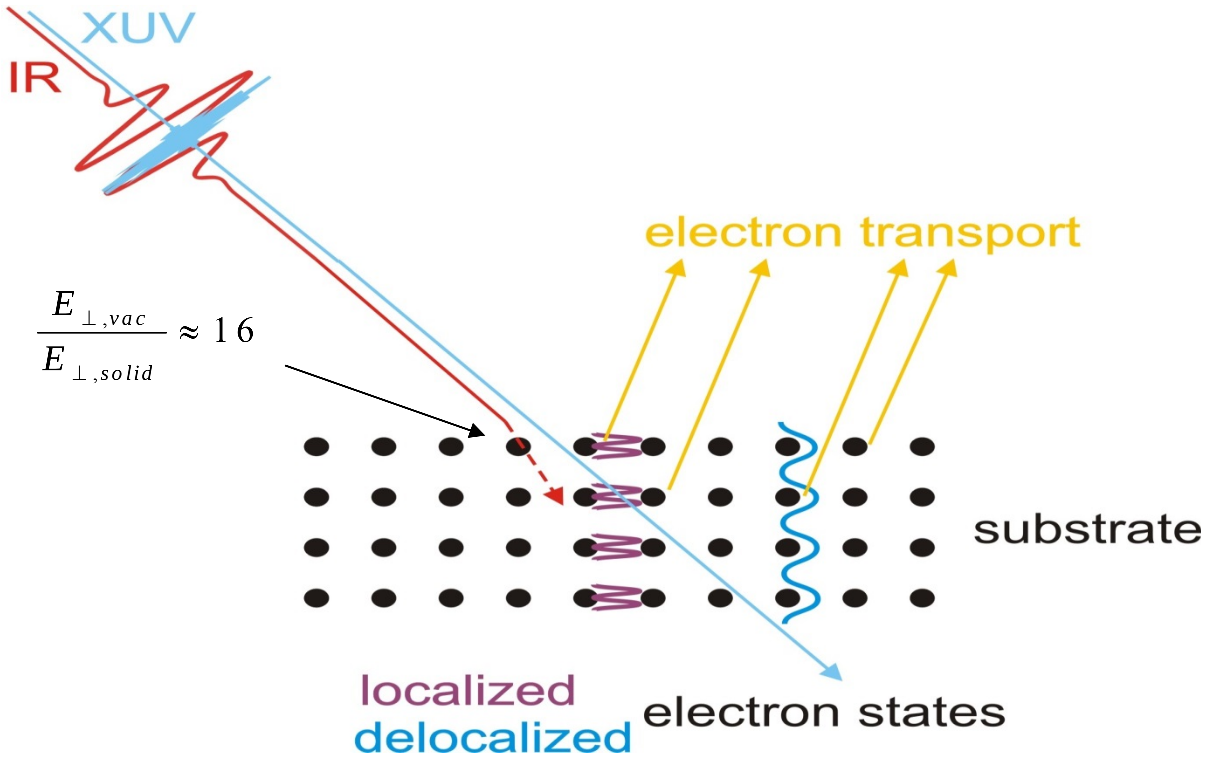
➢ Mg2*p*-W4*f* time delay significantly increases as a function of the Mg overlayer thickness

delay of W4*f* electrons in Mg/W(110)



S. Neppl, al., *Nature* ***517***, 342 (2015)

penetration of the normal field component



assumption: streaking acts outside the solid

propagation model for Mg/W(110)

$$\Delta\tau(d) = \tau_{2p} - \tau_{4f} = \frac{\langle z\rangle_{2p}}{v_{2p}} - \frac{\langle z\rangle_{4f} + d - \delta}{v_{4f}}$$

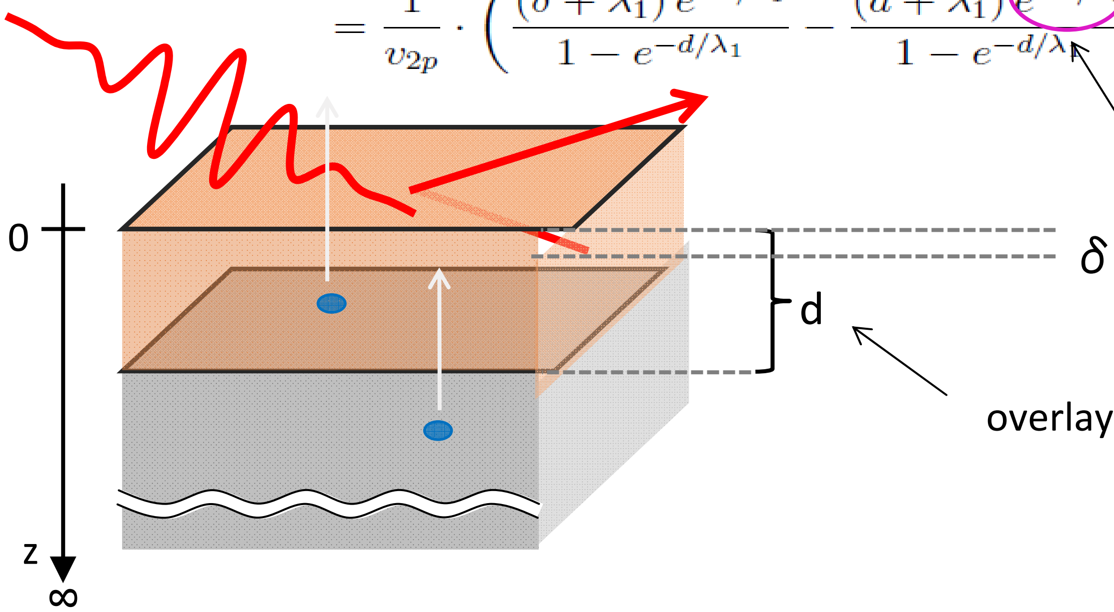
free electron dispersion

$$= \frac{1}{v_{2p}} \cdot \frac{\int_\delta^d z\, e^{-z/\lambda_1}\, dz}{\int_0^d e^{-z/\lambda_1}\, dz} - \frac{1}{v_{4f}} \cdot \left(\frac{\int_d^\infty (z-d)\, e^{-(z-d)/\lambda_2}\, dz}{\int_d^\infty e^{-(z-d)/\lambda_2}\, dz} + d - \delta \right)$$

$$= \frac{1}{v_{2p}} \cdot \left(\frac{(\delta + \lambda_1)\, e^{-\delta/\lambda_1}}{1 - e^{-d/\lambda_1}} - \frac{(d + \lambda_1)\, e^{-d/\lambda_1}}{1 - e^{-d/\lambda_1}} \right) - \frac{\lambda_2 + d - \delta}{v_{4f}}$$

IMFP of W4f electrons in W (3.9Å)

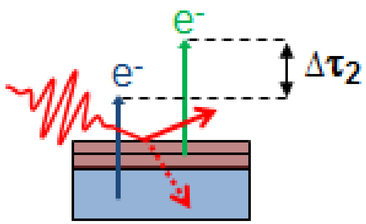
IMFP of Mg2*p* electrons in Mg (3.7Å)

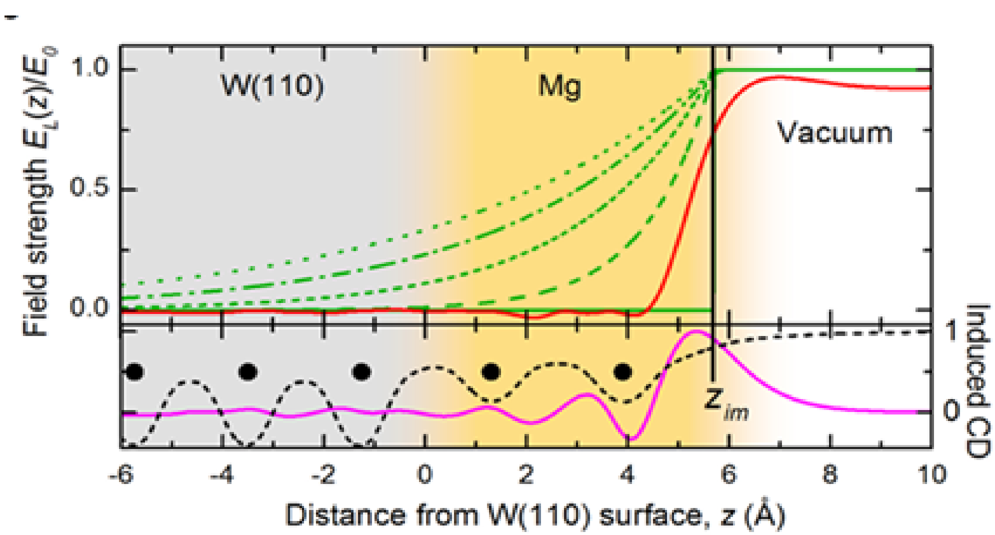


'effective' NIR screening length

overlayer thickness

Delay of W4*f* electrons in Mg/W(110)





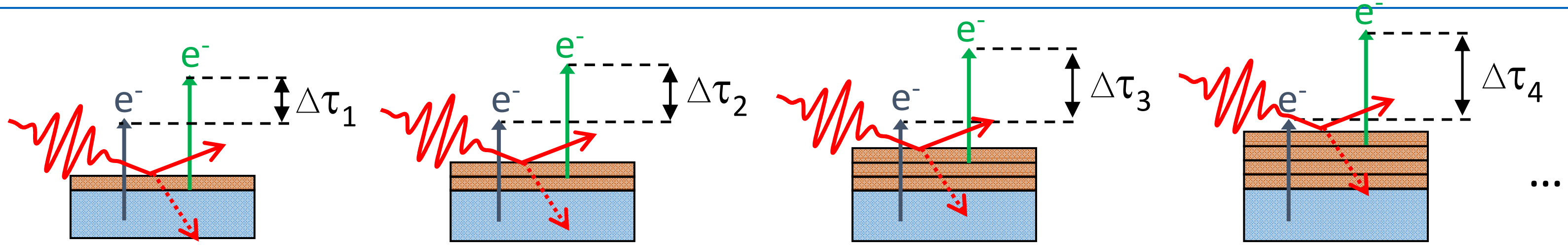
Electric field in 1D calculation
3D calculation

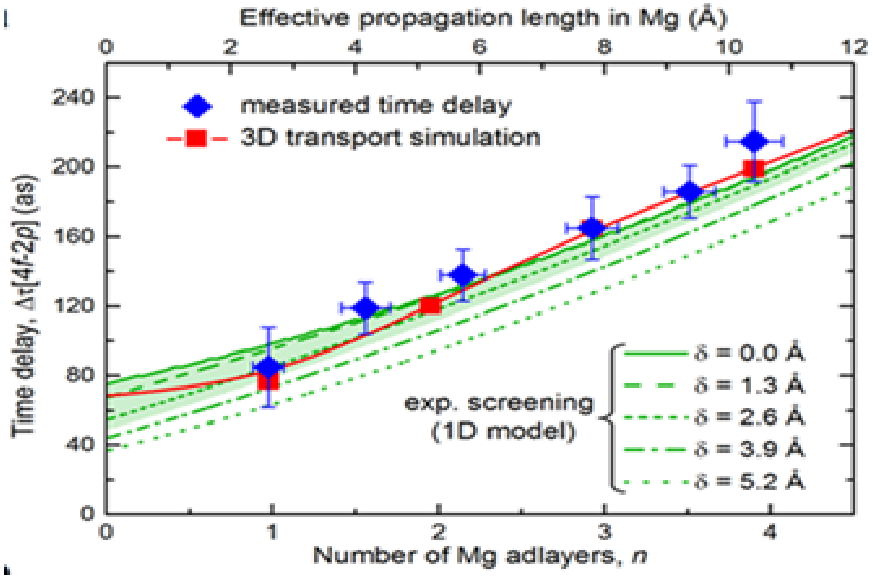
Charge density at maximum of IR pulse

Lattice potential used in DFT calculations

S. Neppl, al., *Nature* ***517***, 342 (2015)

delay of W4*f* electrons in Mg/W(110)



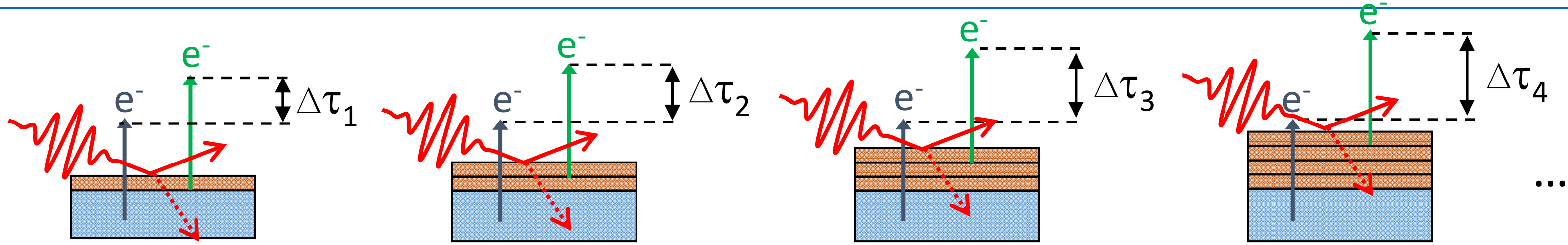


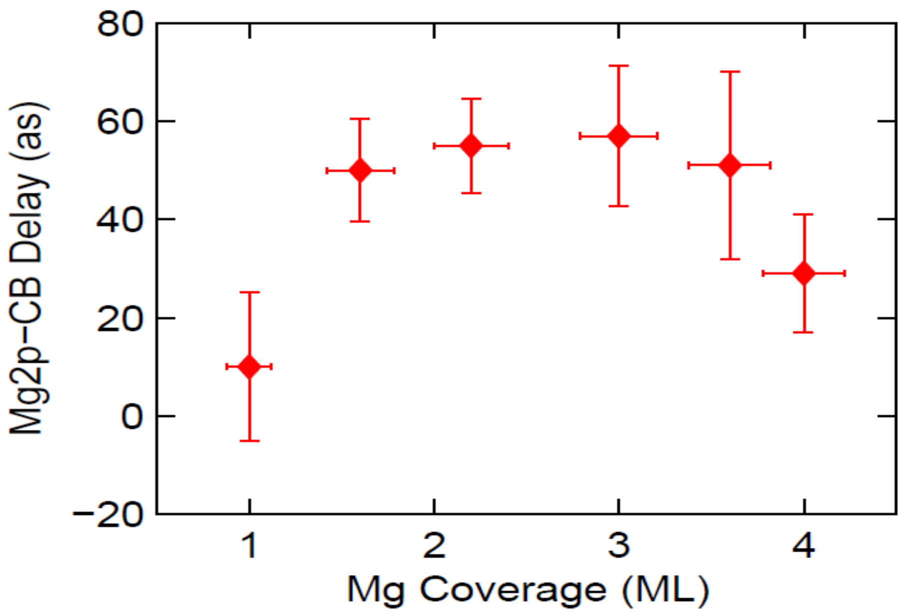
exp. decaying field
& 3D calculations:

screening 1.3 - 2.6 Å

S. Neppl, al., *Nature* ***517***, 342 (2015)

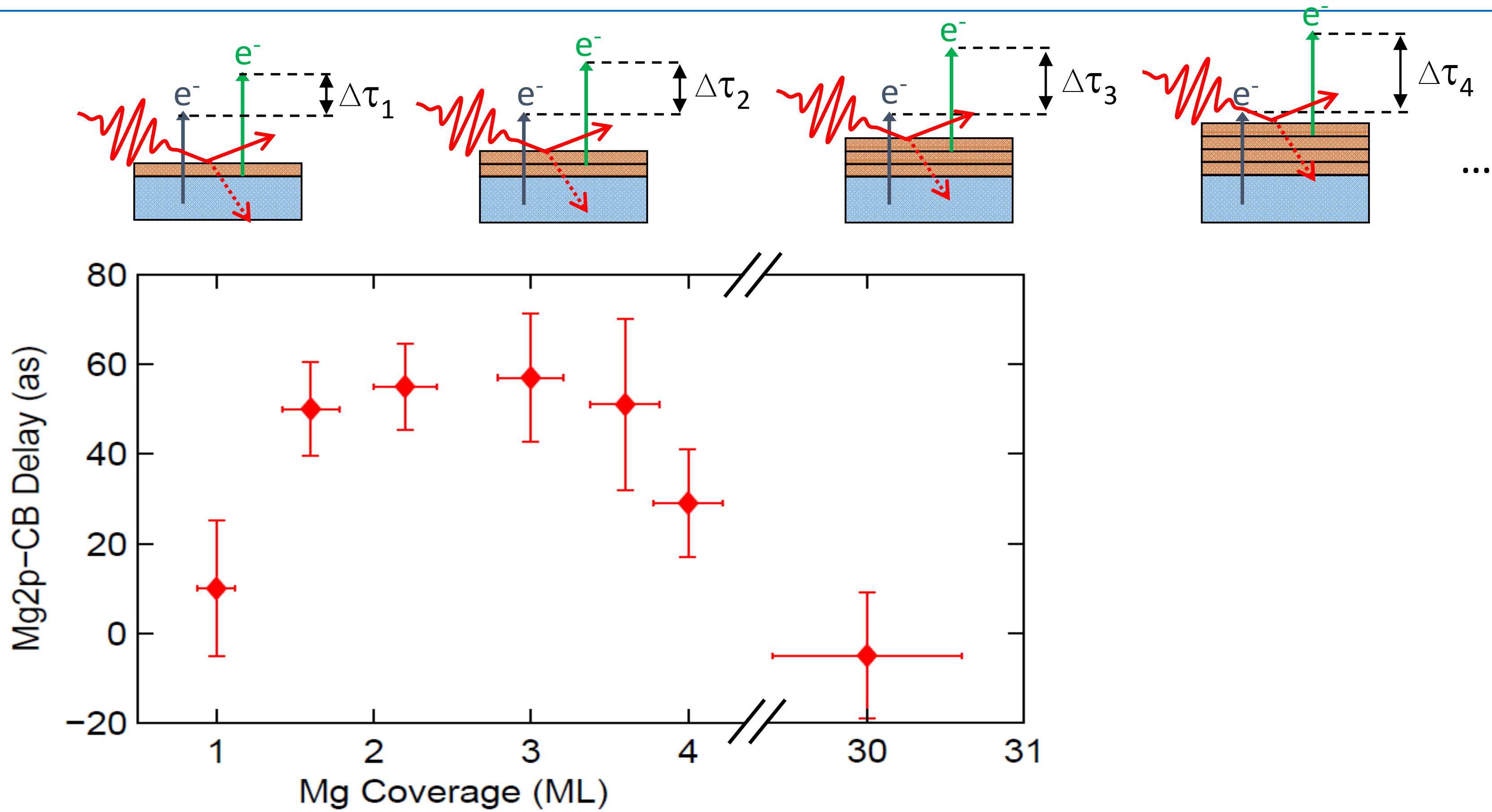
delay of CB electrons in Mg/W(110)



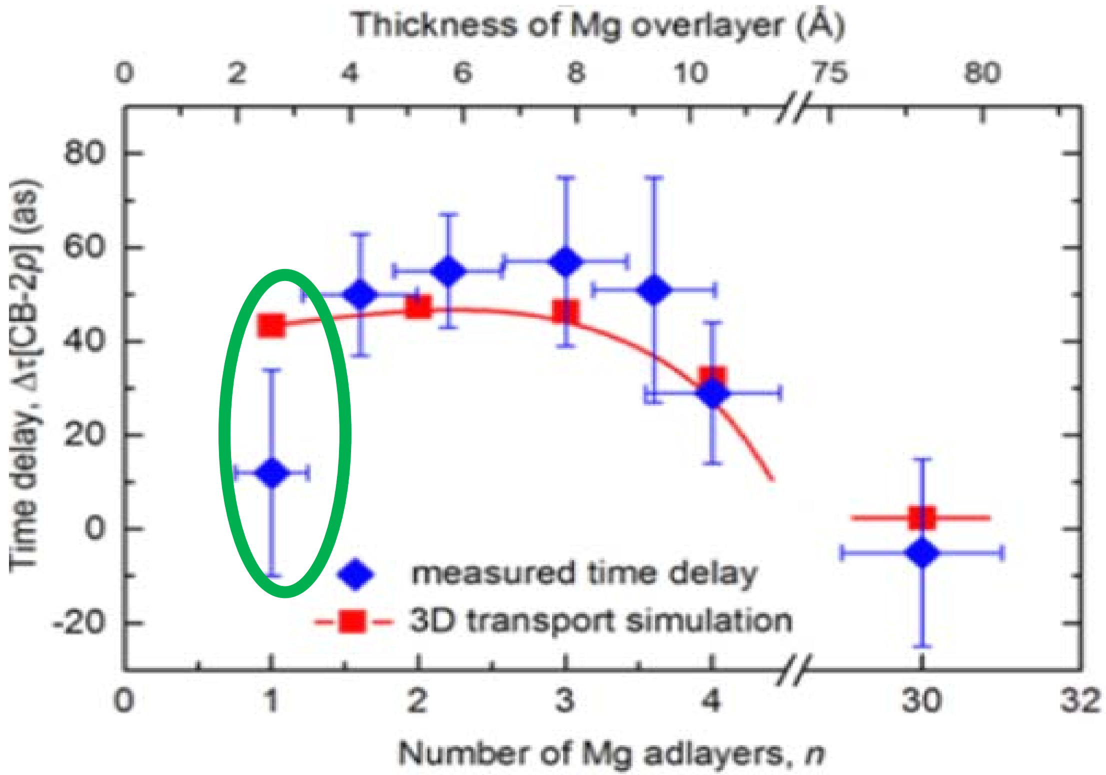


- More complex: superposition of W and Mg CB states
- Delay should converge to bulk Mg(0001) value for increasing coverage

delay of CB electrons in Mg/W(110)

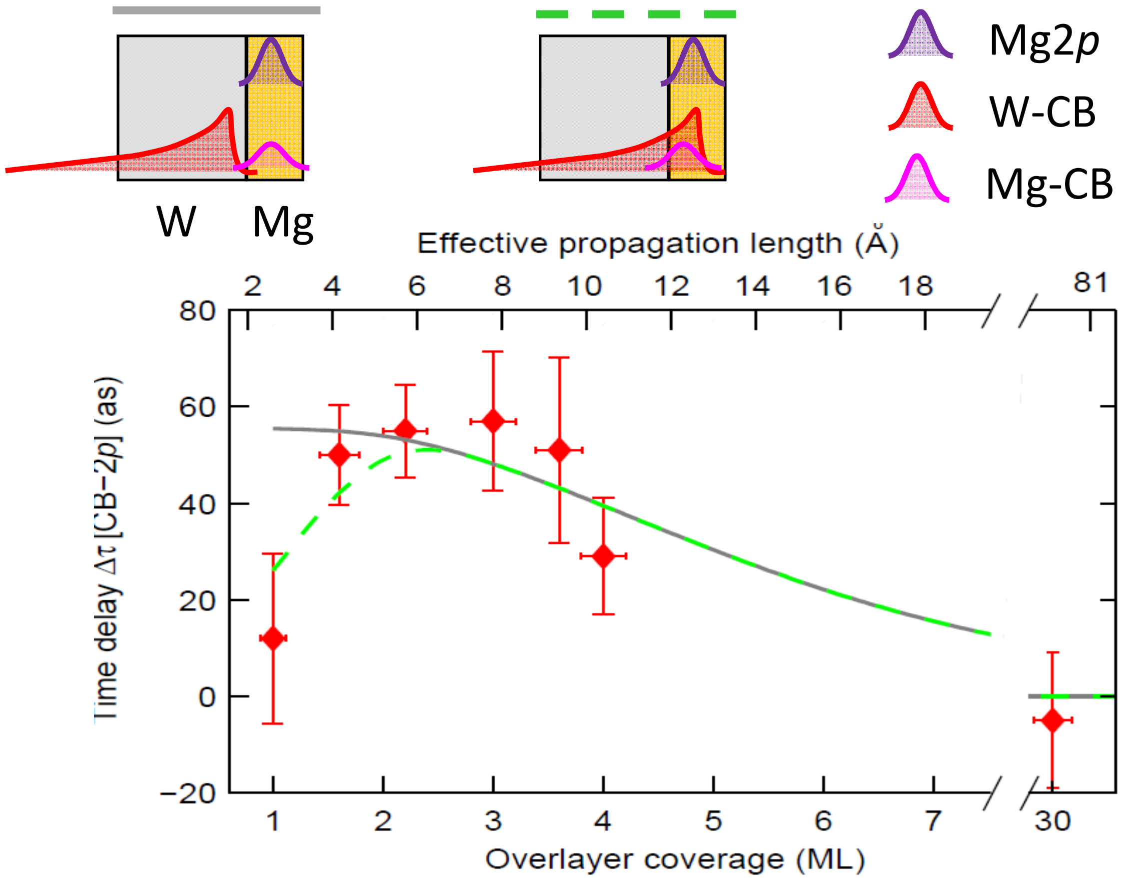


delay of CB electrons in Mg/W(110)
3D transport simulation



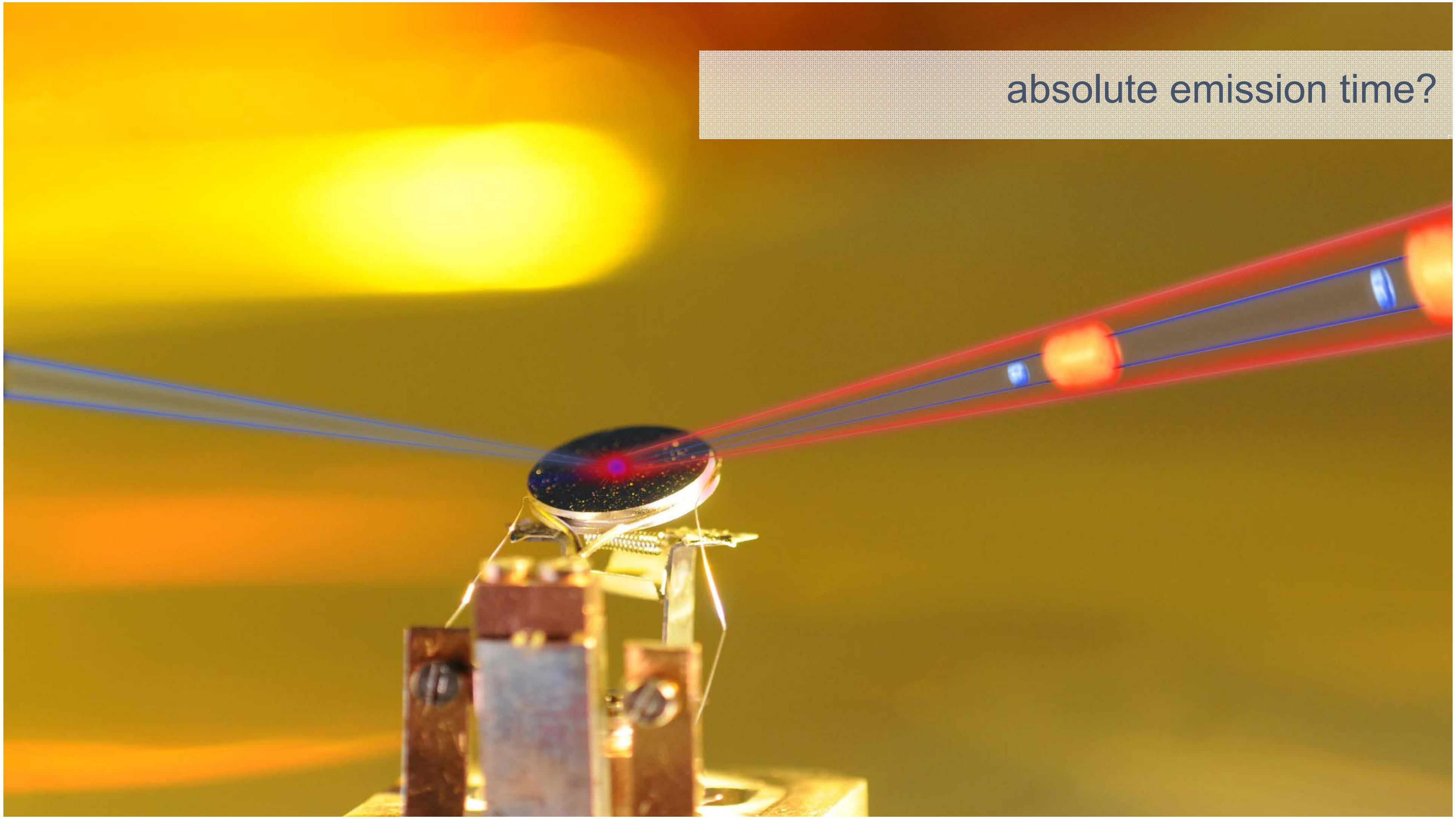
S. Neppl, al., *Nature* ***517***, 342 (2015)

Delay of CB electrons in Mg/W(110)



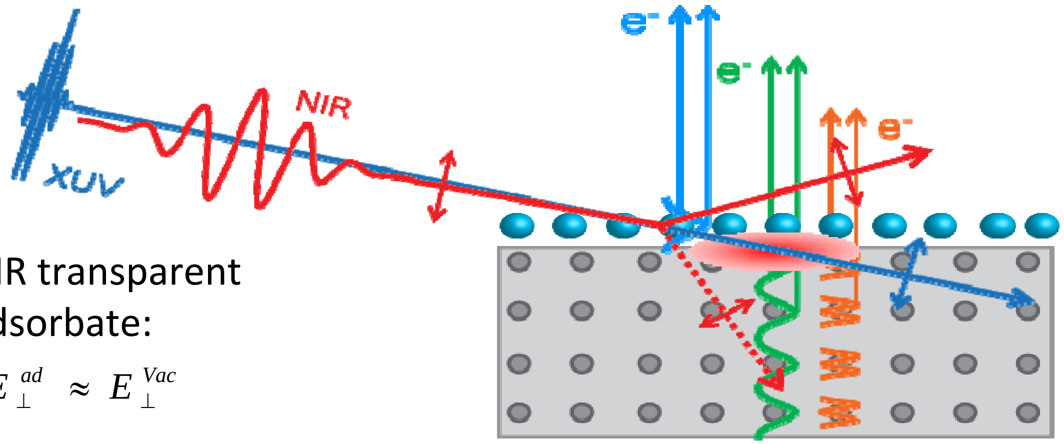
hybridization?

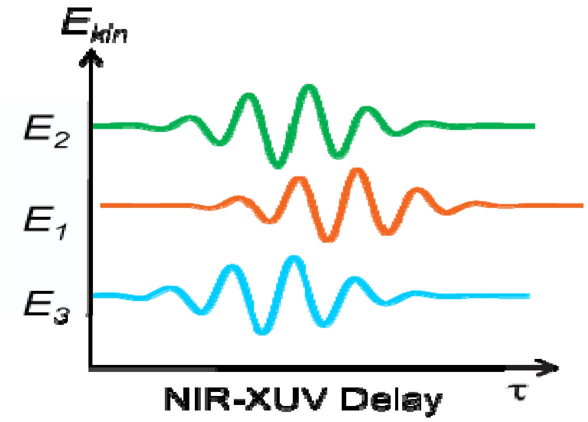
penetration of the W-CB electron wavefunktion into the adlayer


absolute emission time?

What about absolute emission times?
– streaking of adsorbed iodine

The chronoscope / „time-zero-marker“





NIR transparent adsorbate:

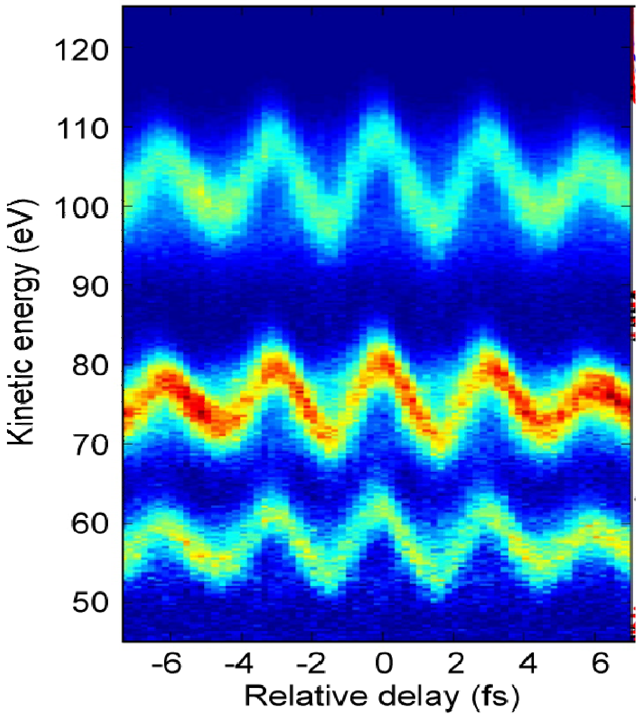
$E_{\perp}^{ad} \approx E_{\perp}^{Vac}$

- Photoelectrons released from the adsorbate immediately respond to the streaking field at the surface
  → time “zero” for clocking substrate emission

streaking experiments on I/W(110)

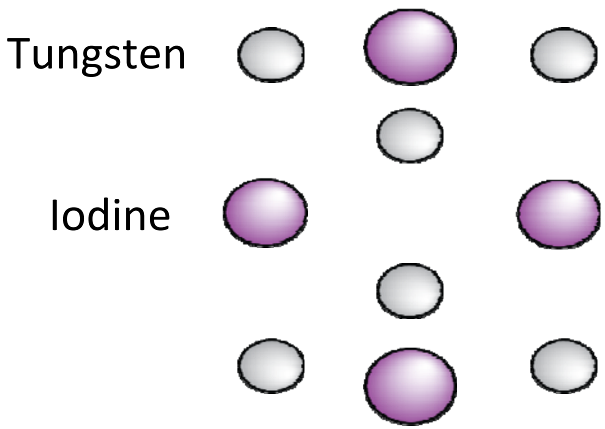
✓ Ideal system for streaking experiments:

- Sufficient energy separation
- Large cross sections
- Ease of sample preparation
- Long-term stable

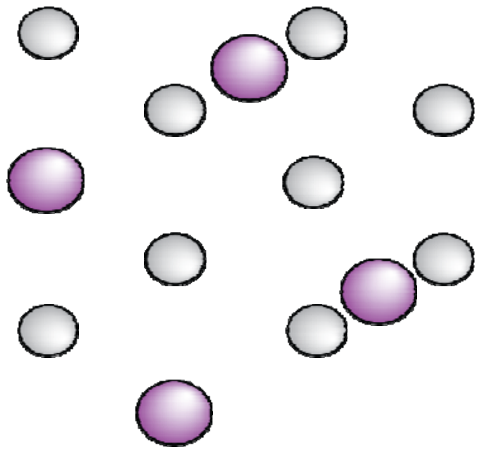


sub-monolayer structures of Iodine/Tungsten (110)

The surface coverage of the I/W(110) adlayer system is varied via thermal desorption after the growth of a saturated atomic monolayer at room temperature.



α-Structure, 1 ML – 0.8 ML



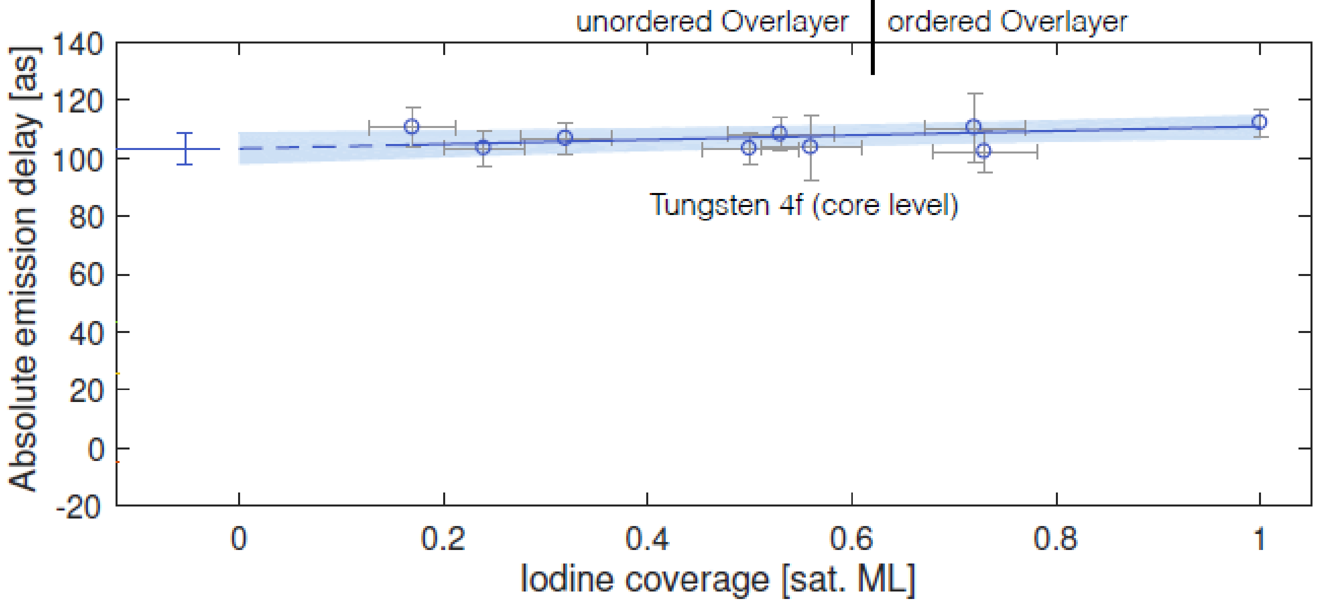
β-Structure, 0.8 ML – 0.5 ML

For a further increase in desorption temperature island formation is expected.

R.G. Jones, **Halogen adsorption on solid surfaces, Progress in Surface Science 27, 112-116, (1988)**

Extrapolation towards zero coverage – the undisturbing chronograph

Even for submonolayer adsorbate coverage residual effects of transport and electric field screening are expected to influence the observed delay → Extrapolation to 0 layers!



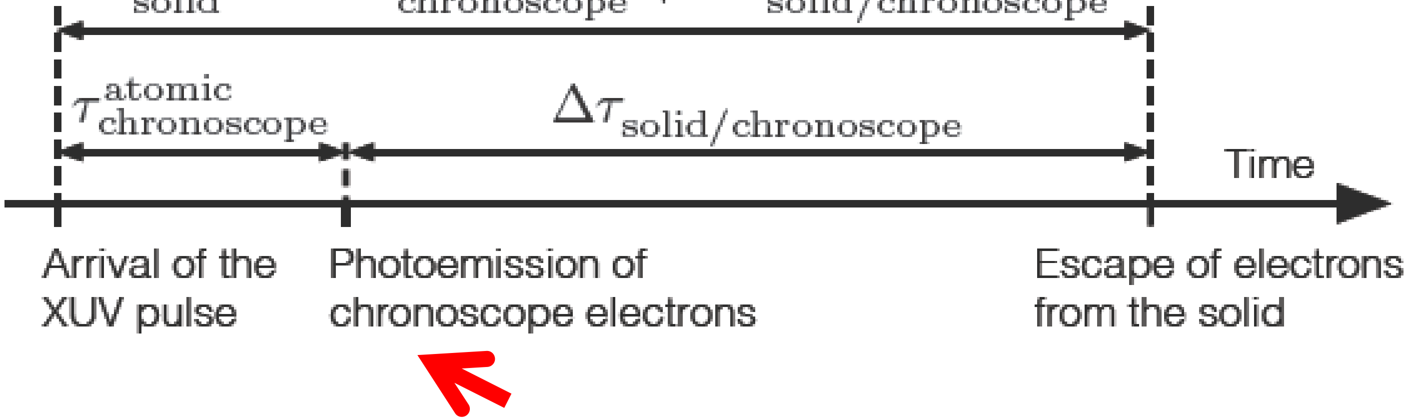
$$\tau_{W4f} = (103 \pm 6)\mathrm{as} + \theta(8 \pm 7)\mathrm{as}$$

Coverage in ML

Extrapolation towards zero coverage – the undisturbing chronograph

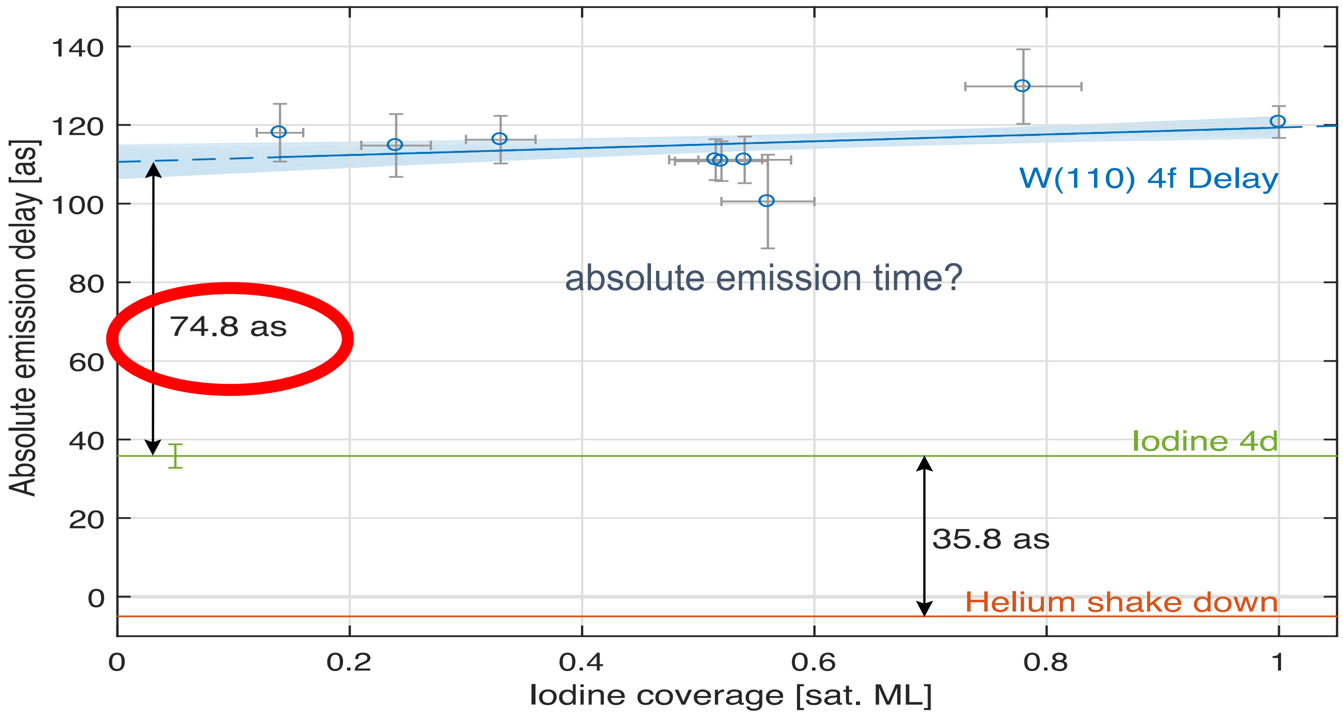
$$\tau_{W4f} = (103 \pm 6)\mathrm{as} + \theta(8 \pm 7)\mathrm{as}$$

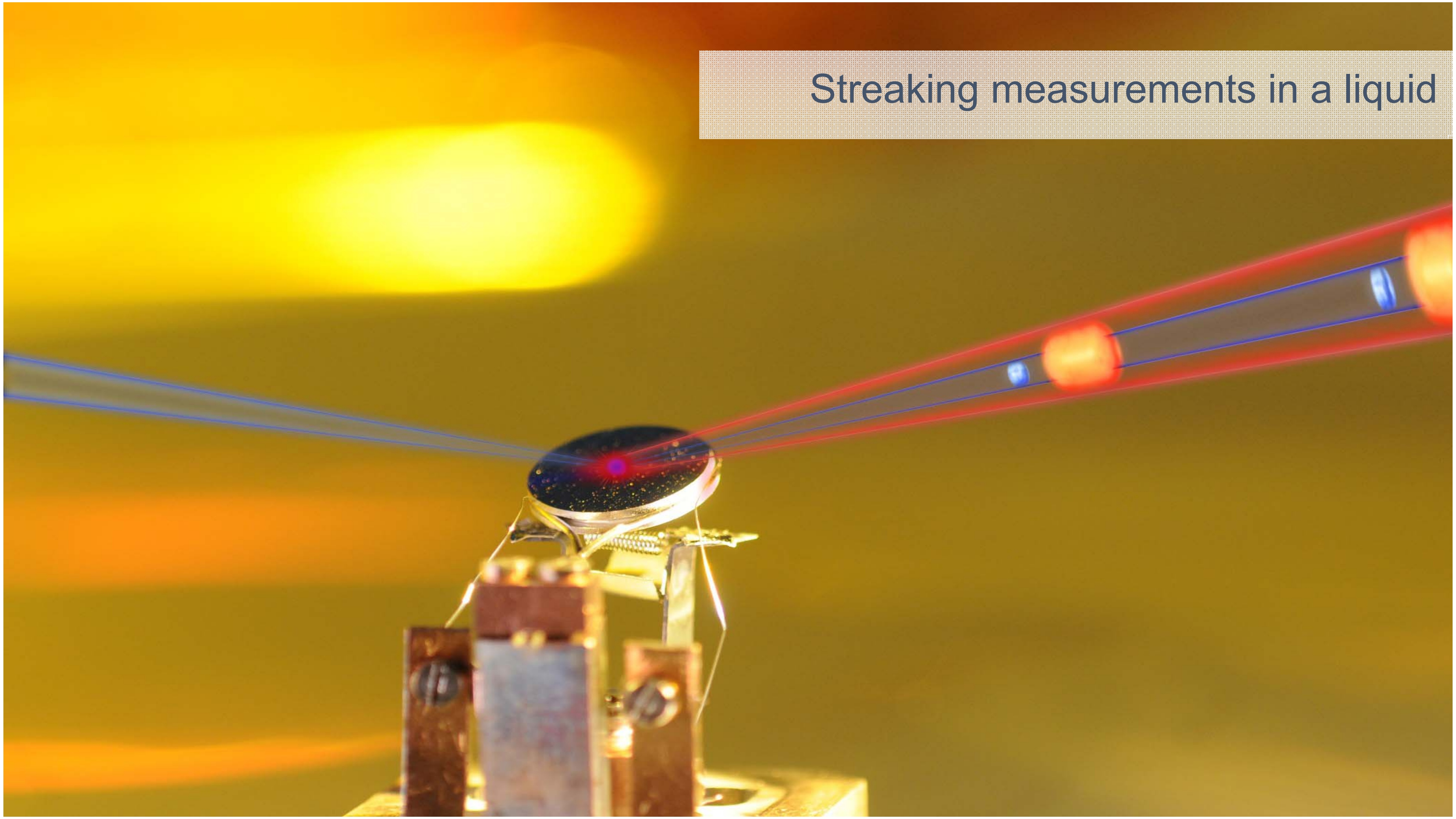
$$\tau_{\mathrm{solid}}^{\mathrm{absolute}} = \tau_{\mathrm{chronoscope}}^{\mathrm{atomic}} + \Delta\tau_{\mathrm{solid/chronoscope}}$$



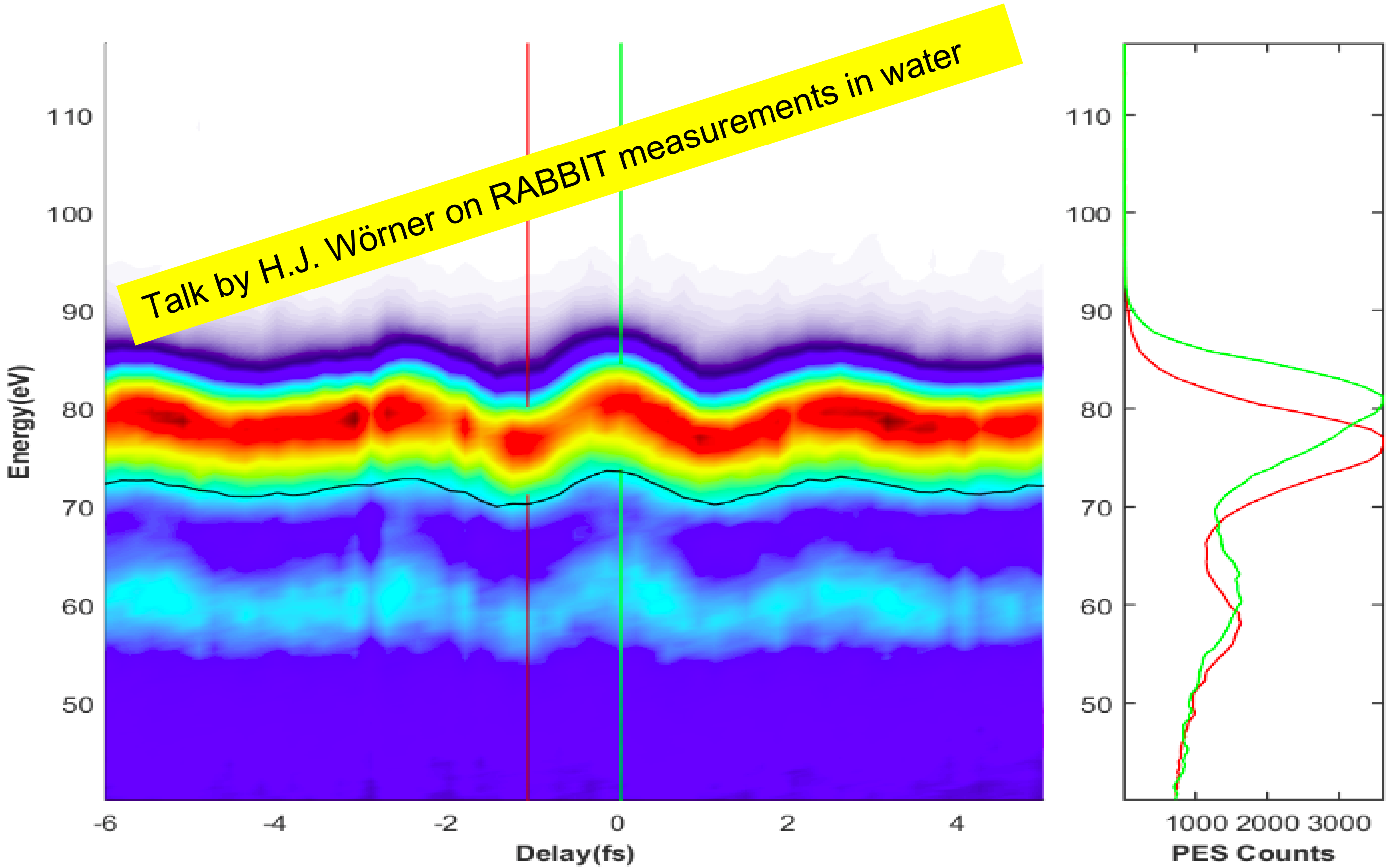
Atomic iodine/helium….

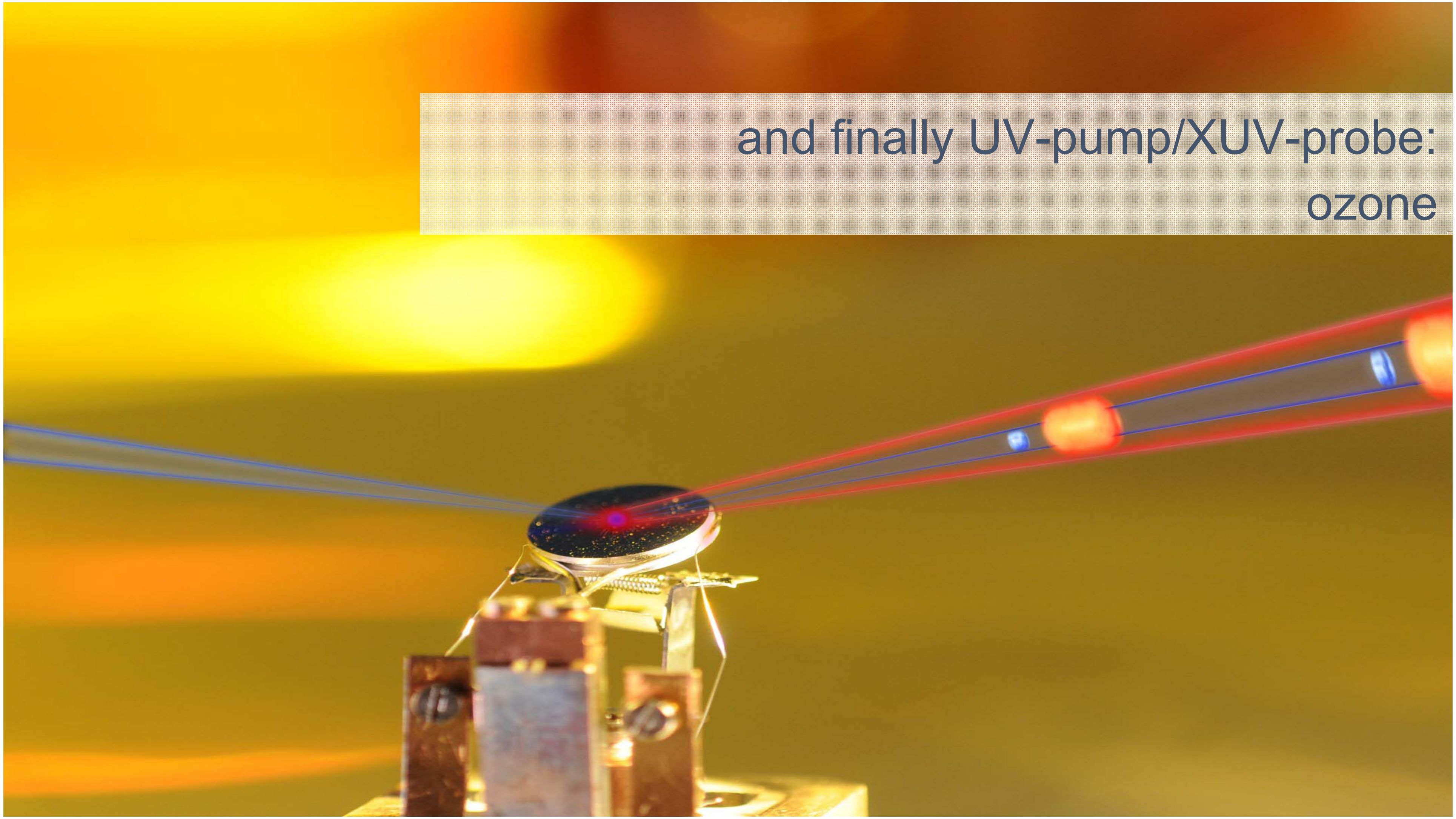
absolute timing of photoemission from surfaces




Streaking measurements in a liquid

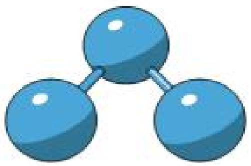
First streaking spectrogram of H_2O



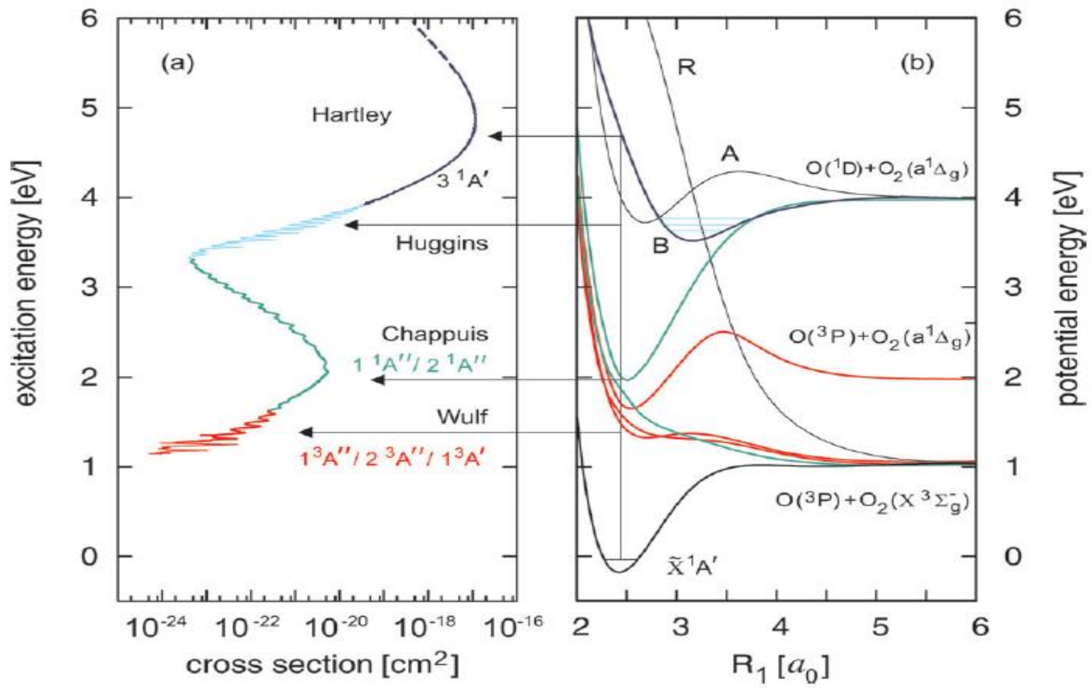

and finally UV-pump/XUV-probe:
ozone

Exciting an electron wavepacket in a molecule
UV+NIR pump / XUV probe

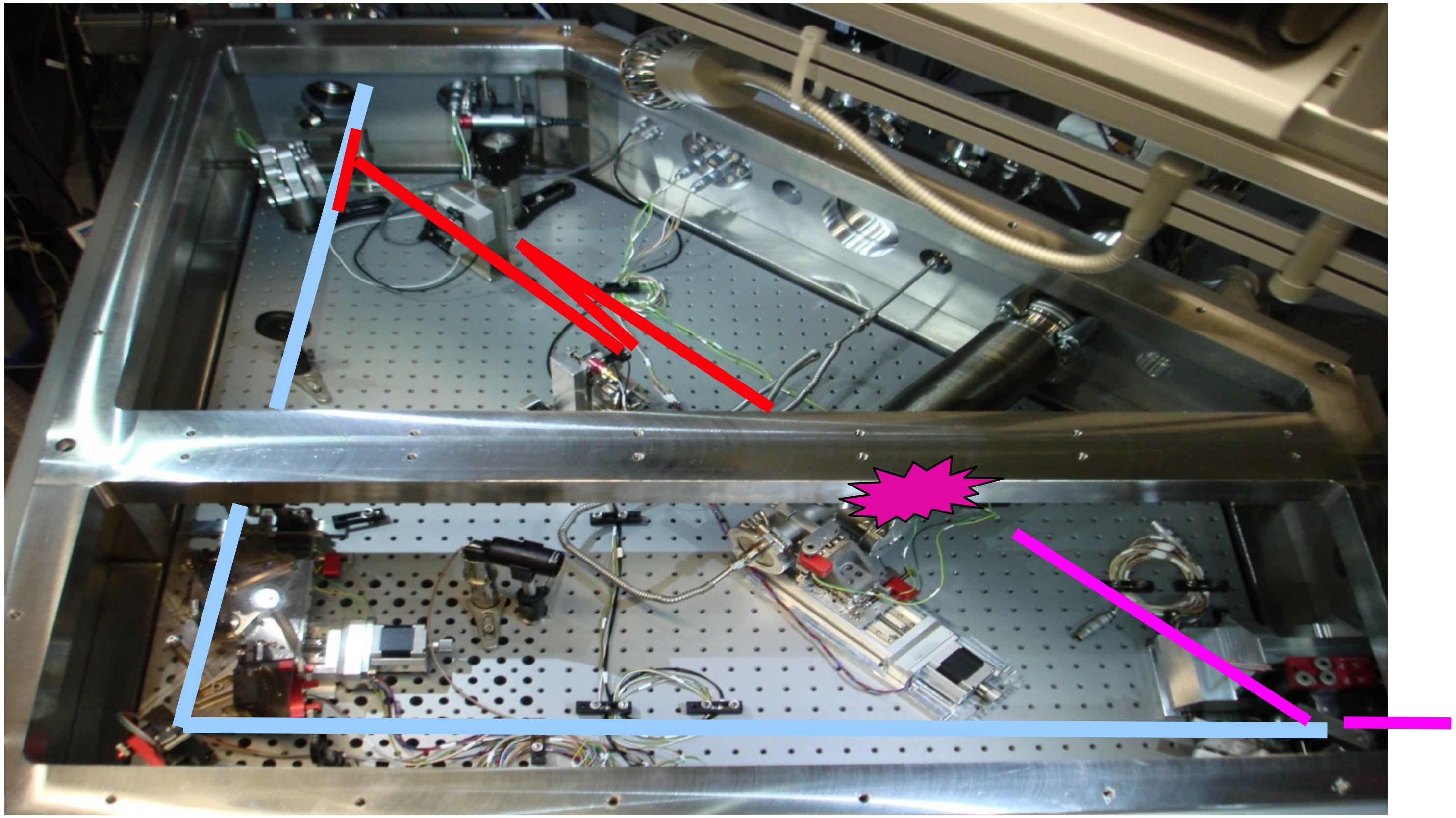
Sampling the evolution with XUV as-pulses



ozone



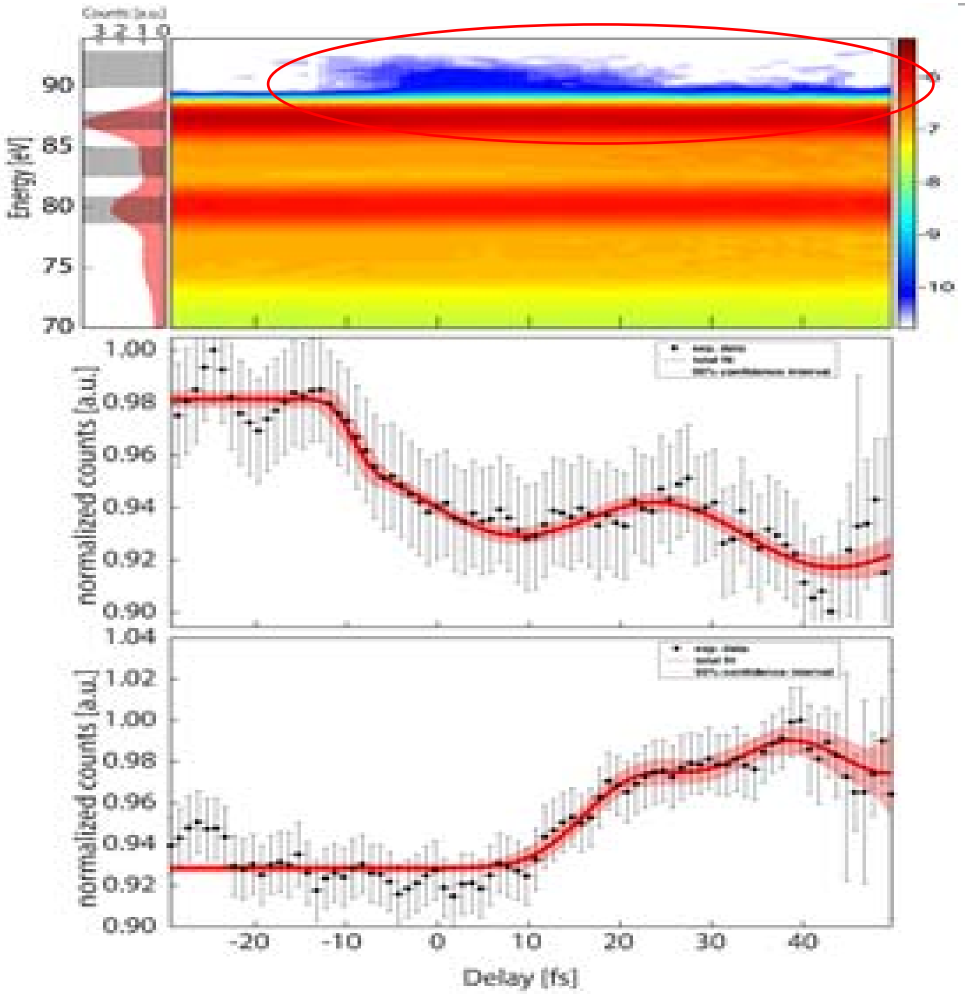
delay & UV-generation



DUV-pump/XUV-probe spectrogramm

ground state

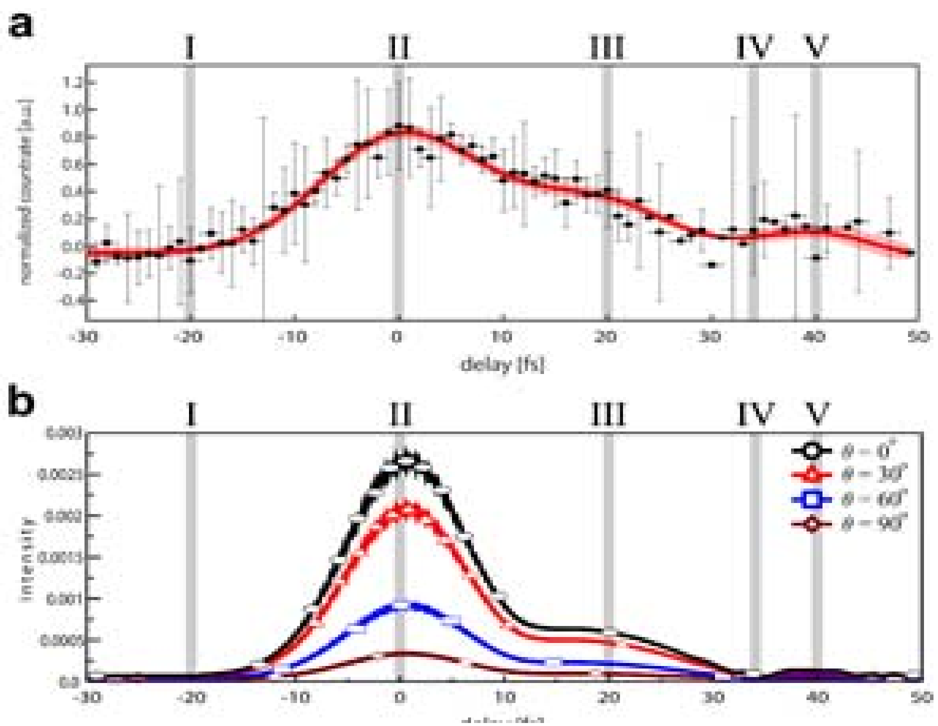
O_2…dissociation



Revival of the excited state

measured

theory



Colleagues & Cooperations

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	R. Ernstorfer, FHI Berlin, Germany
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	Vienna University of Technology, Austria
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	Ludwig Maximilians Univ. Munich, Germany
	P. Echenique, A. Kazansky, E. Krasowski, San Sebastian, Spain
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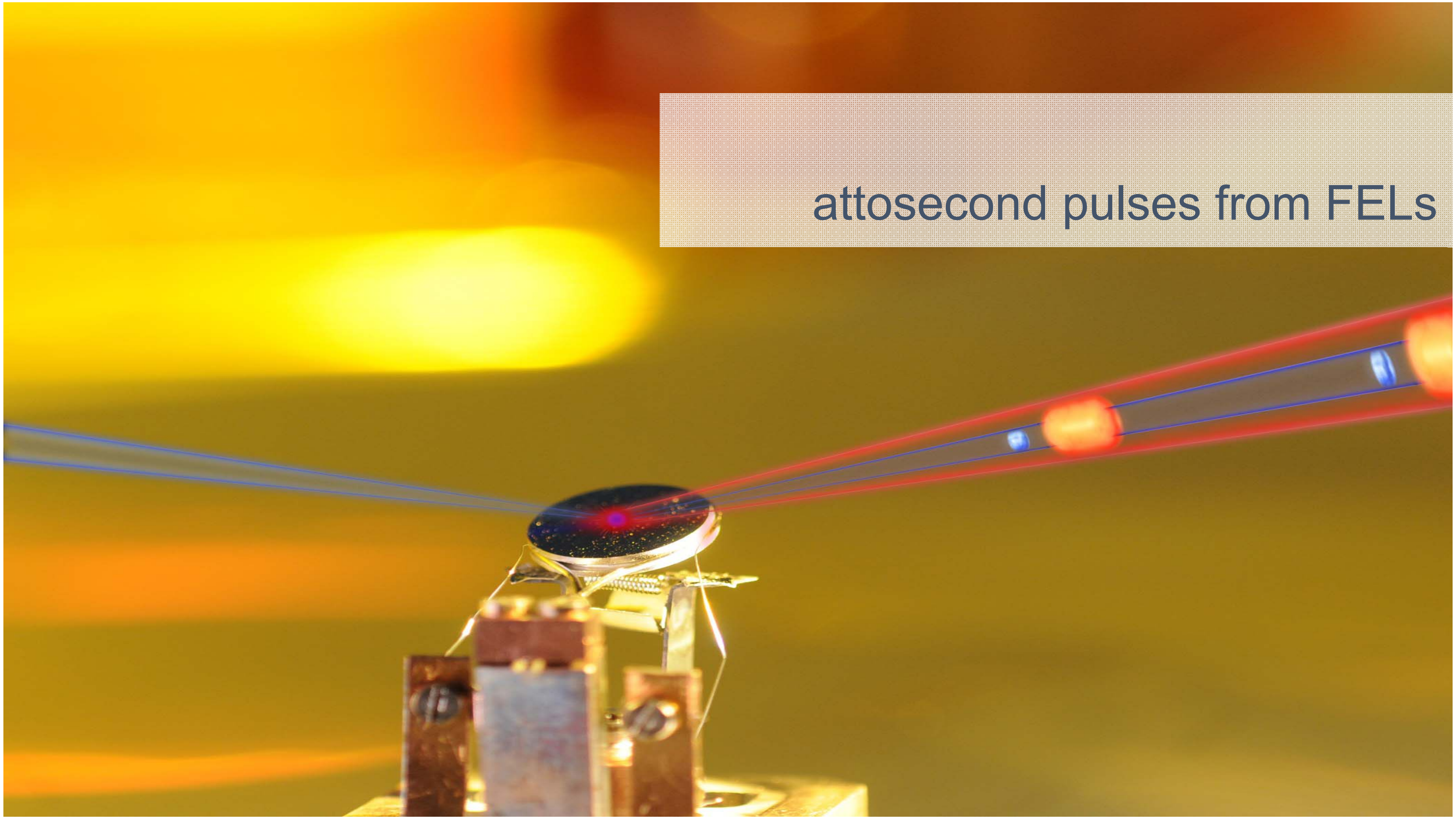
	Ludwig Maximilians Univ. Munich, Germany


Thanks for your attention


attosecond pulses from FELs