Attosecond electron dynamics on solid surfaces - new techniques, new information, - new questions

Reinhard Kienberger

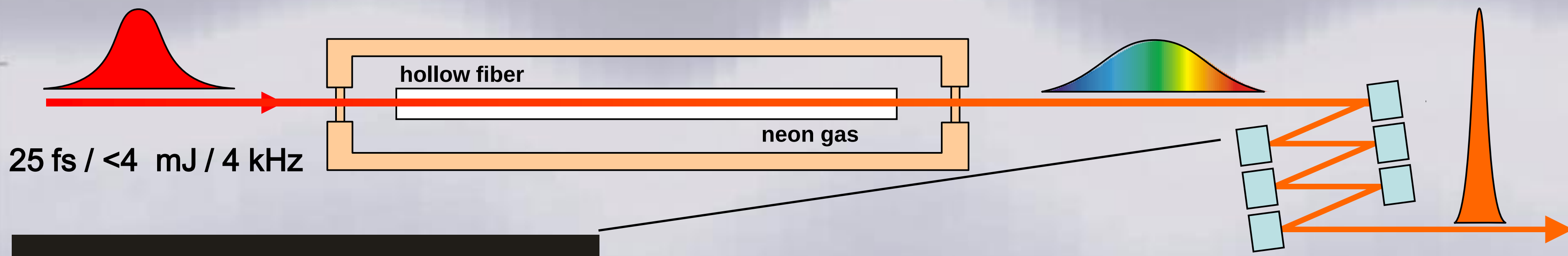
Technische Universität München
Max-Planck Institut für Quantenoptik, Garching

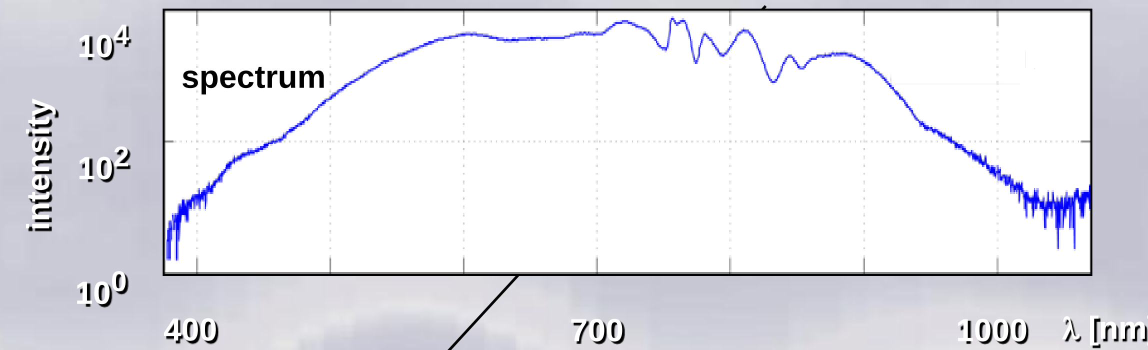
TECHNISCHE UNIVERSITÄT MÜNCHEN
MPQ

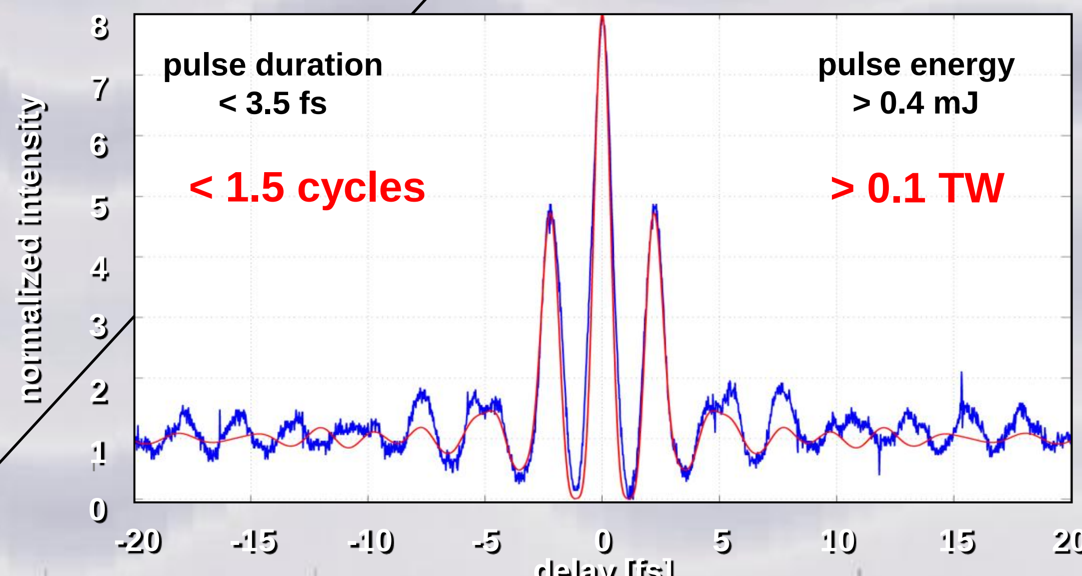
Frontiers of Intense Laser Physics
KITP Santa Barbara, CA, USA
16.09.2014

The tools…

key tools of attosecond technology:
synthesized few-cycle wave & synchronized sub-fs xuv pulse

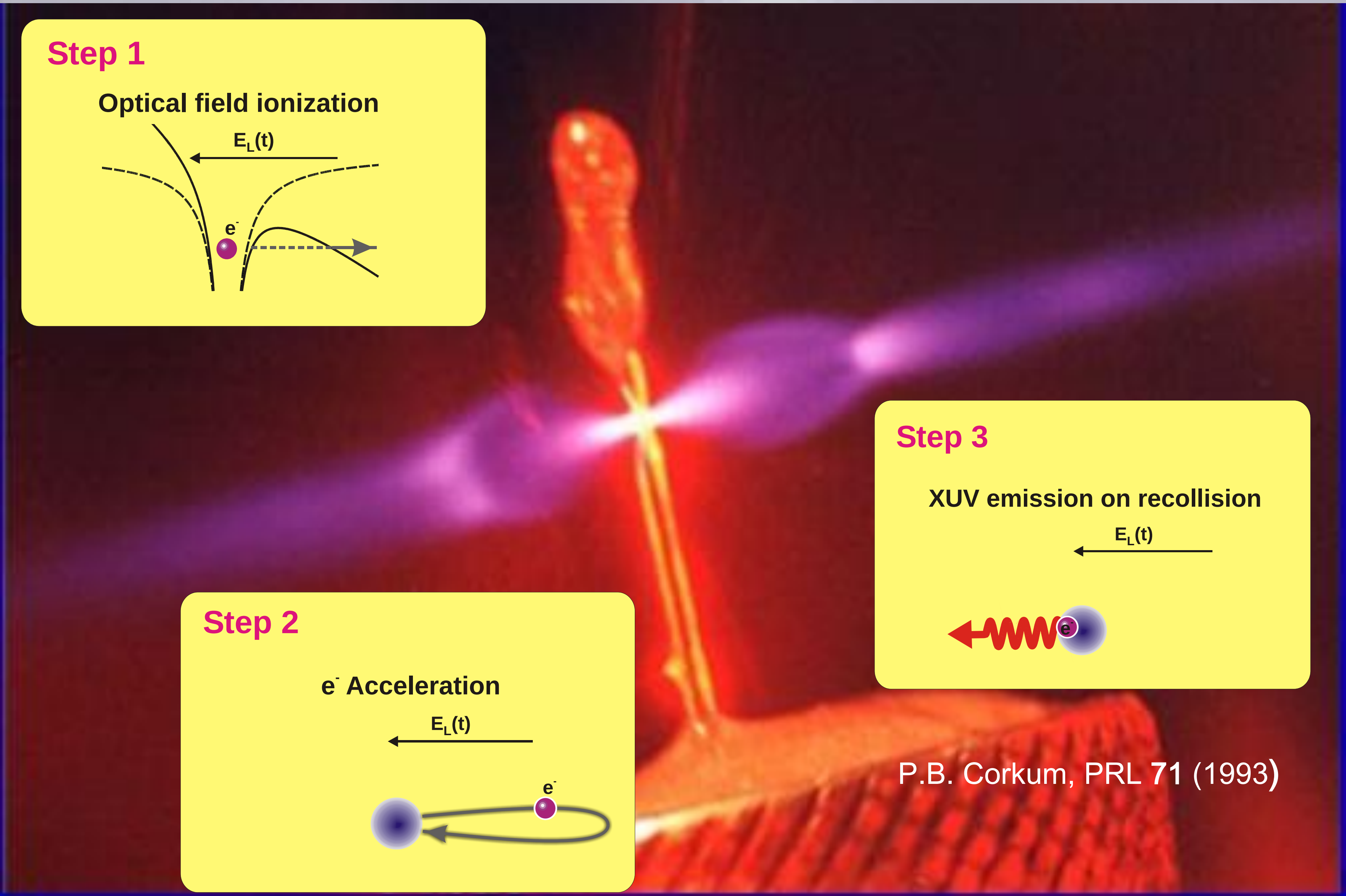






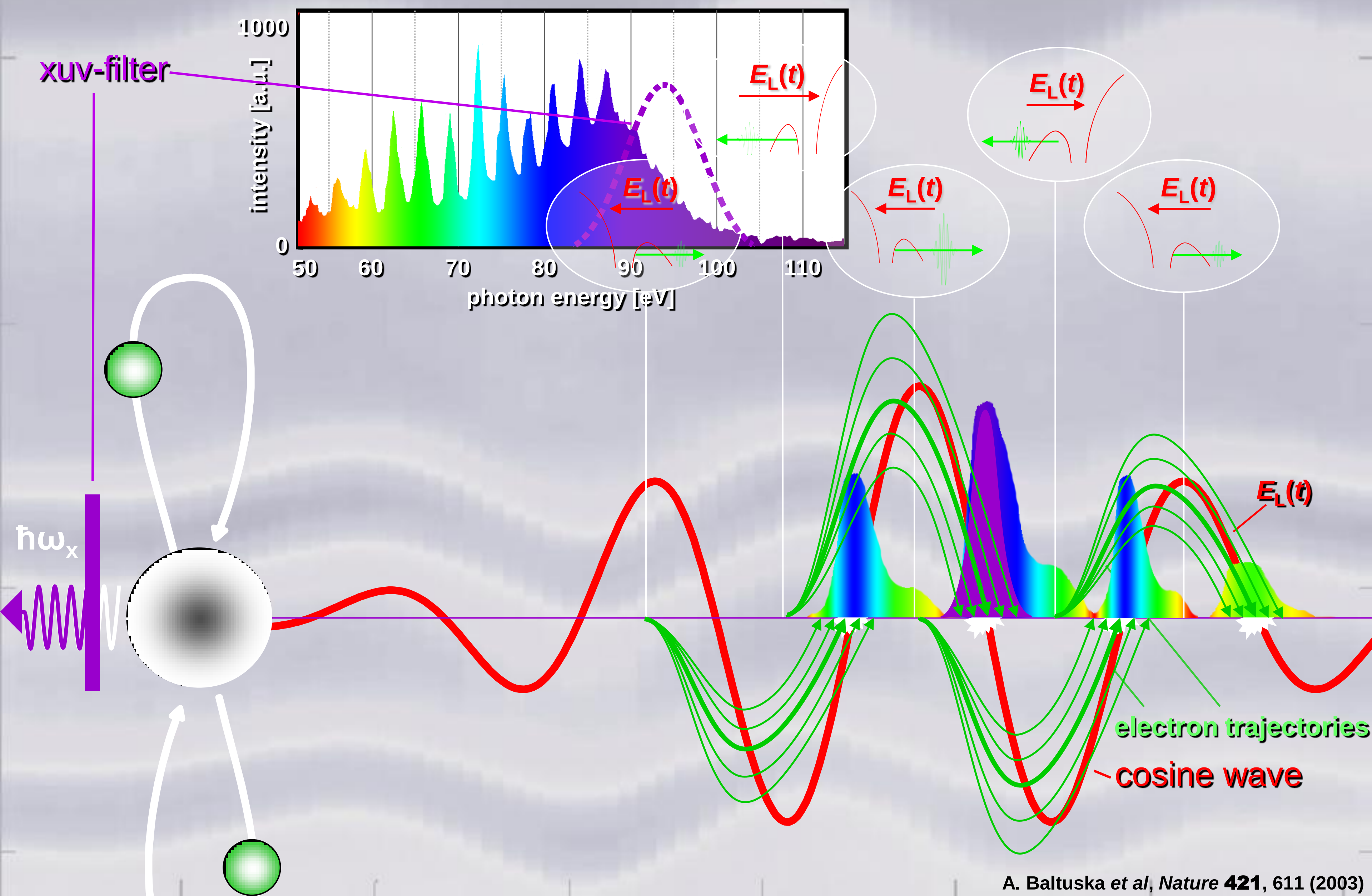
A. L. Cavalieri *et al*, *New J. Phys.* **9**, 242 (2007);
E. Goulielmakis *et al*, *Science* **317**, 769 (2007)

High-order Harmonic Generation in the gas phase



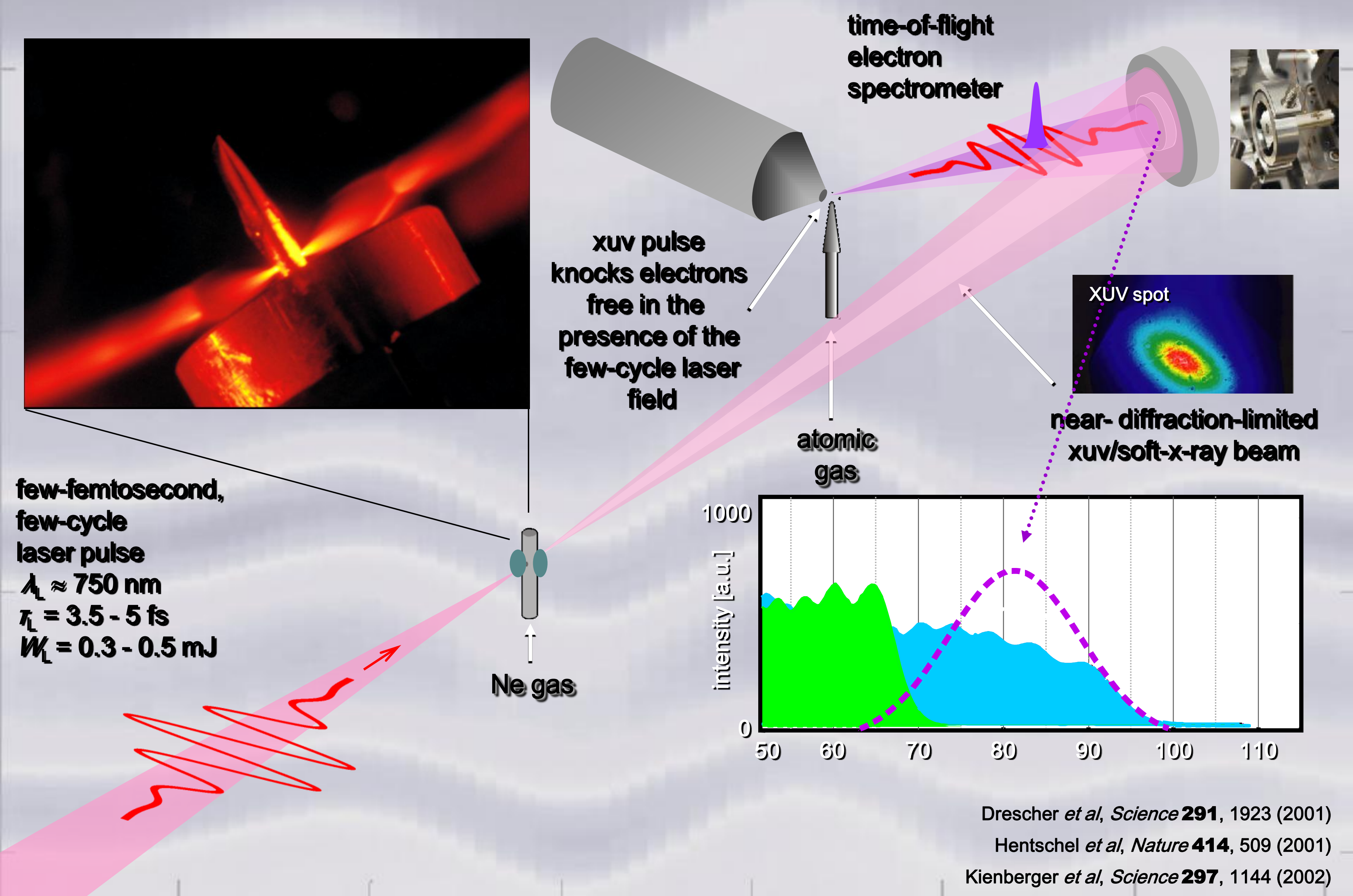
P.B. Corkum, PRL **71** (1993)

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



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)

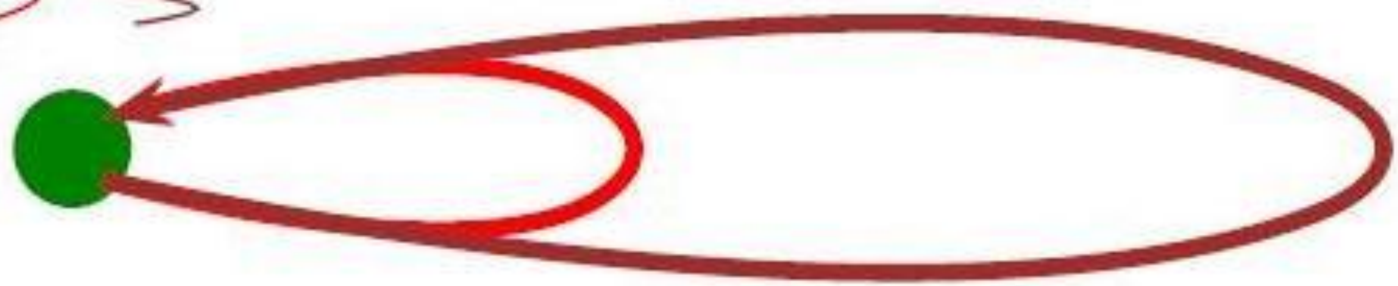
Kienberger *et al*, *Science* **297**, 1144 (2002)

Higher photon energy…

Extending the cutoff

$$(\hbar\omega)_{\max} = I_p + 3.17U_p$$

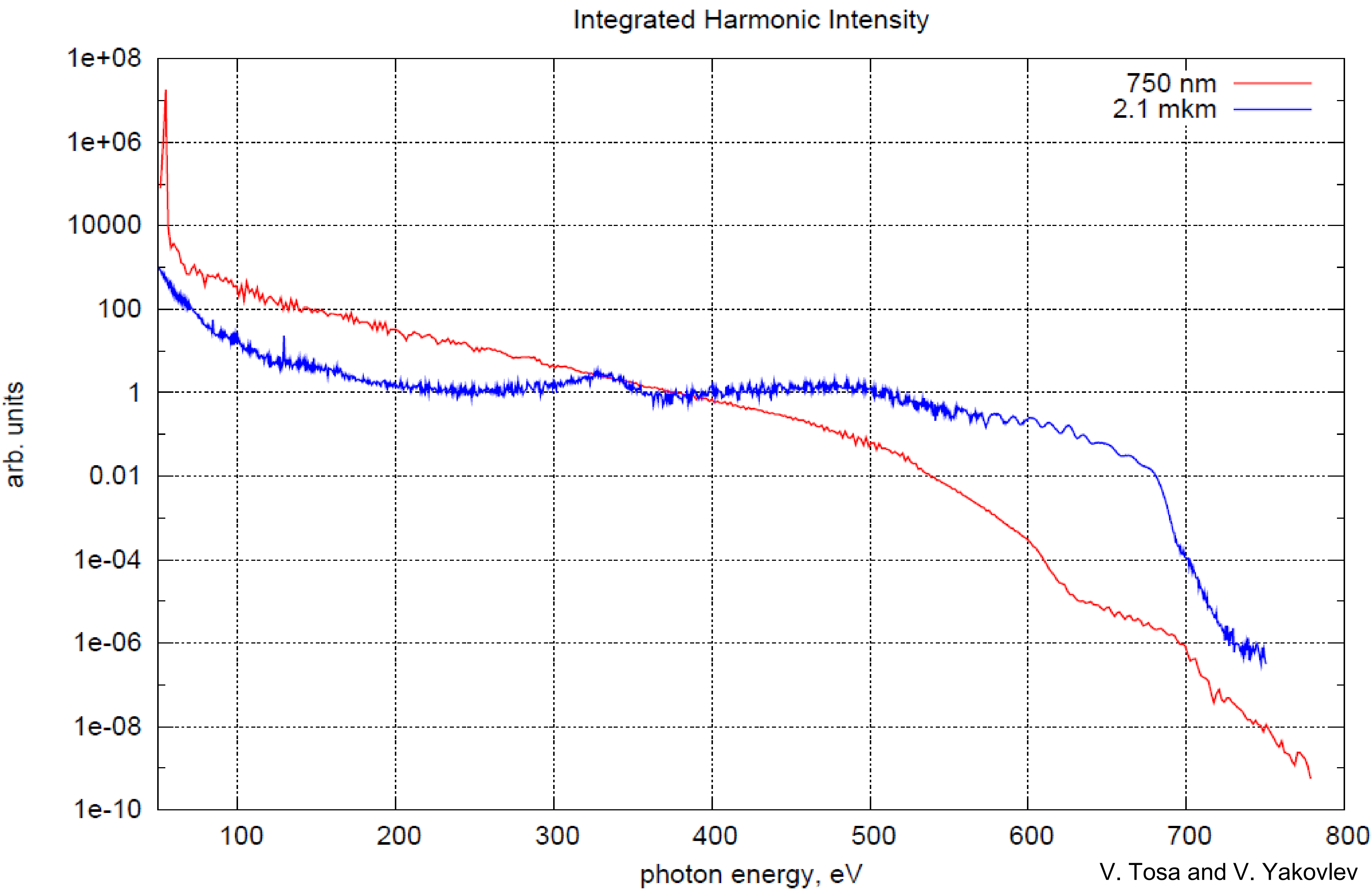
$$U_p = e^2E^2/4m\omega_0^2$$



using an IR driving field

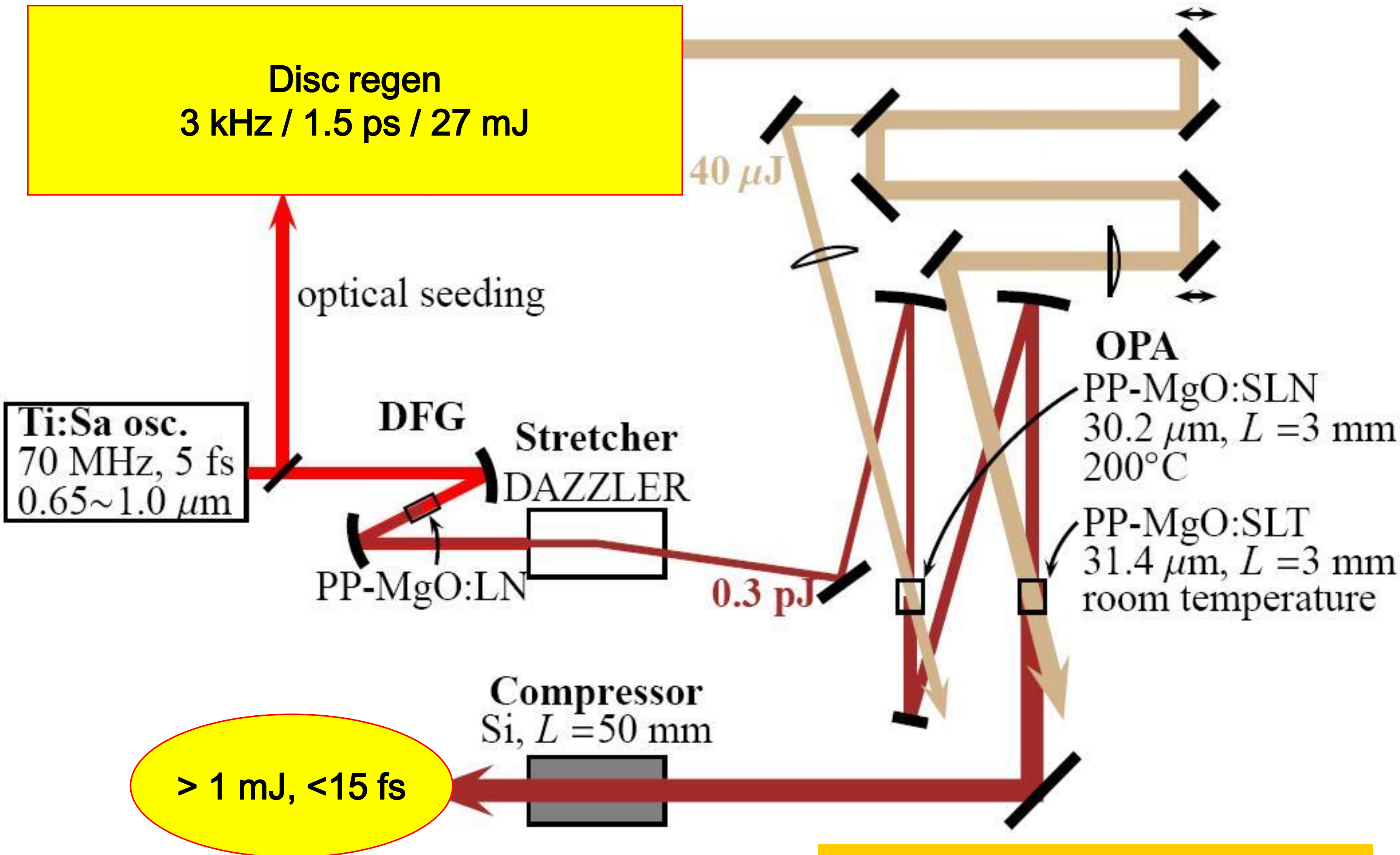
ω:	frequency of HHG
I_p:	atomic ionization potential
U_p:	kinetic energy of the electron
E:	amplitude of electric field
ω_0:	laser frequency
m:	mass of the electron
e:	charge of the electron

Comparison VIS / IR driver



V. Tosa and V. Yakovlev

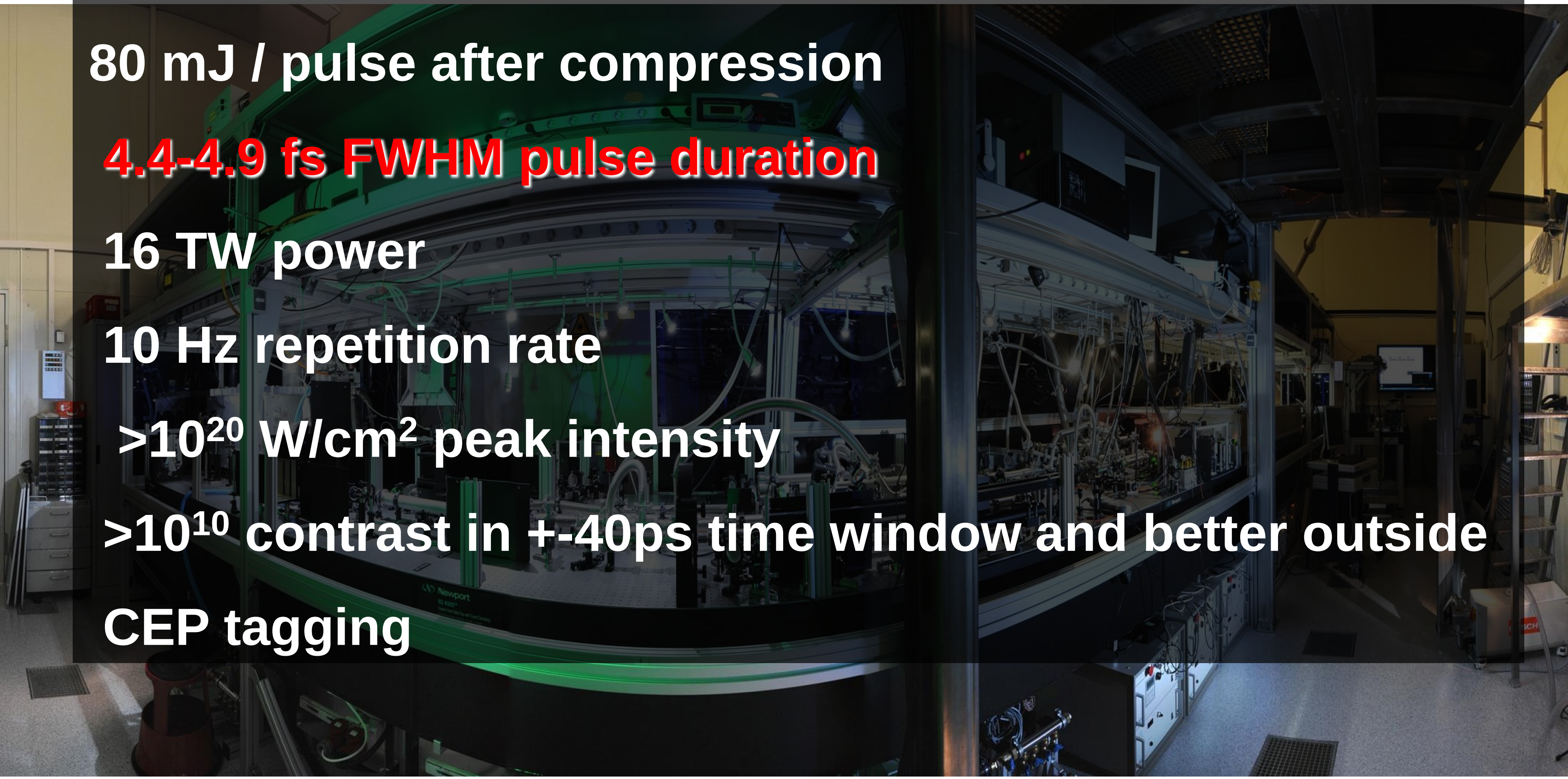
2.1 µm few-cycle OPCPA system



Up to 1 keV, few photons…

Higher XUV intensity

Light Wave Synthesizer-20 (LWS-20) <5 fs

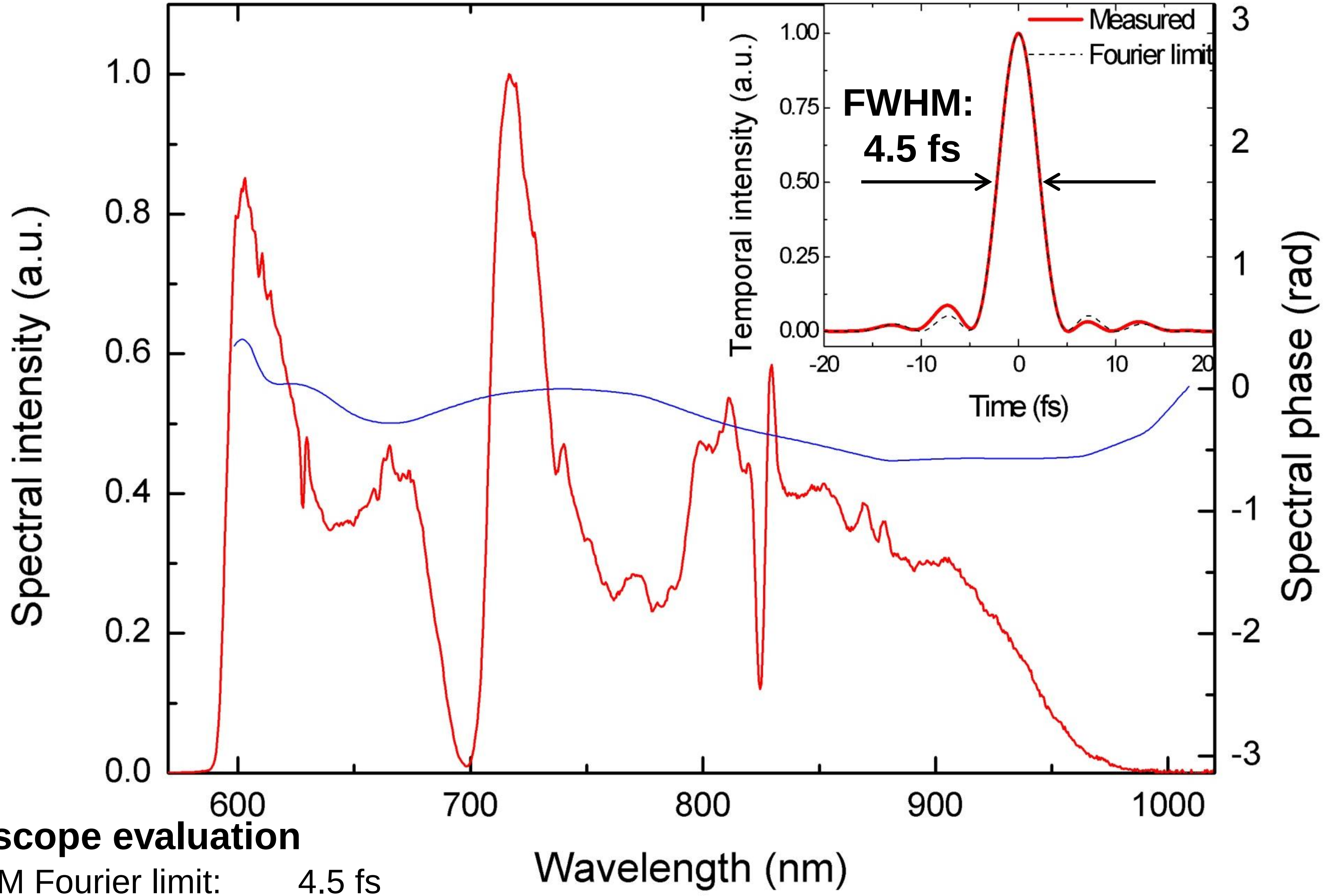


Properties :

- 80 mJ / pulse after compression
- **4.4-4.9 fs FWHM pulse duration**
- 16 TW power
- 10 Hz repetition rate
- $>10^{20}$ W/cm^2 peak intensity
- $>10^{10}$ contrast in +-40ps time window and better outside
- CEP tagging

Courtesy of Laszlo Veisz

Compression



Dazscope evaluation

FWHM Fourier limit: 4.5 fs

FWHM pulse duration: 4.5 fs

SHG AC indicates also this duration!

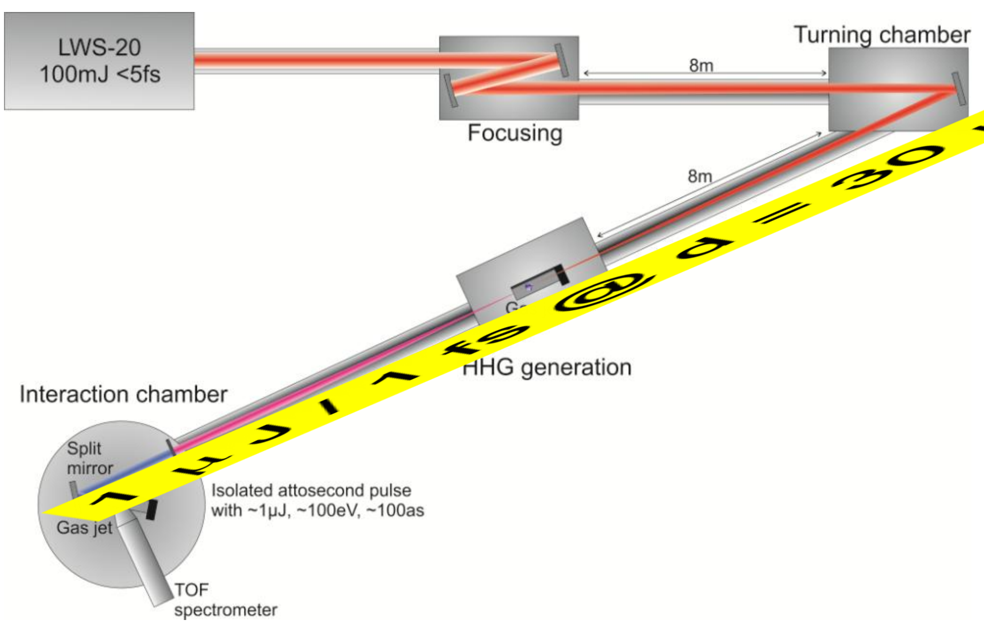
Courtesy of Laszlo Veisz

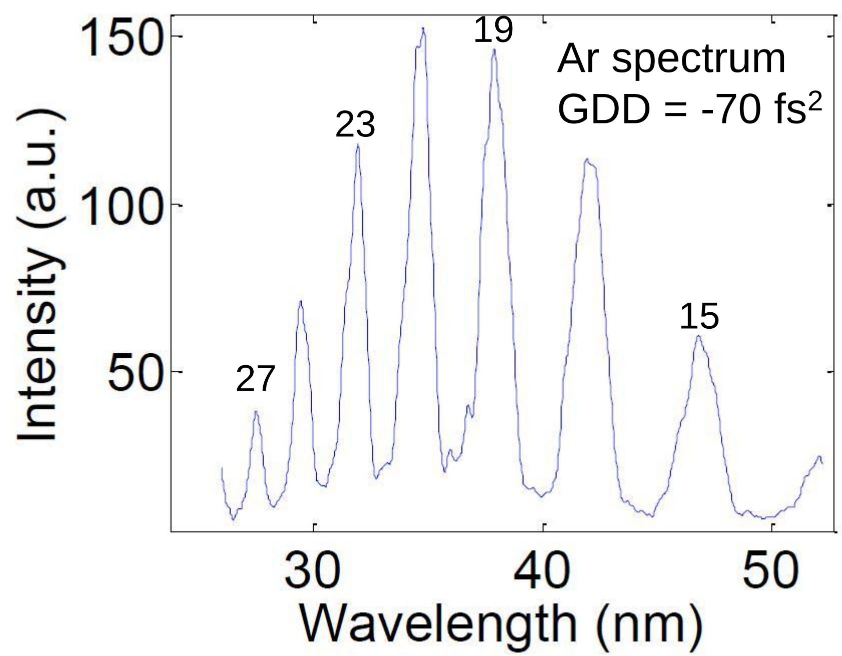
Applications: gas HHG

Applications: gas HHG

High harmonic generation from gas targets

- **Single (few-)100-attosecond pulse** generation
- **up to 1 μJ around 100 eV** in Neon in an isolated attosecond pulse



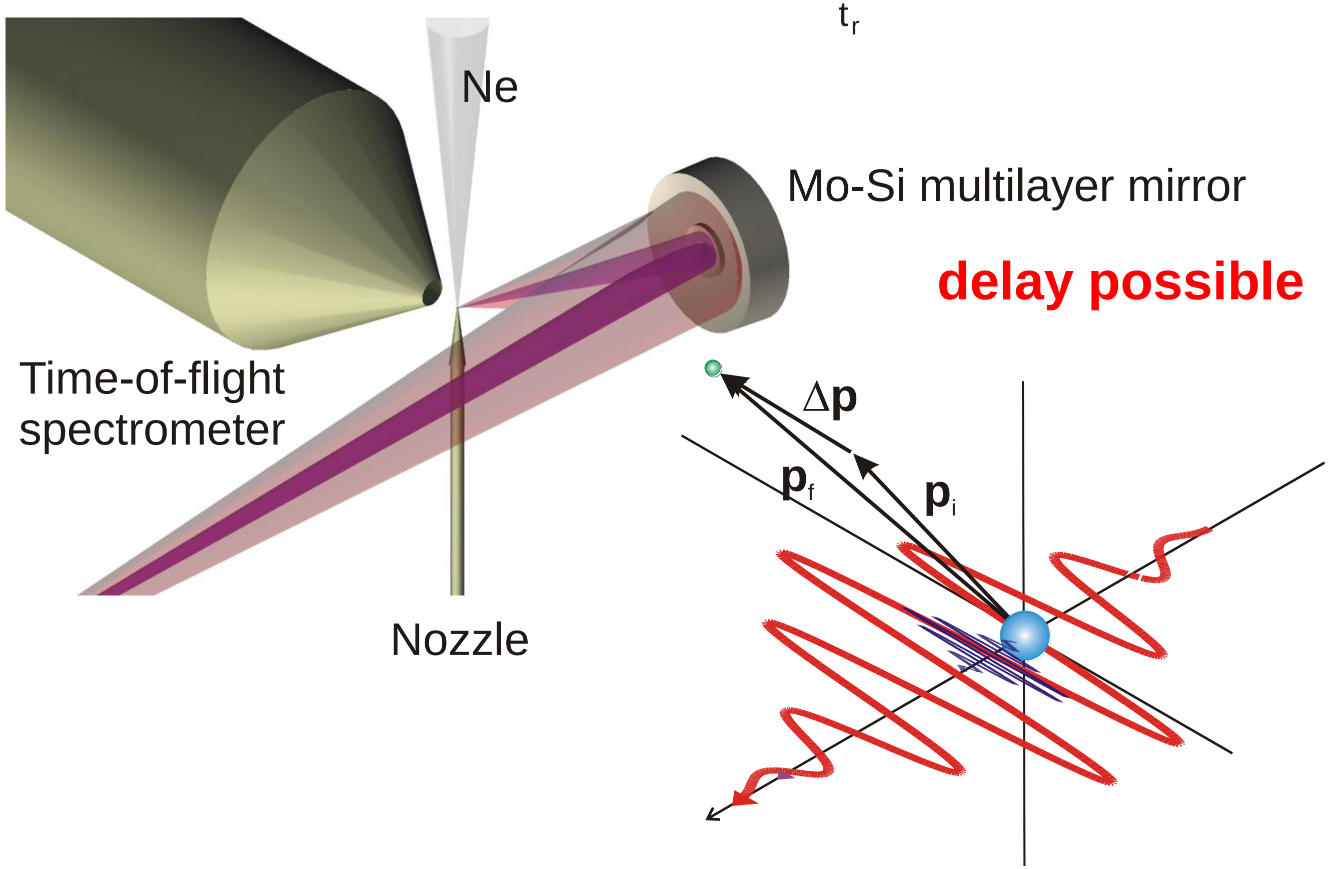


Courtesy of Laszlo Veisz

The method…
XUV pump / NIR field probe

Photoelectrons generated by an as-pulse

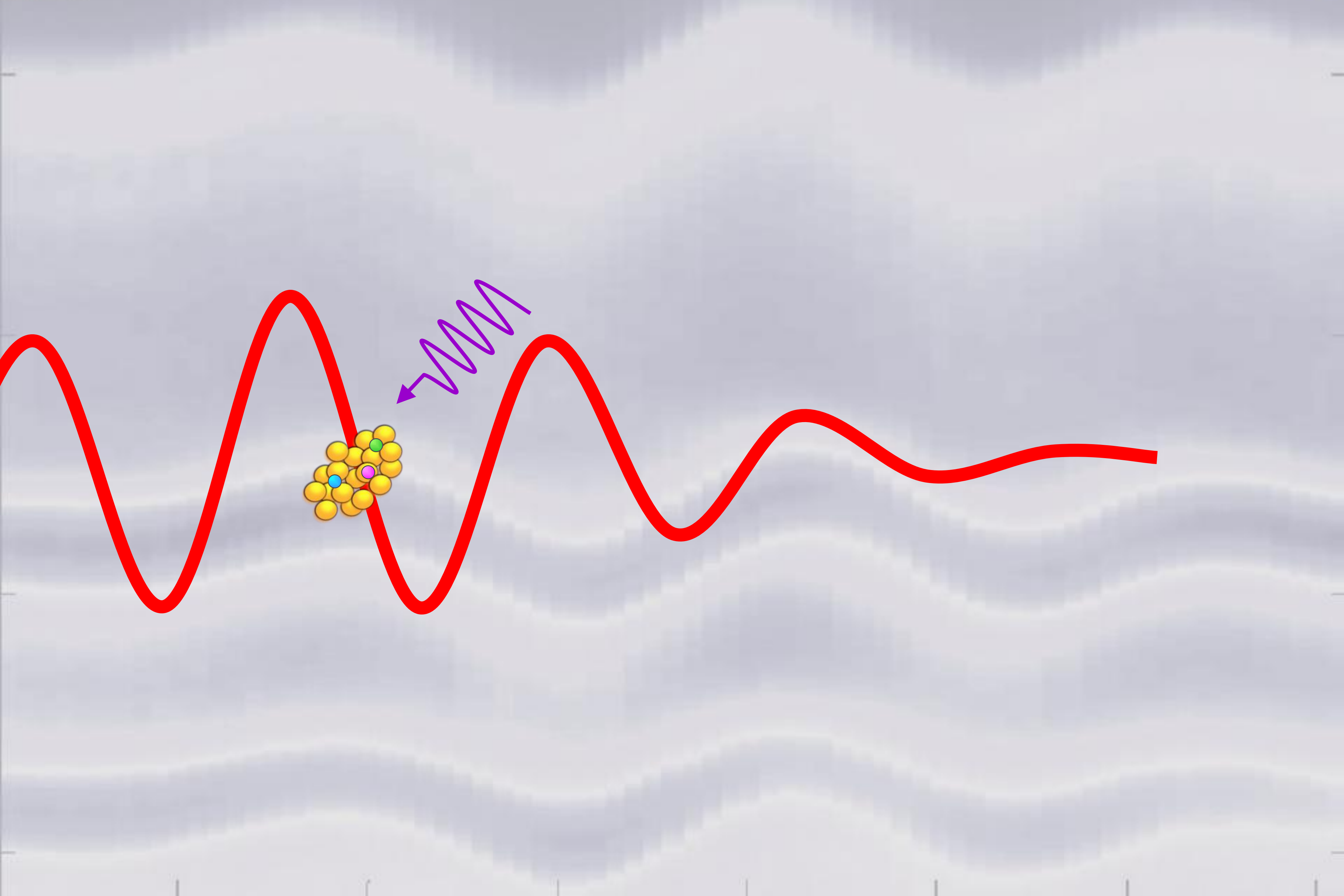
$$\Delta p(t_r) = e\int_{t_r}^{\infty} E_L(t')\,dt'$$



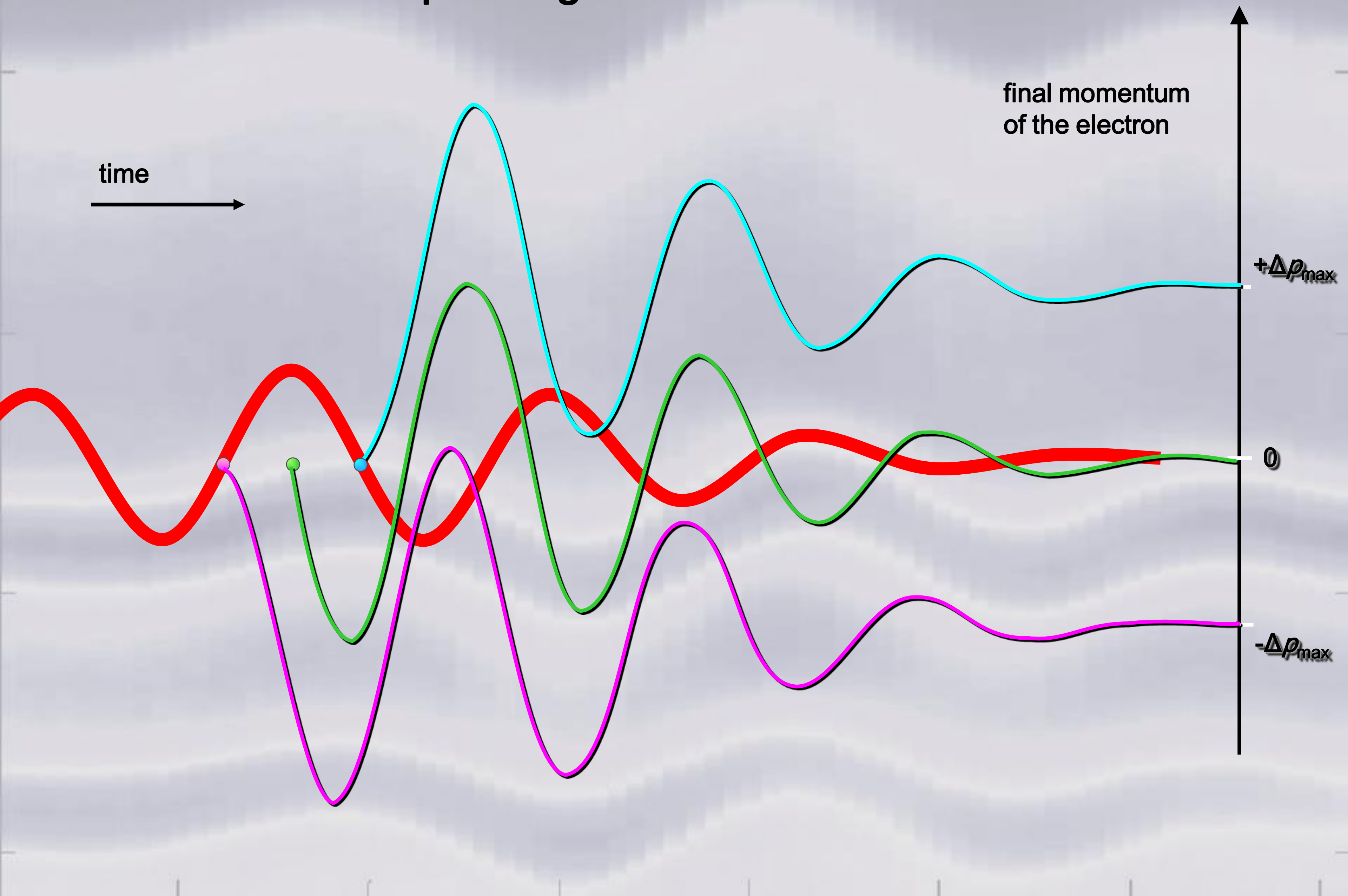
Kienberger *et al.*, Science **297**, 1144 (2002)

XUV cut-off energy: ~100 eV
Mirror reflectivity bandwidthup to: 30 eV (FWHM)

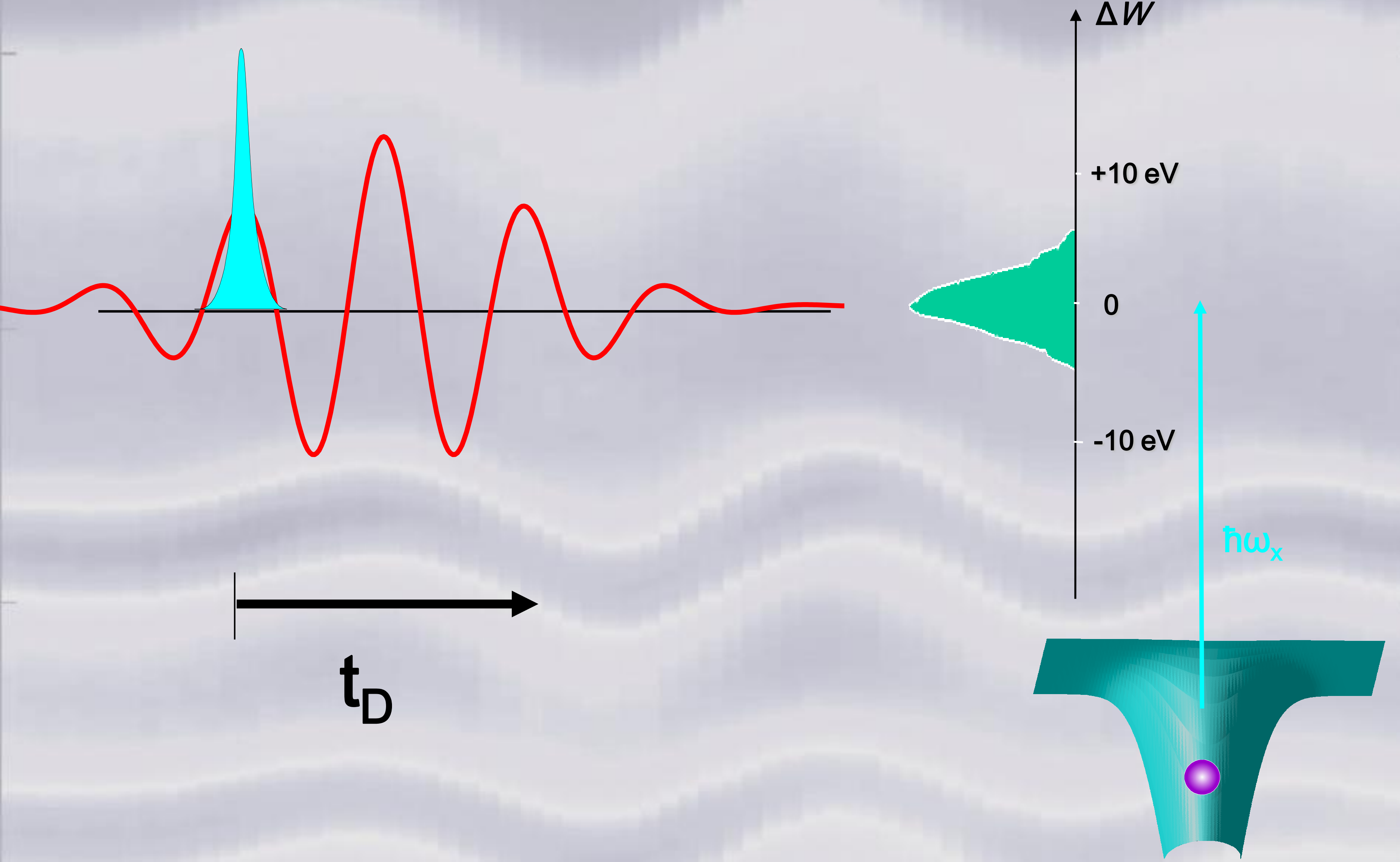
Ionization at different instants of time



final momentum of photoelectrons depending on the release time

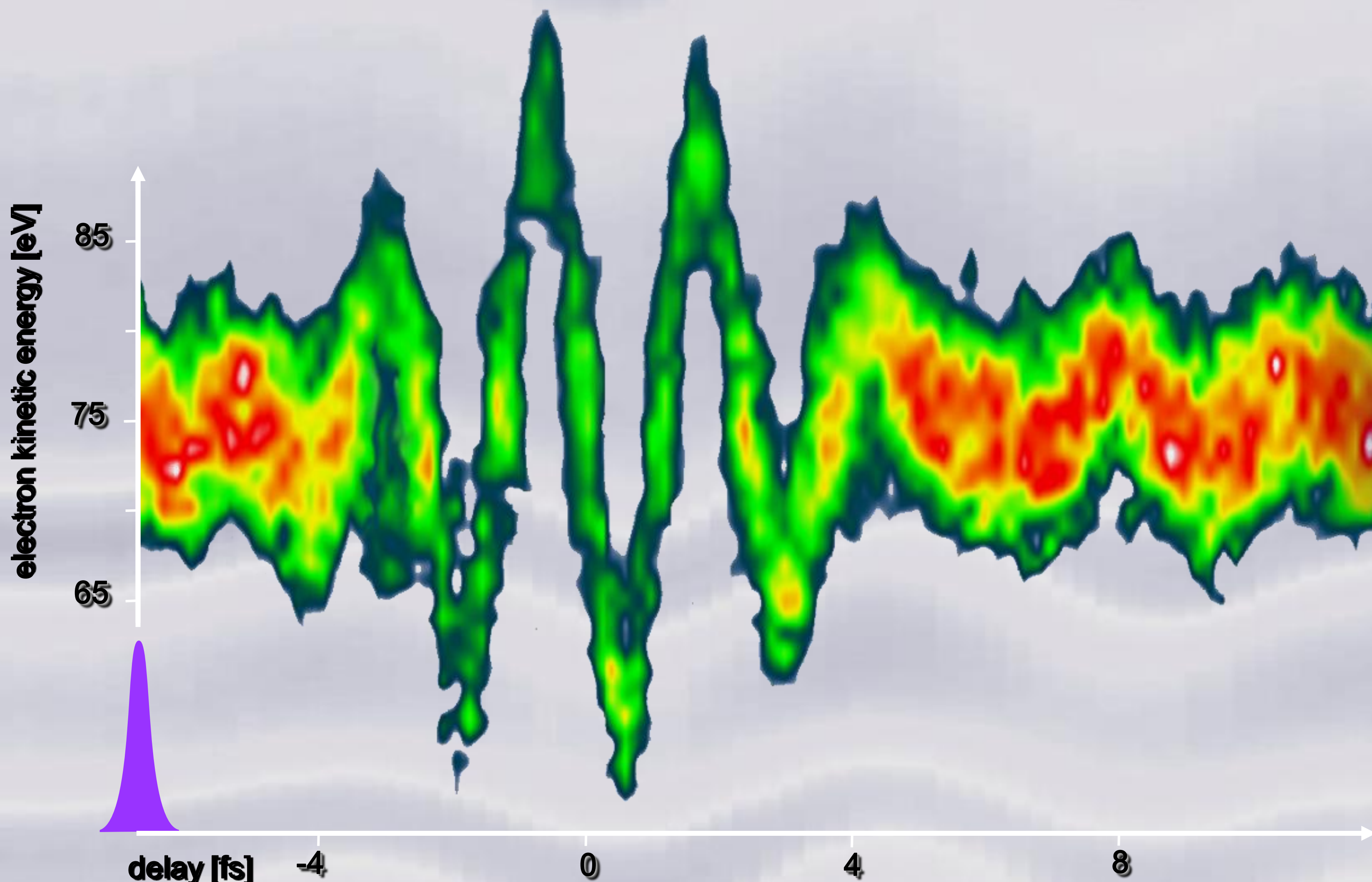


sampling field oscillations



R. Kienberger et al, Nature **294** (2004)

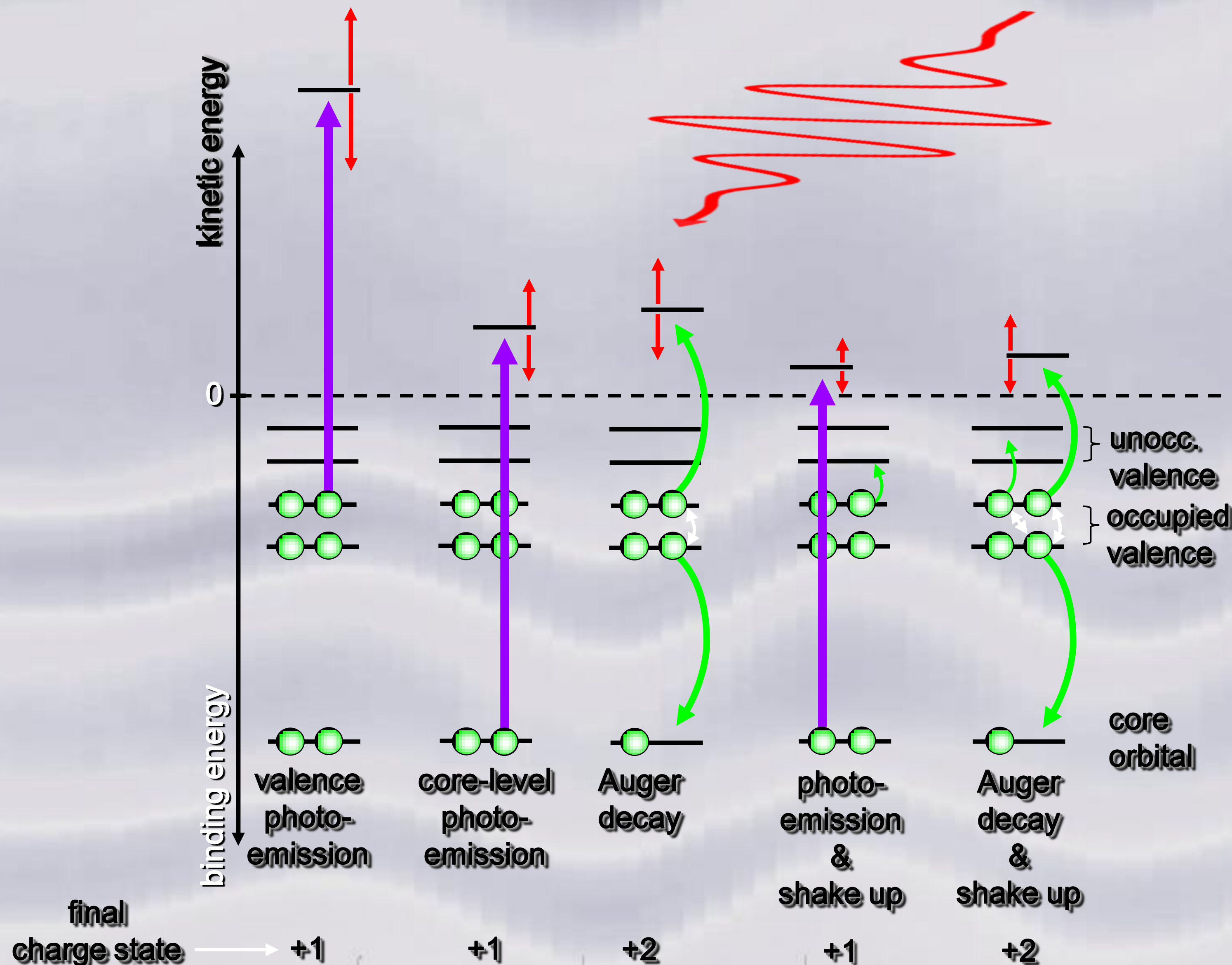
a streaking trace



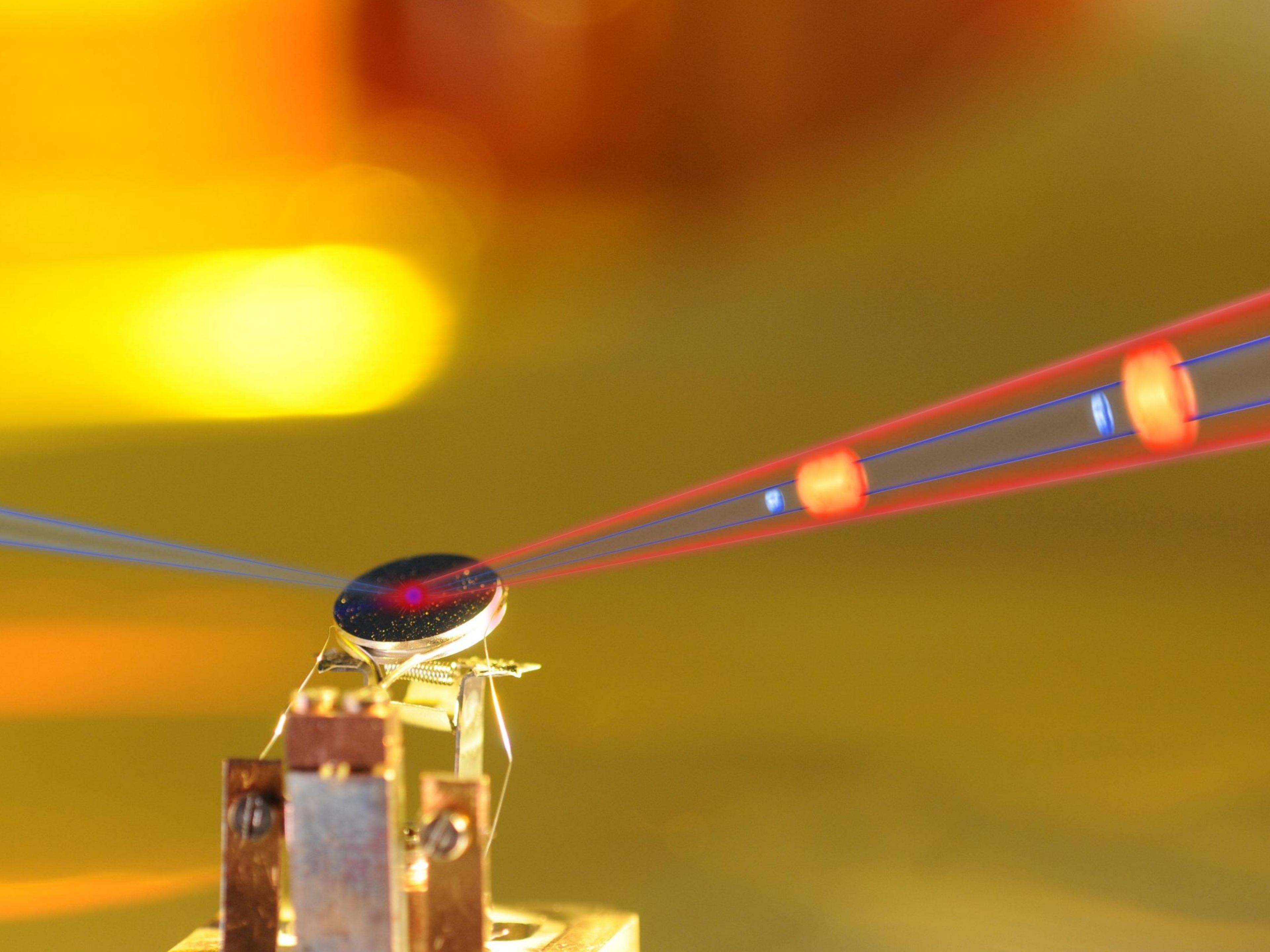
E. Goulielmakis *et al*, *Science* **305**, 1267 (2004)

real-time observation of multi-electron dynamics (excitation, relaxation, correlation) in atoms, molecules & solids?

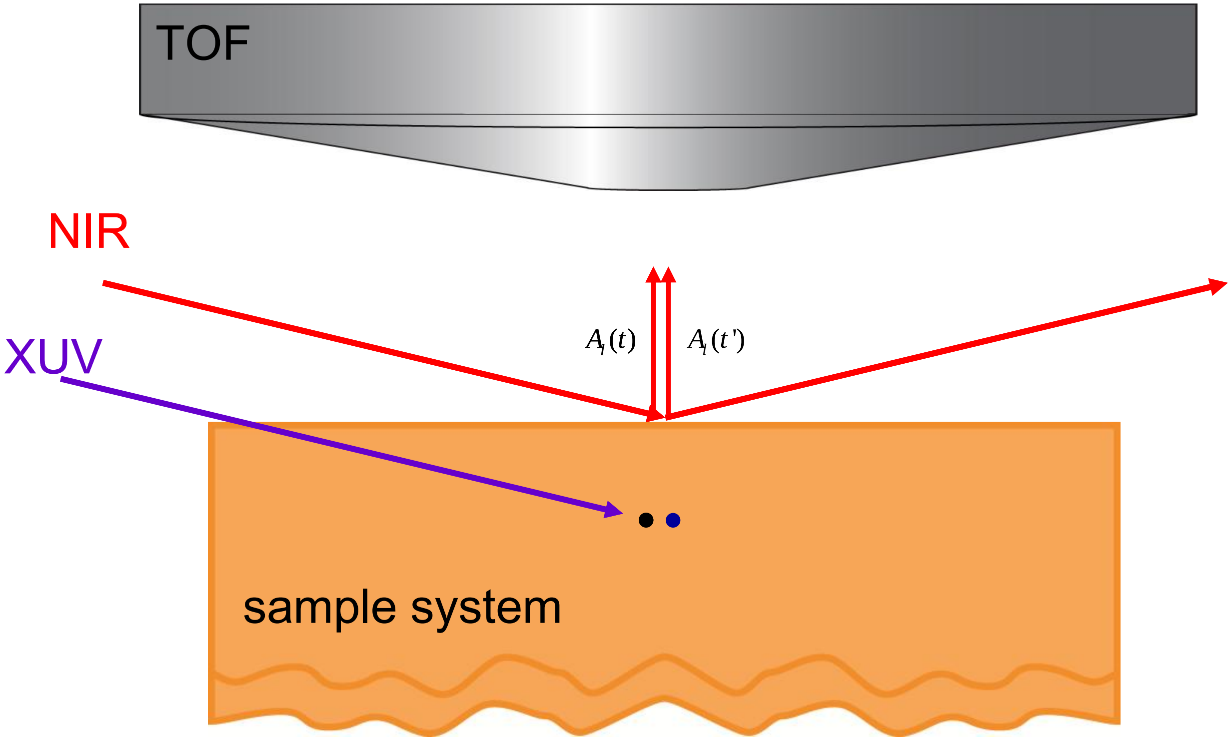
by means of strong-field-induced free-free transitions: streaking

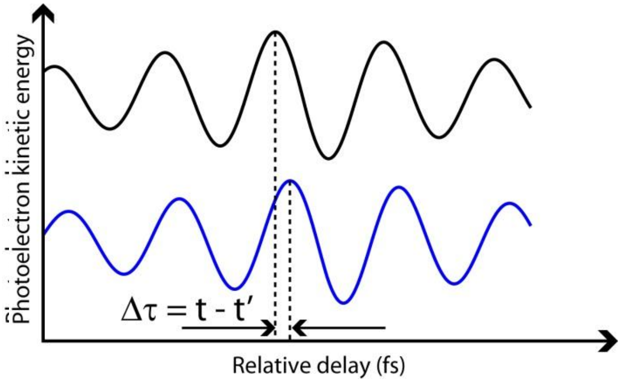


Application in solids…



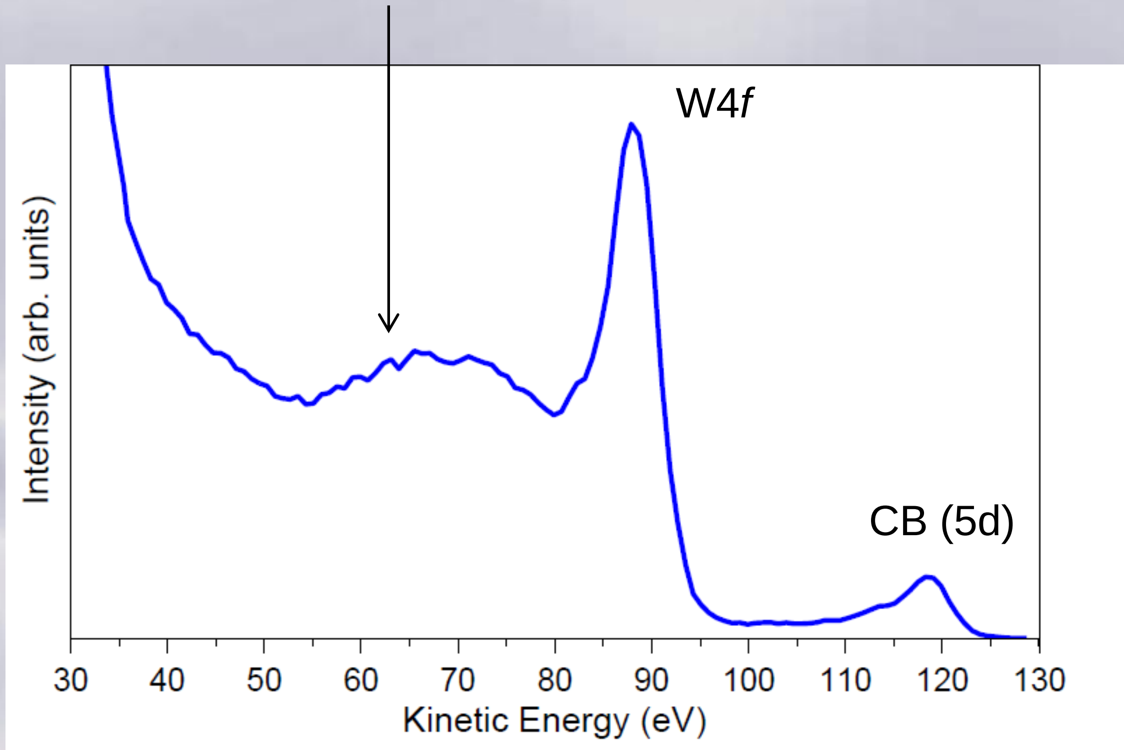
Ionization of electrons in a solid sample

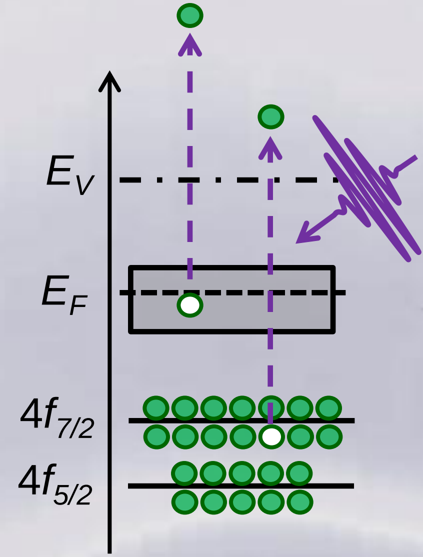




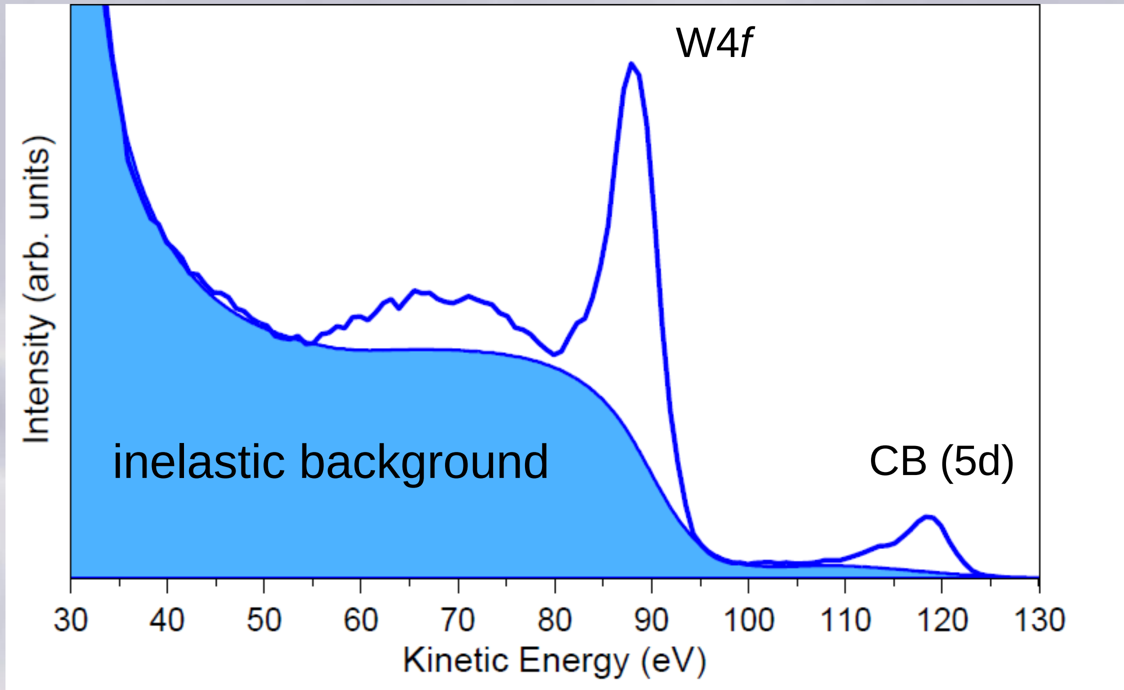
Attosecond Photoemission from W(110)

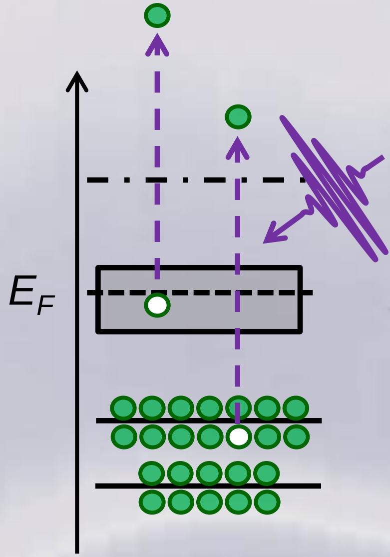
measured with ~400as pulses @ ~120eV





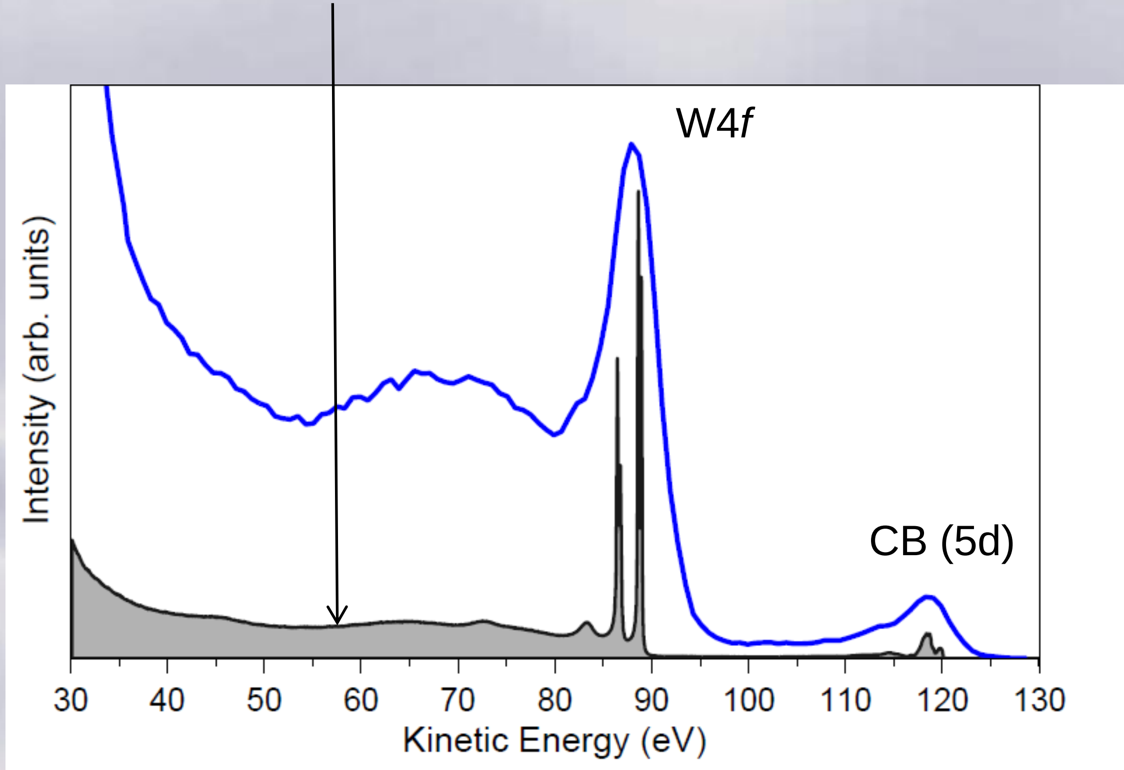
Attosecond Photoemission from W(110)

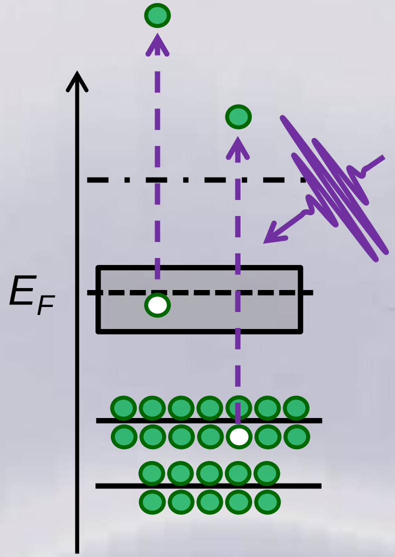




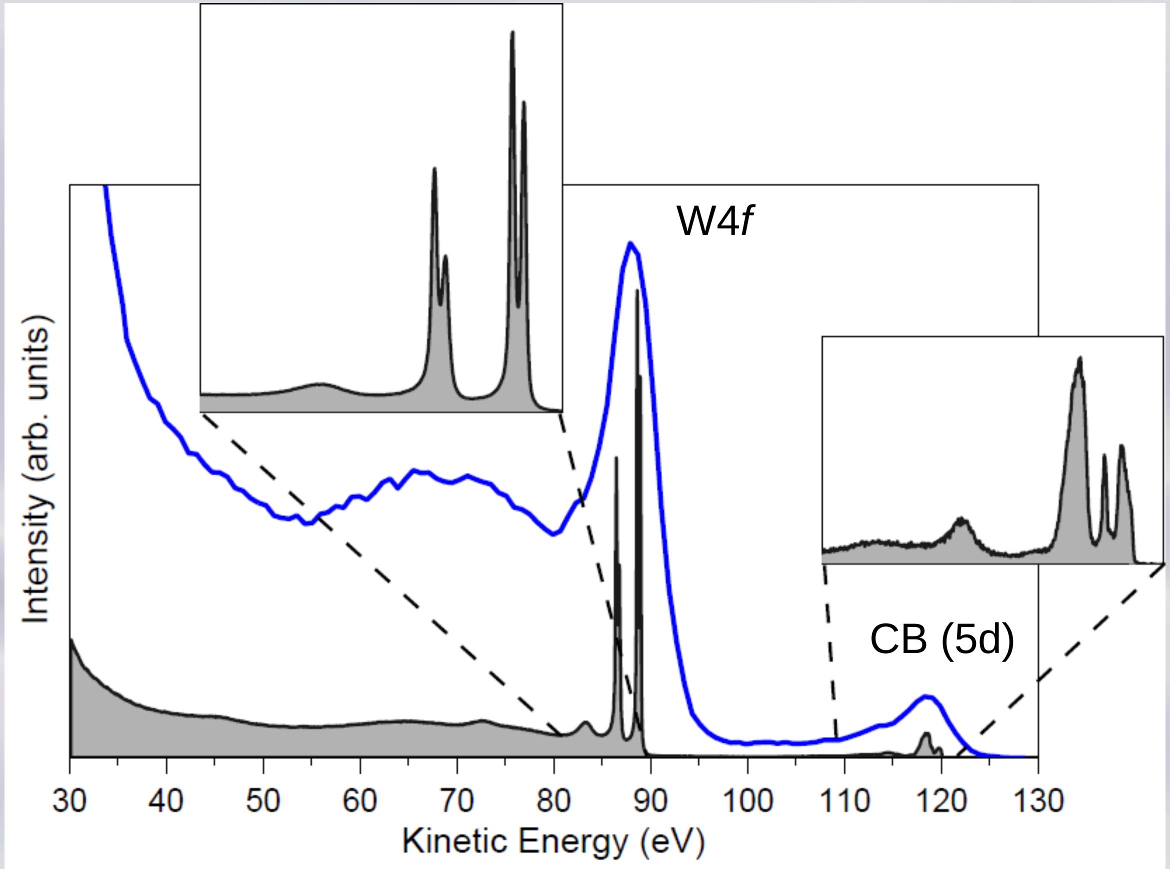
Attosecond Photoemission from W(110)

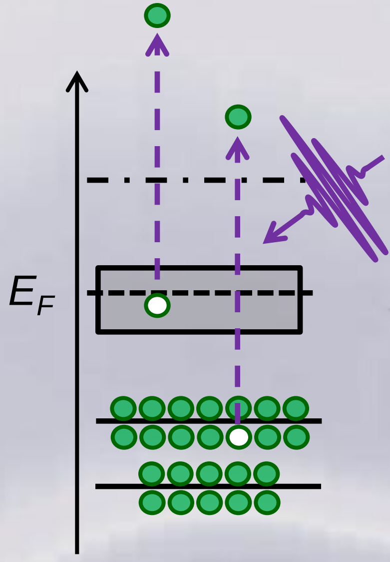
120eV narrow-bandwidth synchrotron radiation



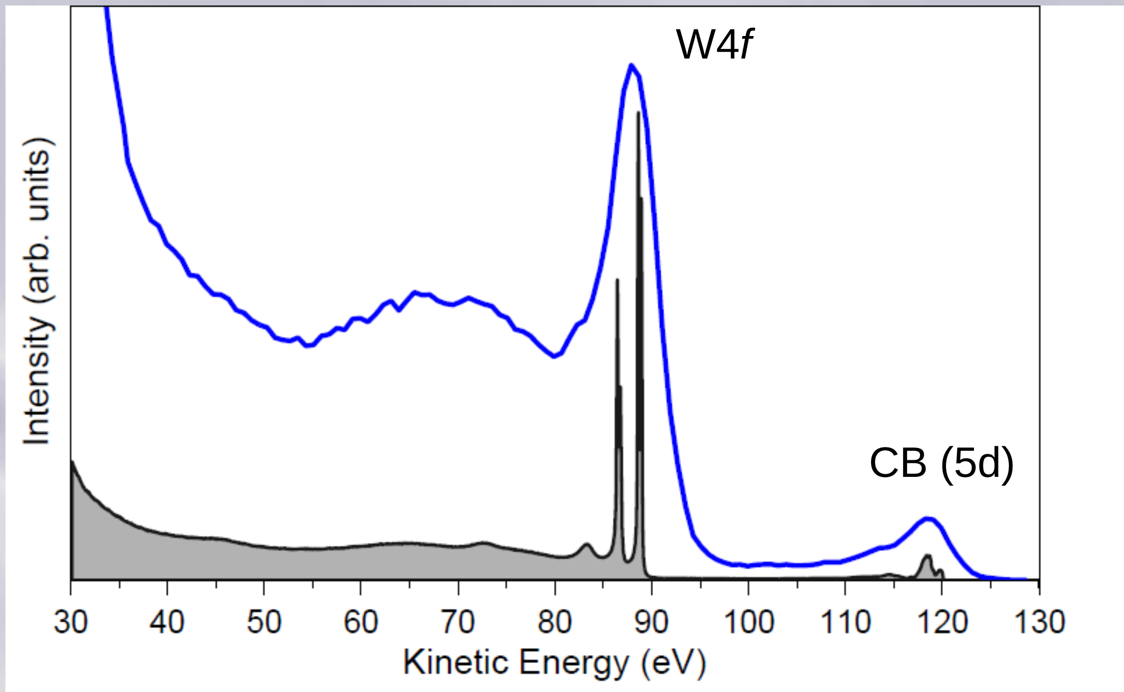


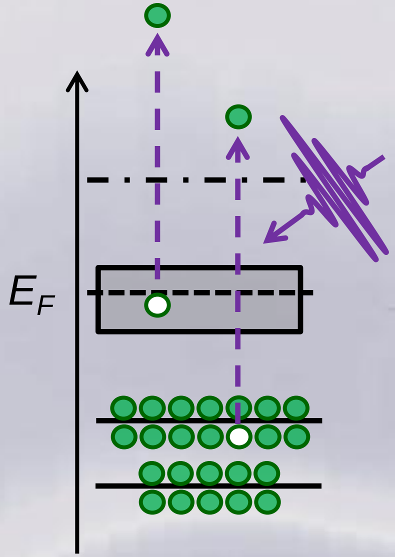
Attosecond Photoemission from W(110)





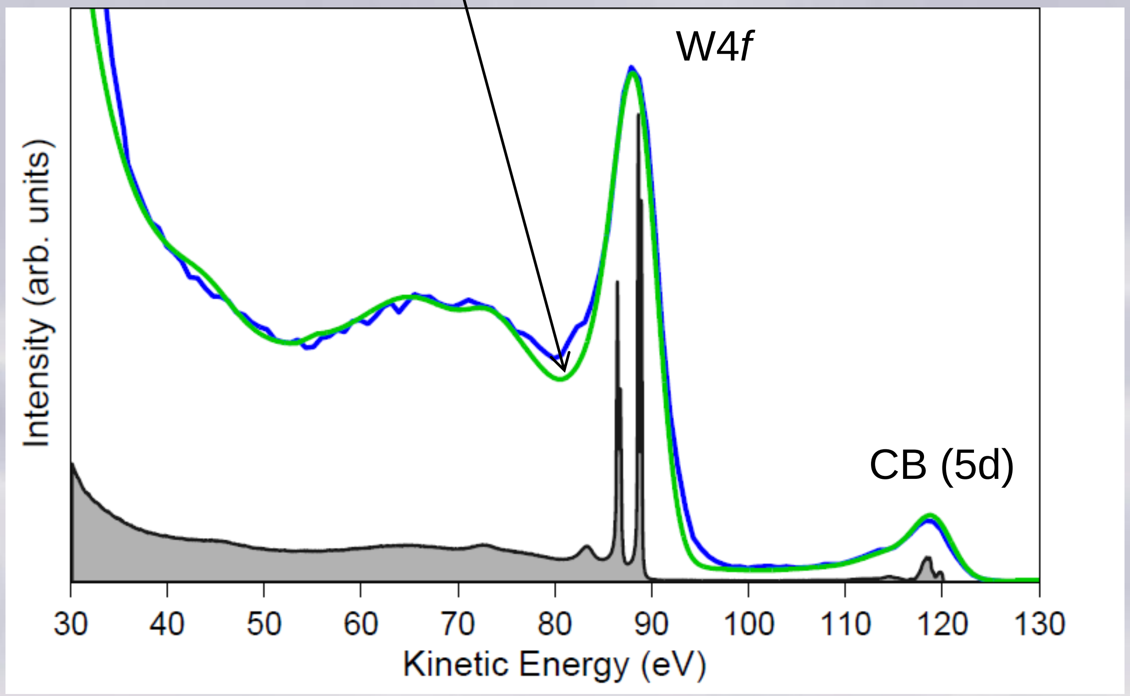
Attosecond Photoemission from W(110)

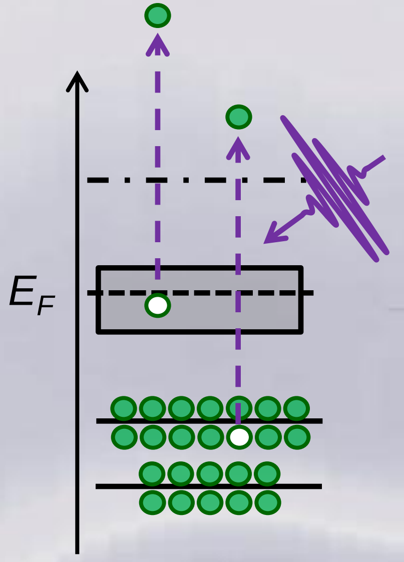




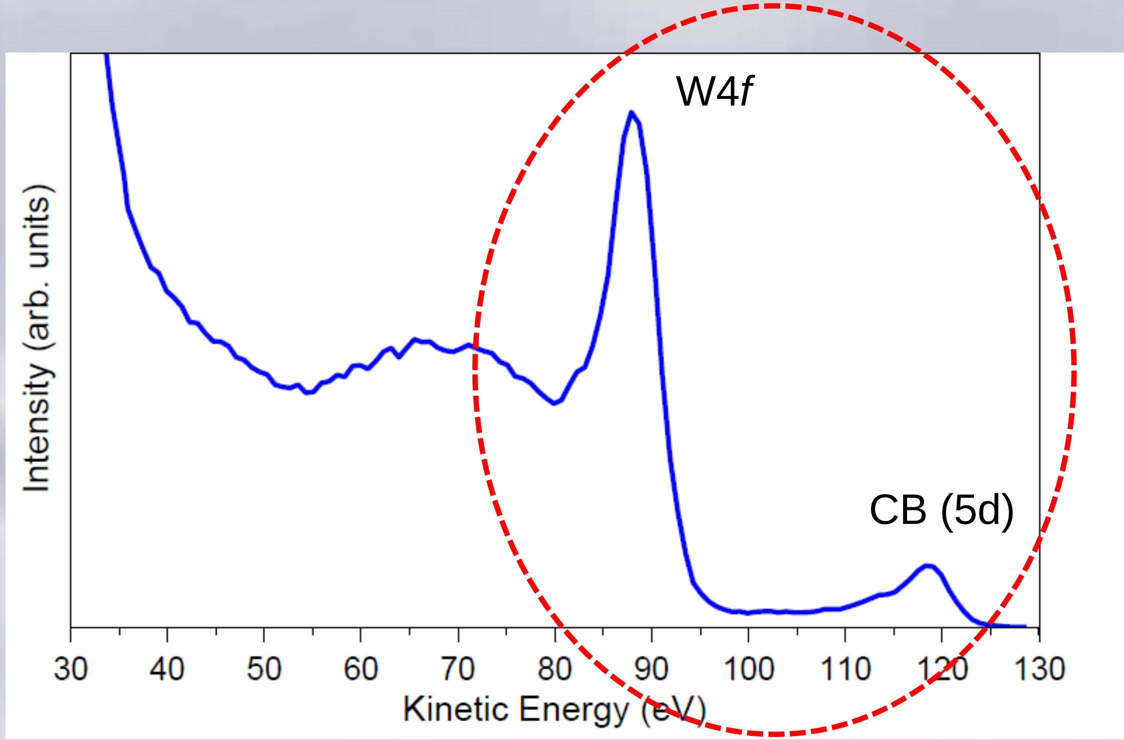
Attosecond Photoemission from W(110)

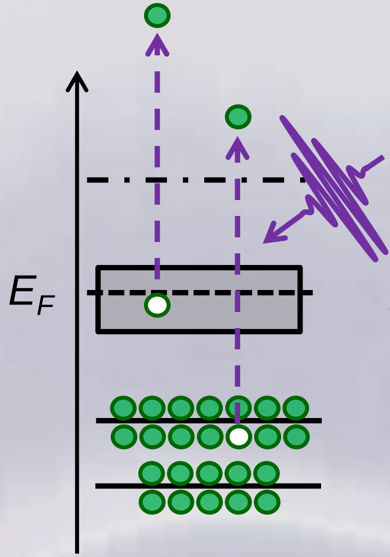
convoluted with spectral
bandwidth of the as-pulse



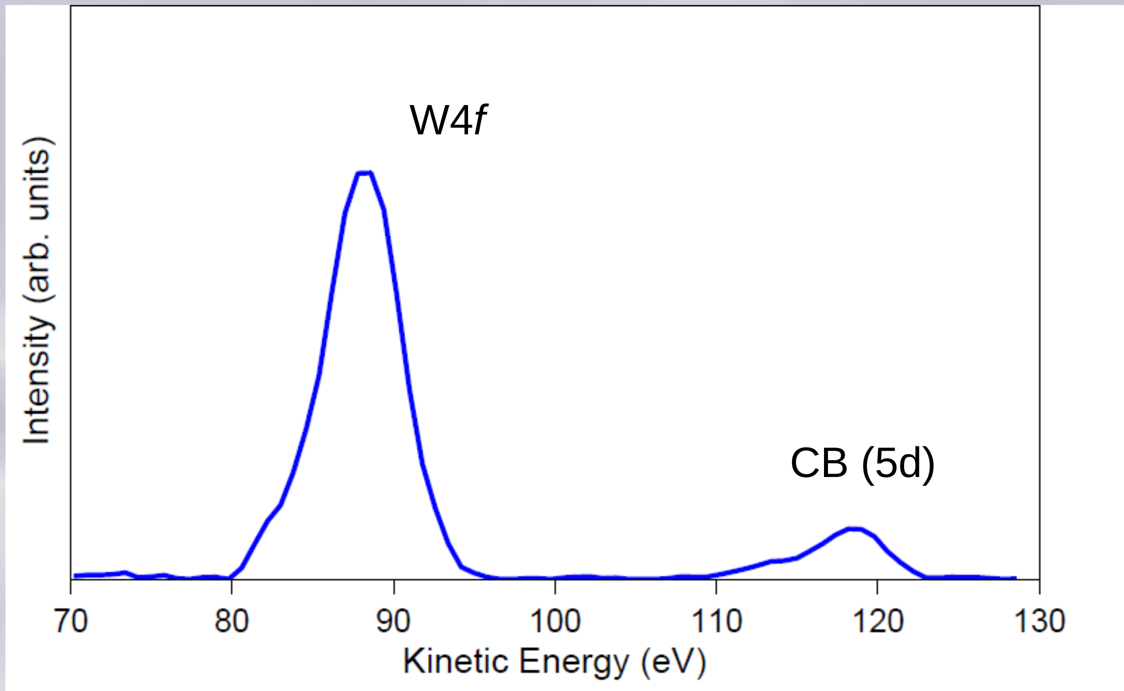


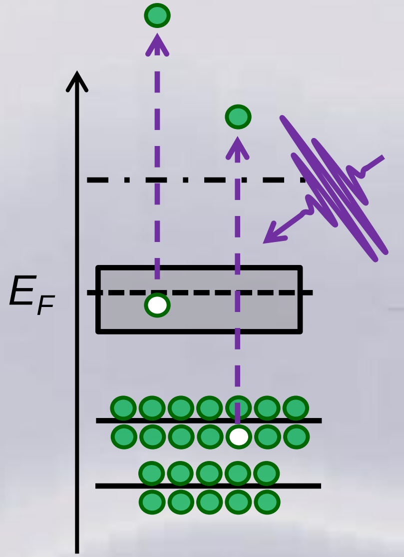
Attosecond Photoemission from W(110)



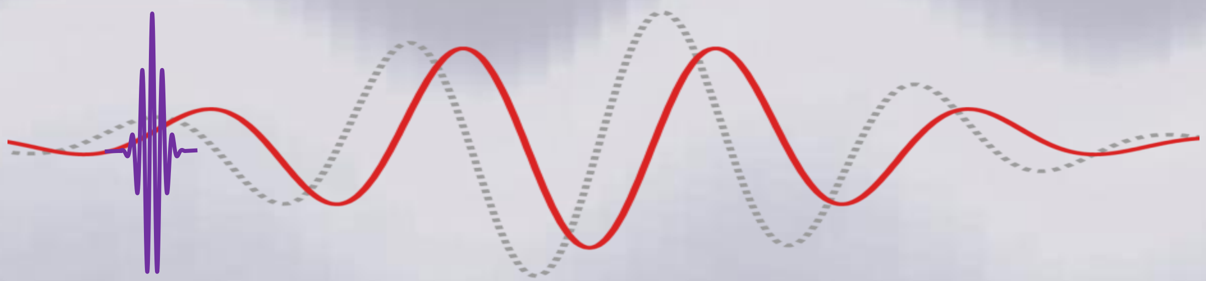


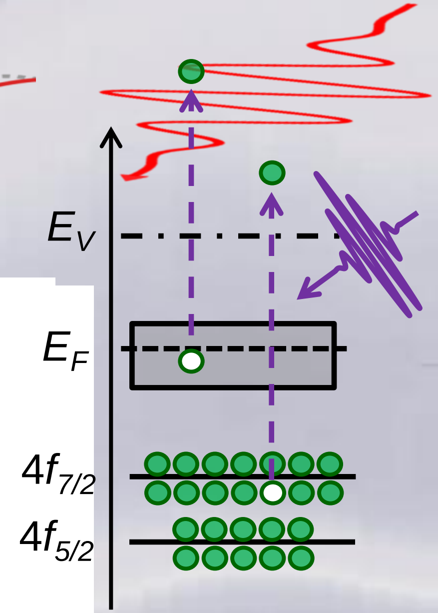
Attosecond Photoemission from W(110)

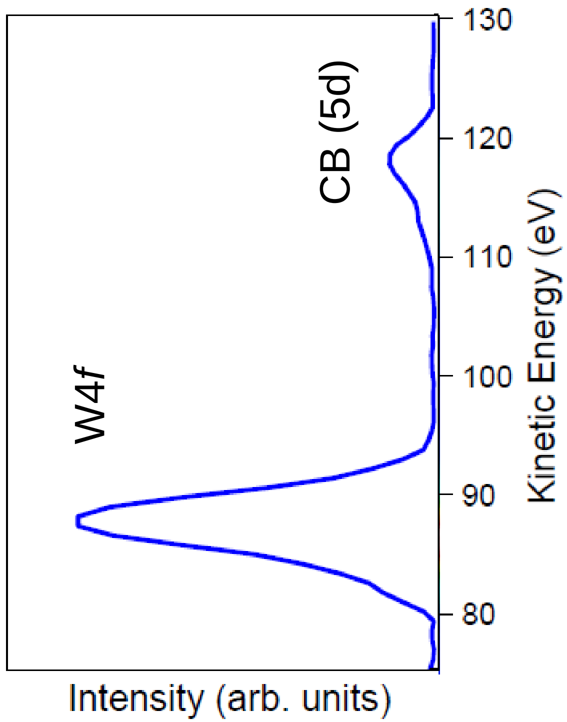




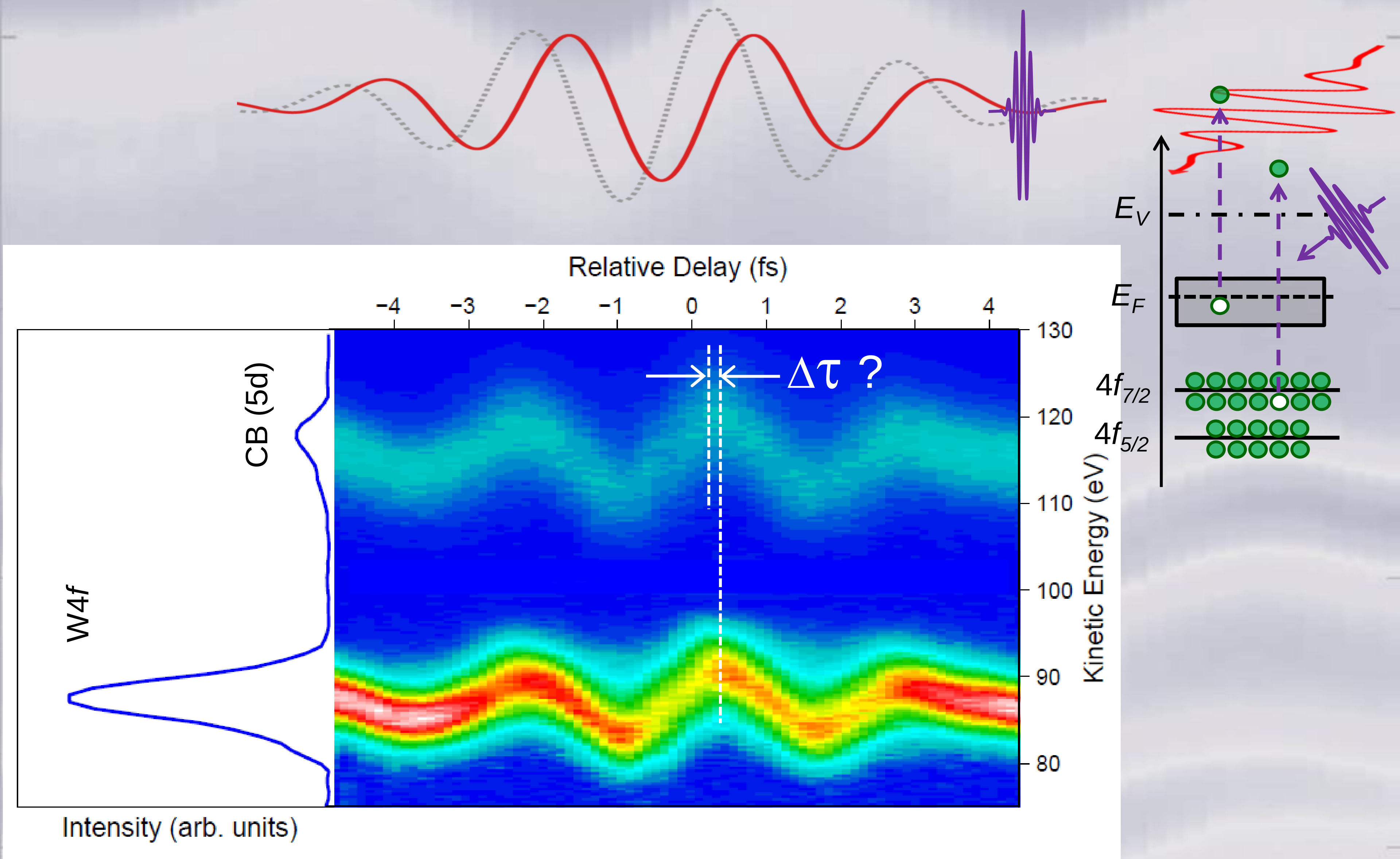
Streaked Photoemission from W(110)



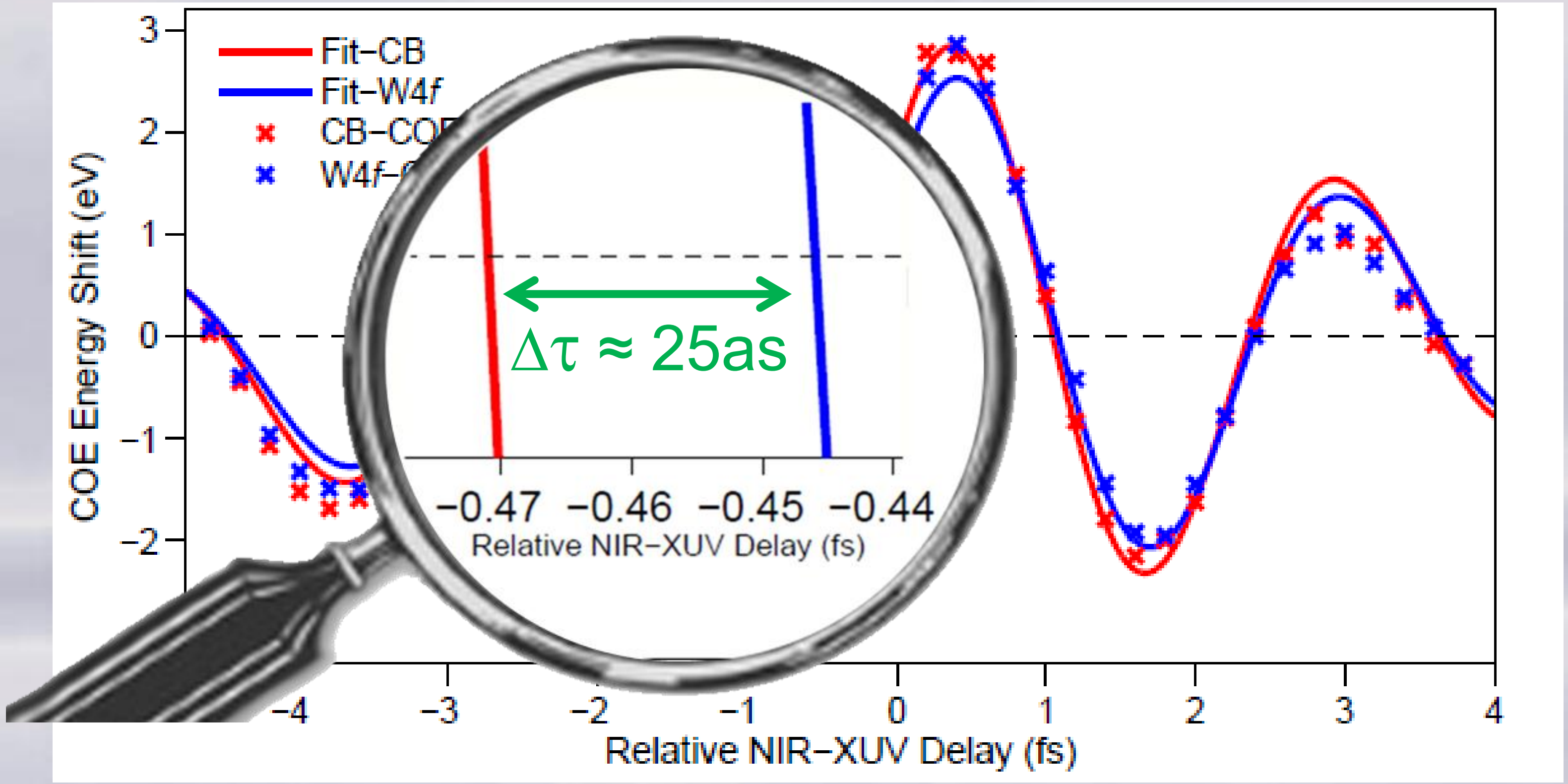




Streaked Photoemission from W(110)



Timing Analysis



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

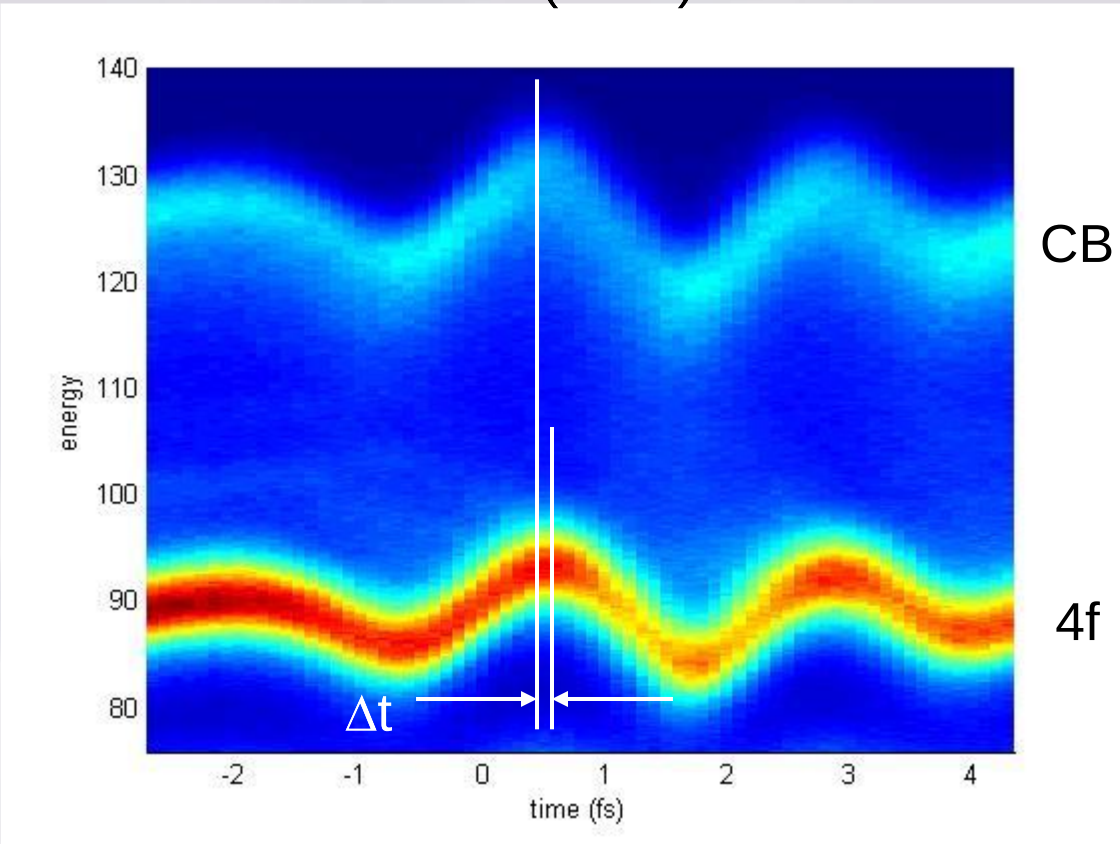
as spectroscopy of clean metal surface

Samples:
Re(0001) and W(110)

Photon energies:
93, 105, 120, 145 eV

Core level electrons delayed by 10 – 100 as.

Re (0001)



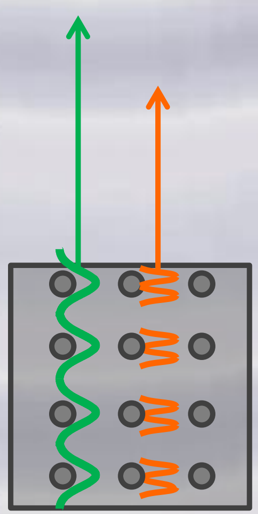
Spectrogram analysis:
retrieval algorithm based on analytical solution of TDSE

Delay of 4*f* core levels relative to CB:
64 ± 10 as

Current state of interpretation

Time delay in photoemission form solids is mainly due to:

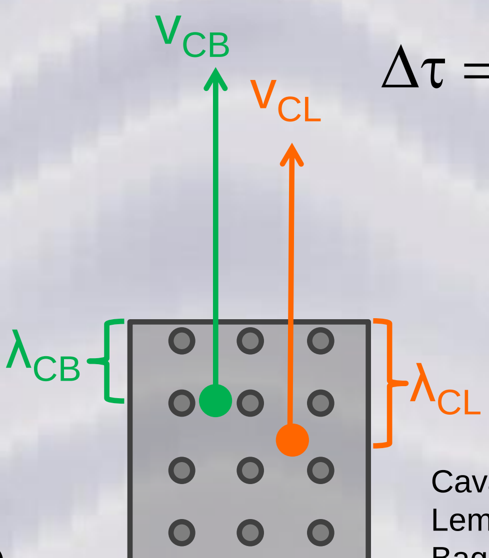
…the difference in the spatial localization of the **initial-state** wave functions of CB and core-level electrons



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:



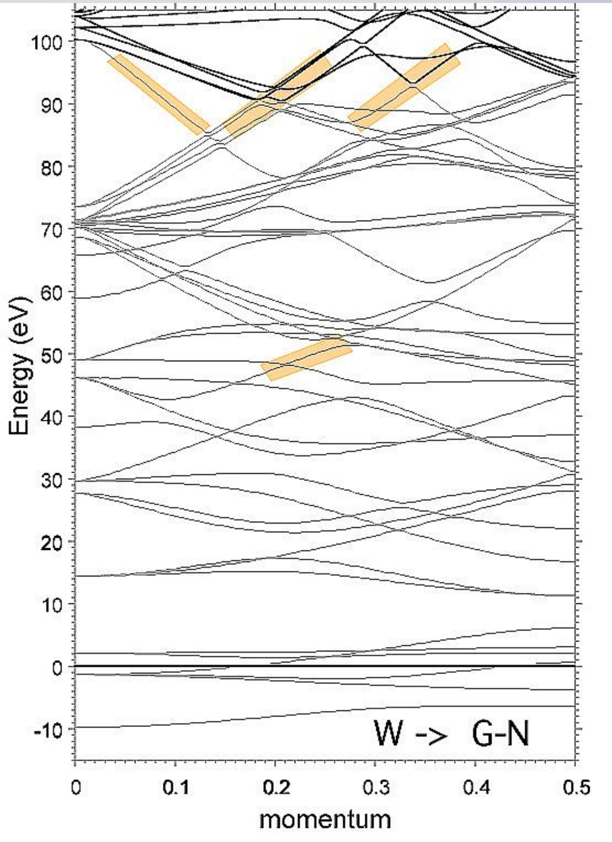
$$\Delta\tau = \lambda_{CL}/v_{CL} - \lambda_{CB}/v_{CB}$$

Impact of **final-state band-structure**?

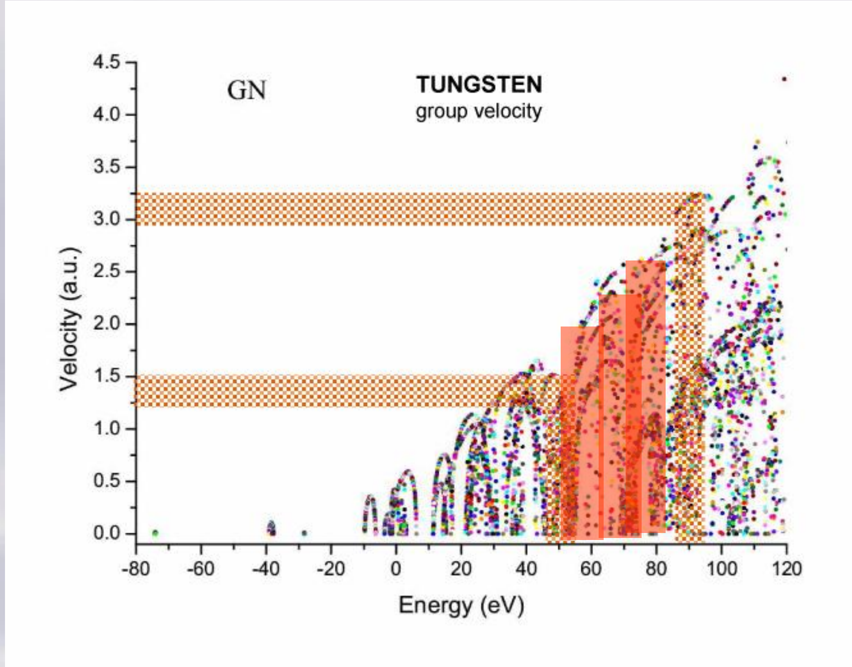
Cavalieri *et al*, Nature **449**, 1029 (2007)
Lemell *et al*., PRA **79**, 062901 (2009)
Baggesen *et al*., PRA **80**, 030901 (2009)

final-state bandstructure???

calculated W bandstructure



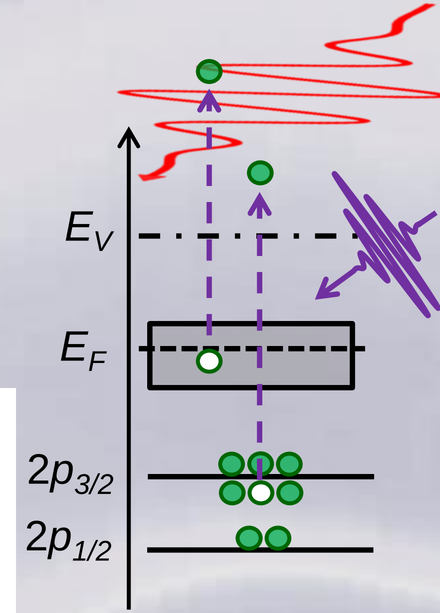
calculated group velocities for hν = 95 eV

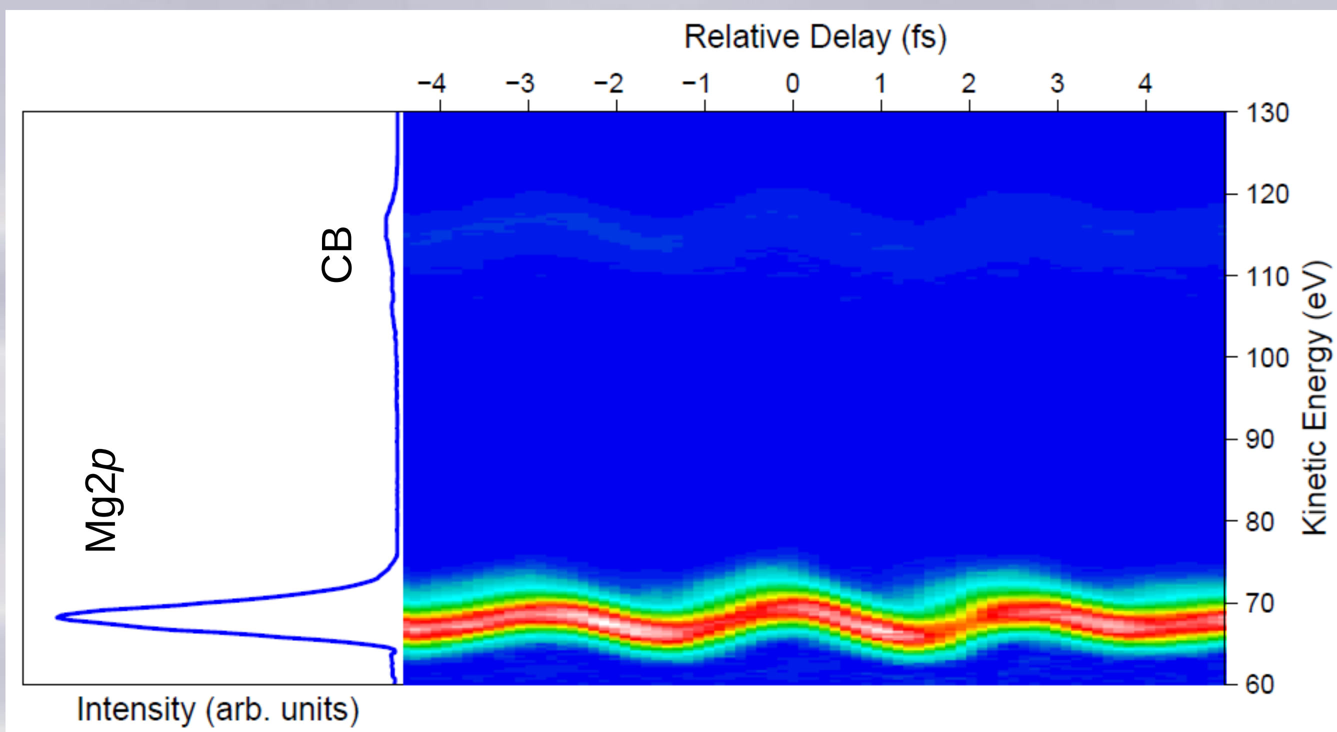


- Conduction-band electrons (from near the Fermi energy) are excited into upper-conduction band states where their group velocity is ~twice that of the excited 4*f*-state electrons
- IMFP of 4f-state electrons is approximately 1 Angstrom deeper than for conduction-band electrons
- Combined effect leads to a estimated delay of ~100 as in agreement with observation

Attosecond Streaking of Mg(0001)

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

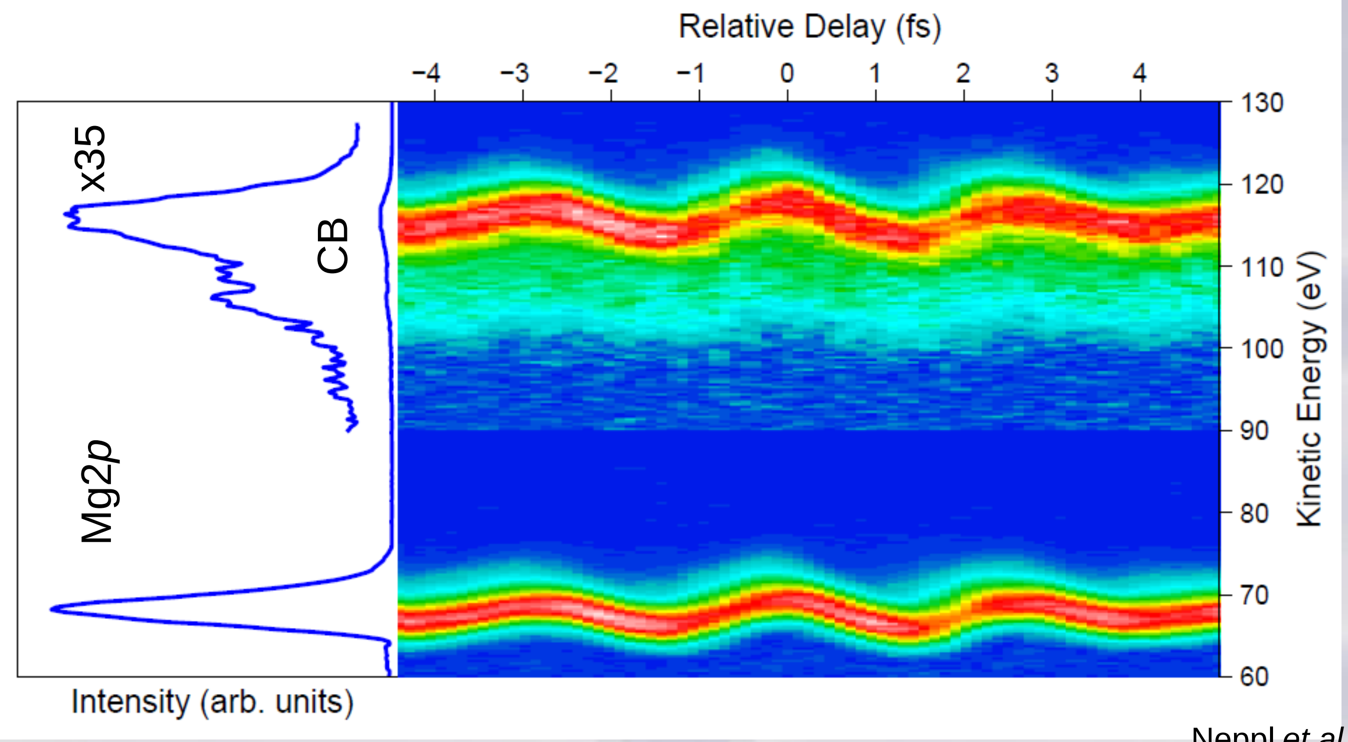


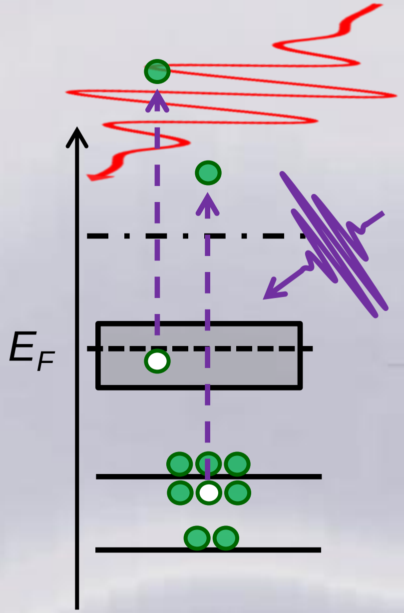


Neppl *et al*, PRL **109**, 087401 (2012)

Attosecond Streaking of Mg(0001)

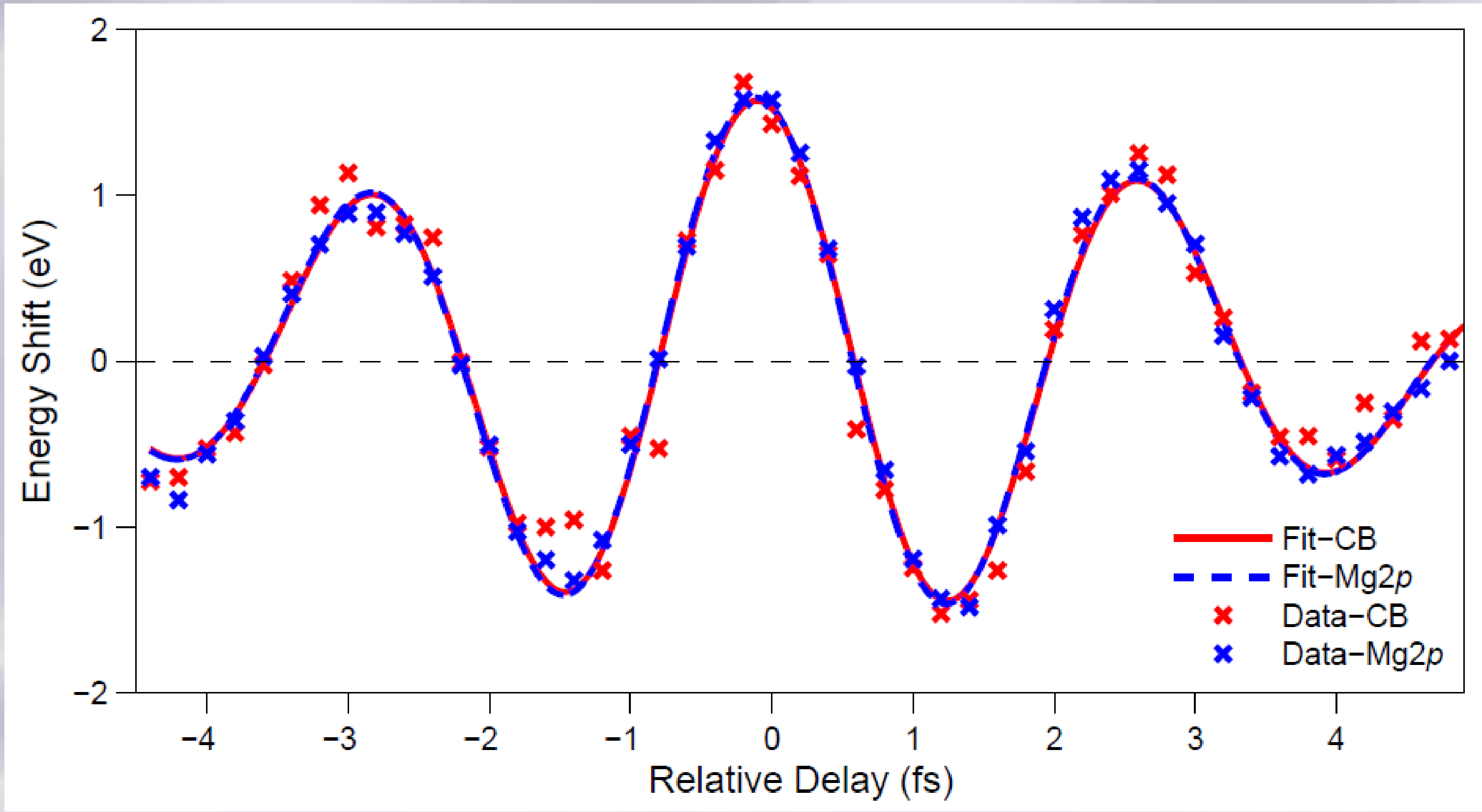
- Ideal test case: truly delocalized CB electrons & localized *2p* core-level electrons
- **Free-electron-like final-state band structure**
- Challenging: reactive material & low CB density of states





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

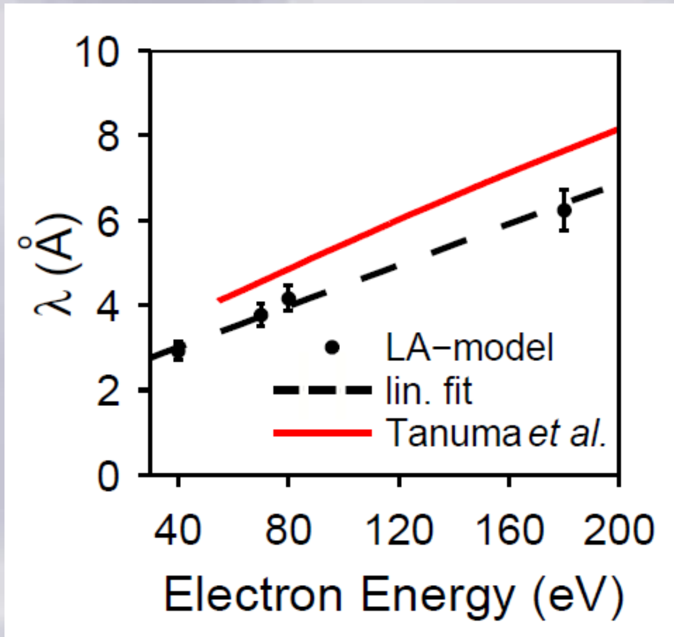
Attosecond Streaking of Mg(0001)



- Vanishing time delay between the release of CB and Mg2*p* electrons
- Strong discrepancy with theories that emphasize the role of the initial-state localization ($\Delta\tau \rightarrow$ 100as !)

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

Attosecond Streaking of Mg(0001)

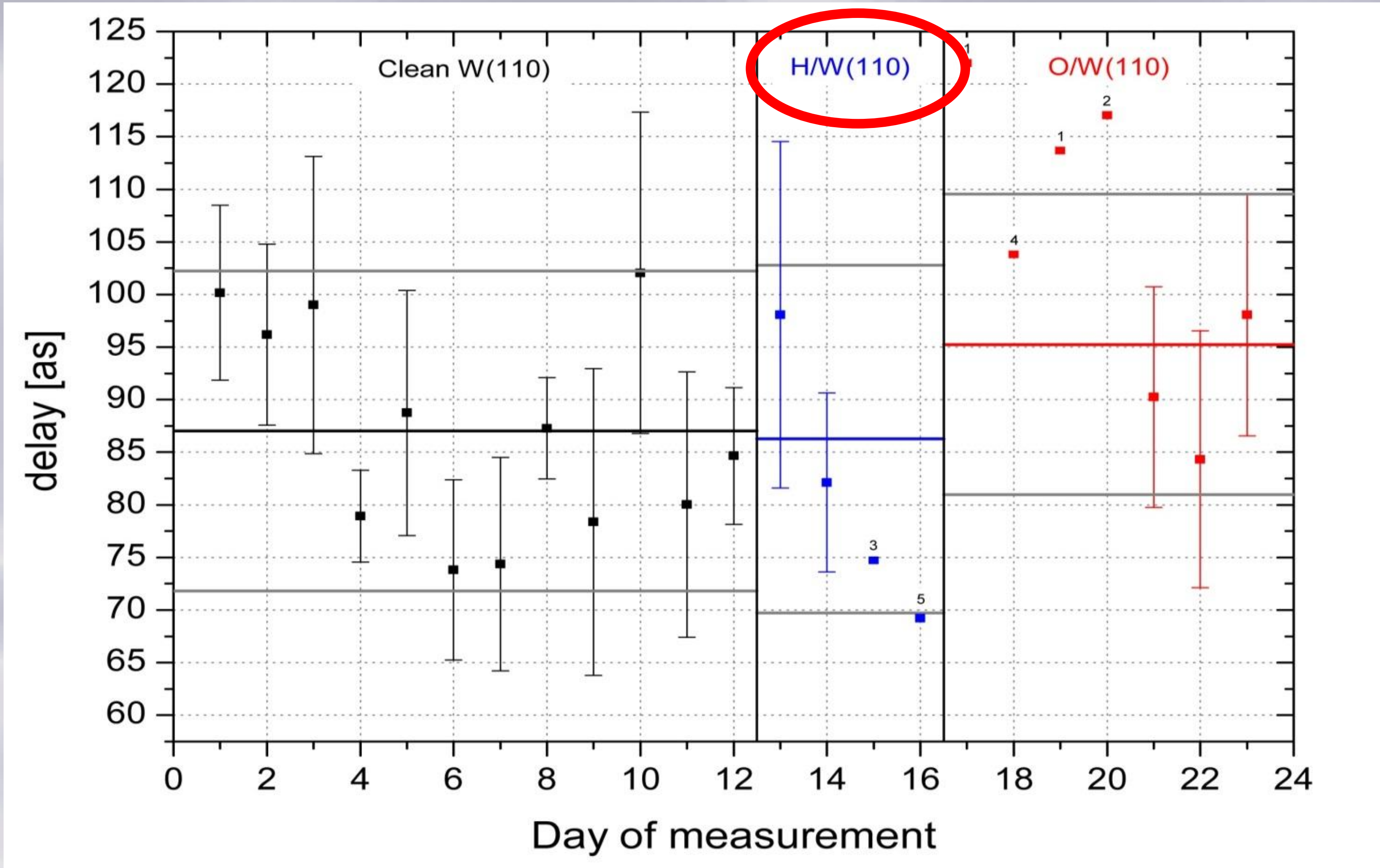


- Perfect agreement with the free-electron propagation limit:
  $\Delta\tau = \lambda_{CB}/v_{CB} - \lambda_{2p}/v_{2p} =$ **0as**

- Cancelation of the electrons' velocities and their average escape depths

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

Surface states?



H is known to strongly quench the contribution of surface states to the CB feature of W

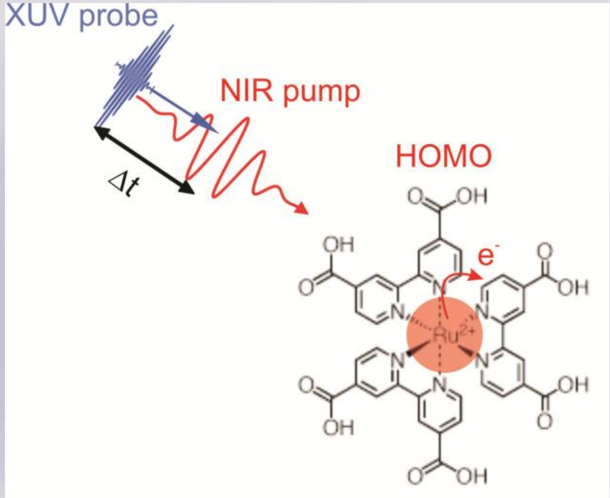
ongoing and future work…

different types of
pump / probe experiments on
molecules

IR pump / XUV probe

candidates: Ru/Os-bipyridine complexes

optical excitaion:
metal-to-ligand charge transfer

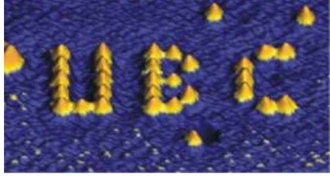


Ground state

Excited state

quasi-oxidation of metal center
→ **dynamic chemical shift**

Sample preparation



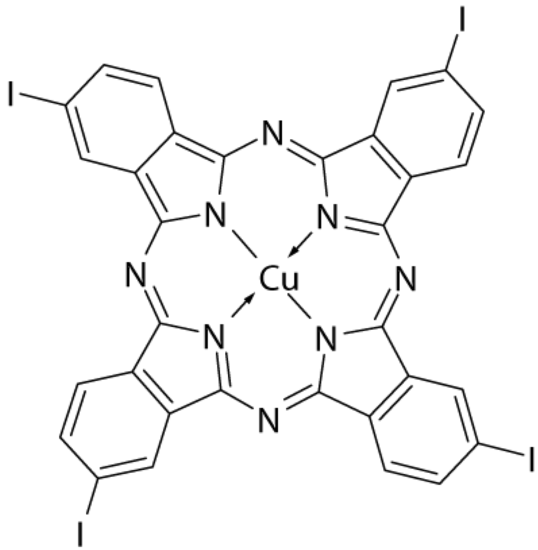
New sample preparation: Sublimation and vapour deposition of molecule

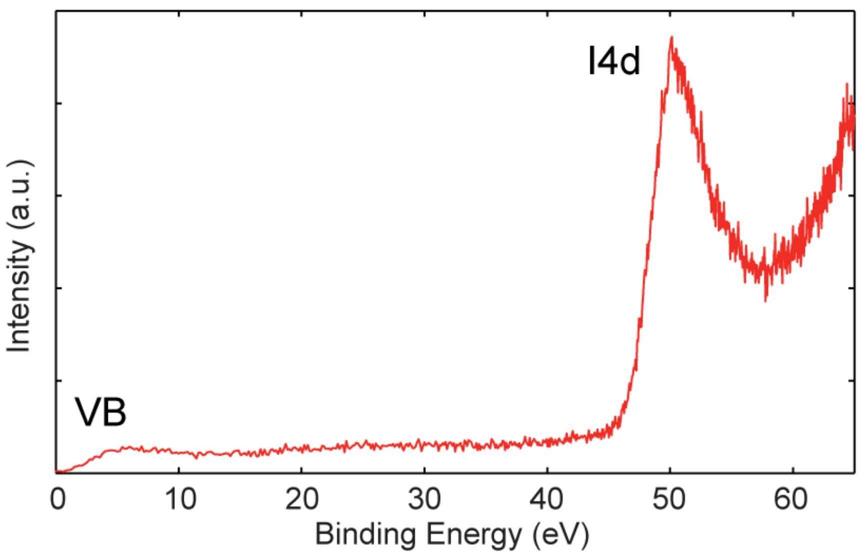
>Uniform thin layers
>Mono- and sub-monolayers possible
>No native oxide on silicon

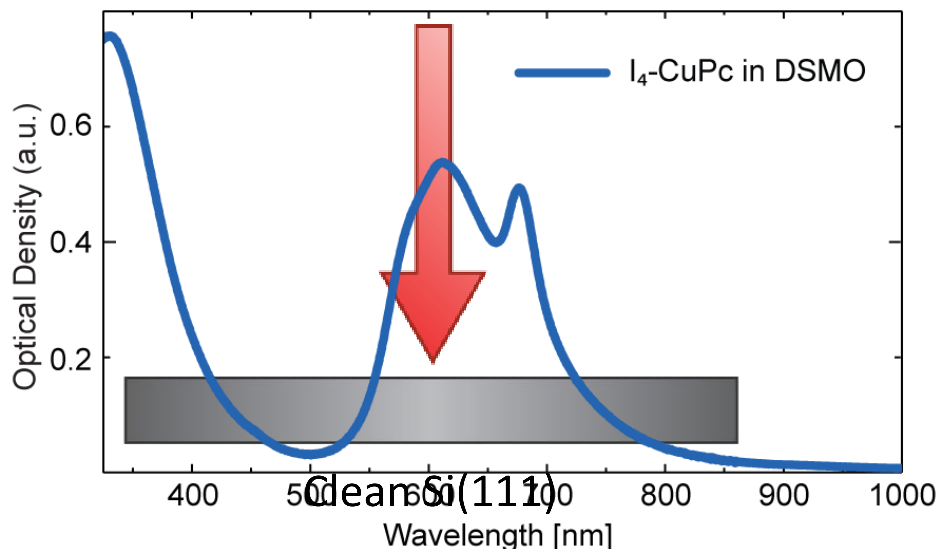
→ Strongly improved signal to noise ratio

→ Interfacial charge transfer accessible

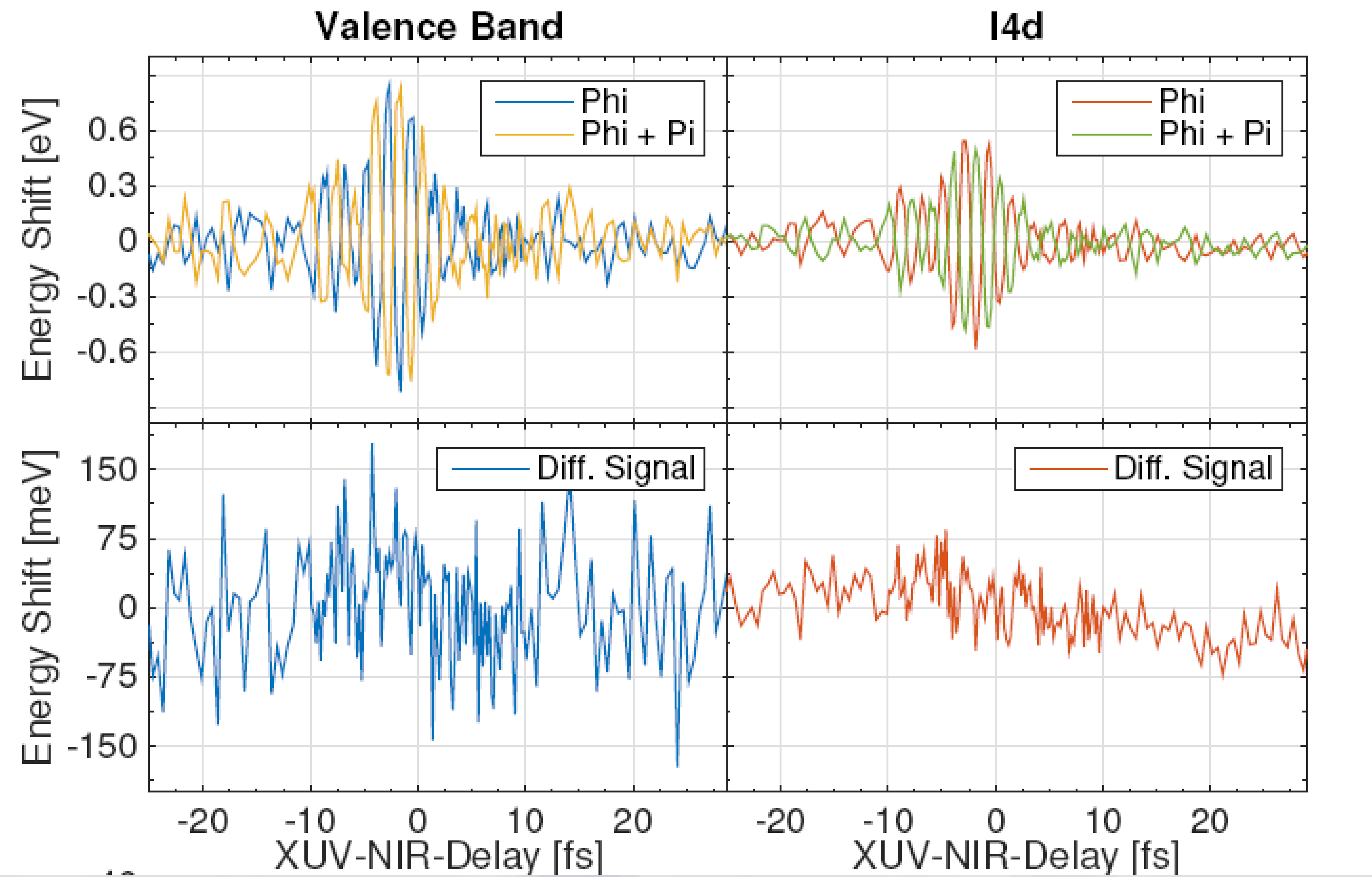
Tetraiodo-Copper(II)-Phthalocyanine



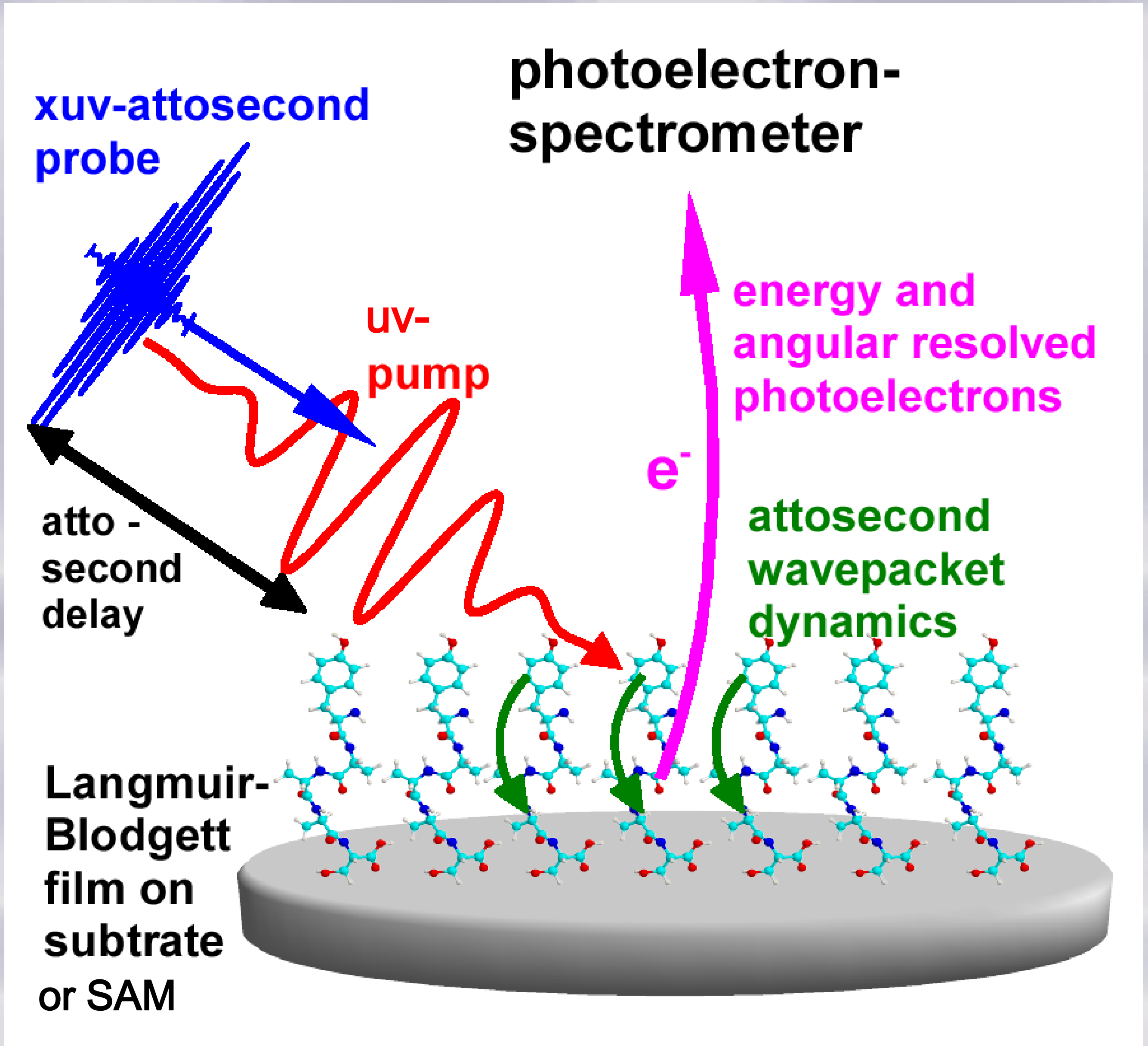




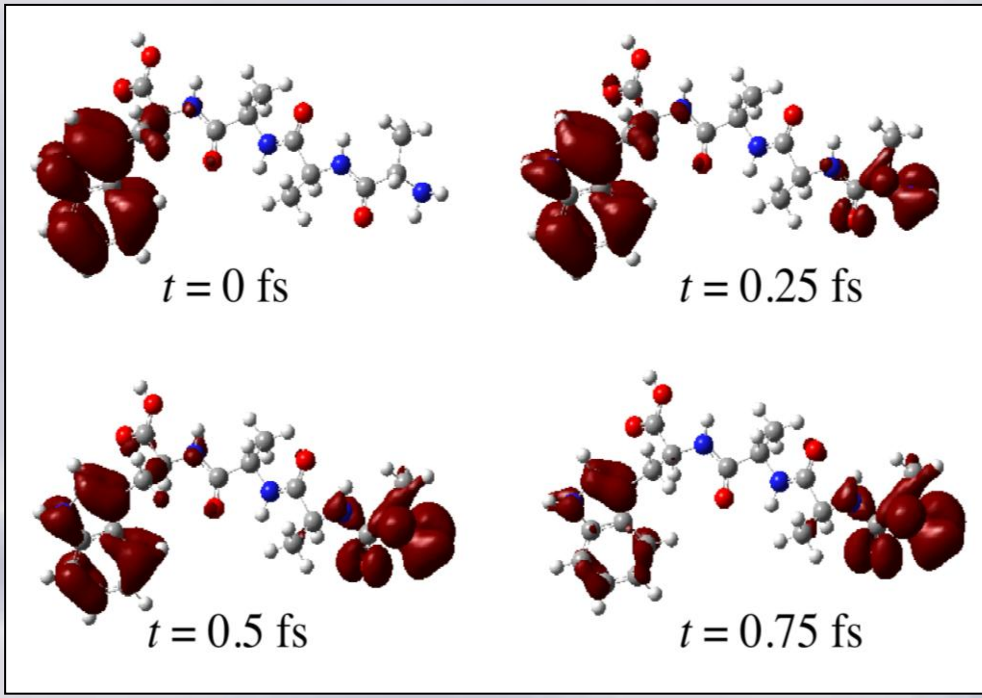
„Differential streaking“

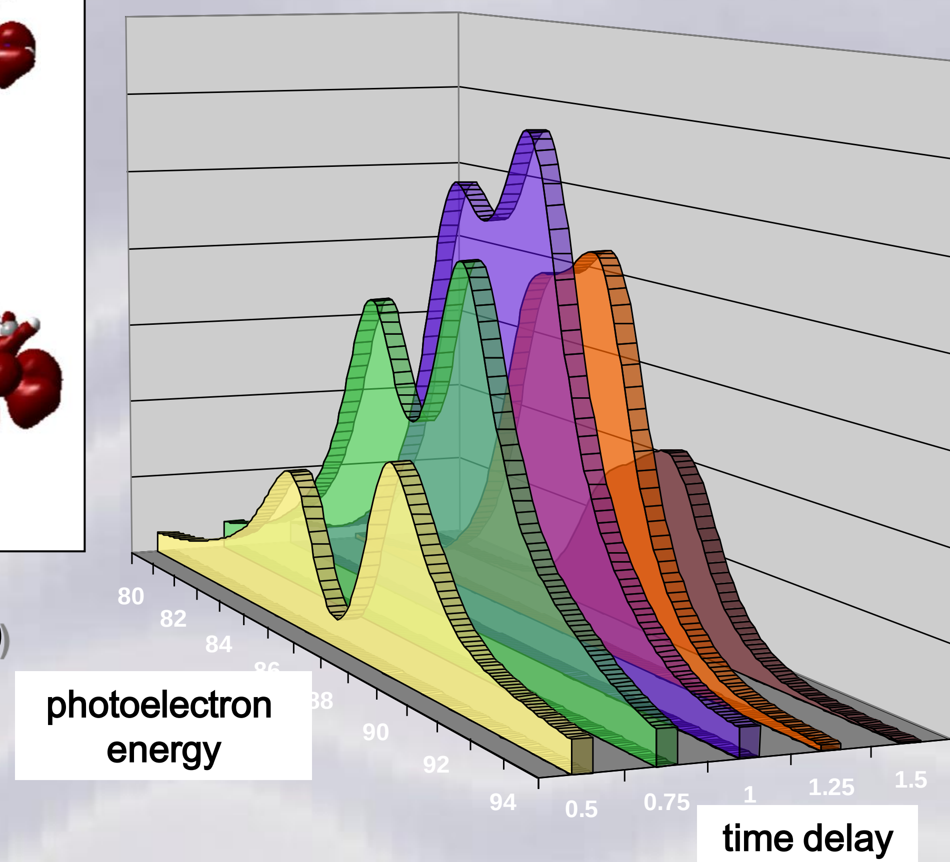


probing charge transfer/migration in complex (bio-)molecules?



probing charge transfer/migration in complex (bio-)molecules?





F. Remacle, R. D. Levine, *PNAS* **103,** 6793 (2005)

Colleagues & Cooperations

W. Helml	M. Jobst	T. Latka	J. Riemensberger	
K. Hütten	R. Heider	D. Hutzler	M. Wagner	H. Iglev
B. Eder	S. Schweyer	S. Prinz	L. Pedrosa	K. Oberhofer

surface dynamics: A. Kim, M. Schäffer, P. Feulner, D. Menzel, J. Barth
TU Munich, Germany

R. Ernstorfer
FHI Berlin, Germany

as- development: A. Sommer, A. Schwarz, N. Karpowicz, S. Keiber
W. Schweinberger, F. Krausz
MPQ Garching, Germany

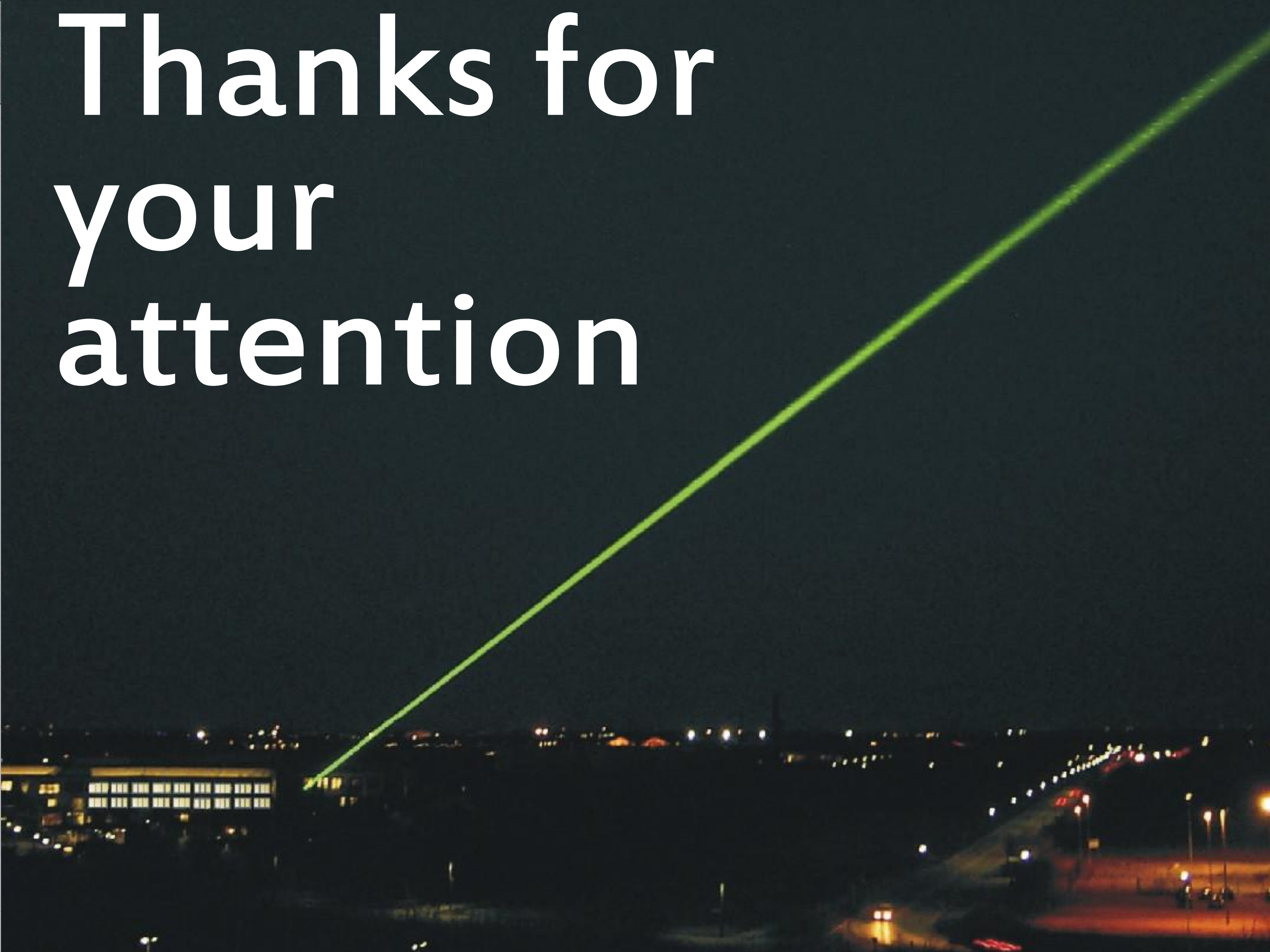
theory: M. Lupetti, A. Scrinzi
Ludwig Maximilians Univ. Munich, Germany

P. Echenique, A. Kazansky
San Sebastian, Spain

Ch. Lemell, J. Burgdörfer
Vienna University of Technology, Austria

J. Gagnon, V. Yakovlev
MPQ Garching, Germany

XUV optics: A. Guggenmos, U. Kleineberg
Ludwig Maximilians Univ. Munich, Germany


Thanks for your attention