Complementary uses of Free Electron Lasers and High-Harmonic Sources

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SCIENCE@FELs Villigen, 17-9-2014

Max-Born-Institut

Source parameter comparison FEL vs. HHG

Parameters	Flash FEL (Hamburg 2005)	Fermi (Trieste, 2010)	LCLS (Stanford, 2009)	SACLA (Hyogo, 2011)	EU XFEL (Hamburg, 2015)
E_e	1.25 GeV	1.24 GeV	13.6 GeV	8 GeV	17.5 GeV
γ	2,450	2,300	26,600	15,700	35,000
$\hat{I}$	1.3 kA	300 A	3.4 kA	3 kA	5 kA
λ_u	27.3 mm	55 mm	30 mm	18 mm	35.6 mm
N	989	216	3733	4986	4000
L_u	27 m	14 m	112 m	90 m	200 m
$\hbar\omega$	30-300 eV (4-40 nm)	20-60 eV (20-60 nm)	250 eV - 12 keV (1-50 Å)	4.5-15 keV (0.8-2.8 Å)	4-12 keV (1-3 Å)
$\lambda/\Delta\lambda_{FWHM}$	100	1000	200-500	200-400	1000
$\Delta\tau_{FWHM}$	25 fsec	85 fsec	70 fsec	30 fsec	100 fsec
$\dot{\mathcal{F}}$ (ph/pulse)	3×10^{12}	5×10^{12}	2×10^{12}	5×10^{11}	10^{12}
rep rate	5 Hz	10 Hz	120 Hz	60 Hz	5 Hz/27 kHz
$\hat{P}$	1 GW	1 GW	25 GW	30 GW	20 GW
L	260 m	200 m	2 km	710 m	3.4 km
Polarization	linear	variable	linear	linear	variable (?)
Mode	SASE	Seeded (3ω Ti: saphire)	SASE	SASE	SASE

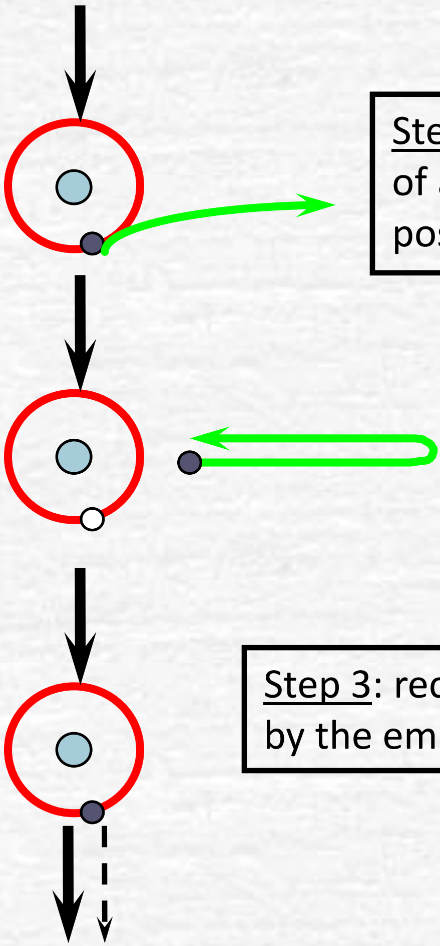
Flash II, Fermi II, SLS FEL, LCLS II, ...

FreeElectronLasersChart_June2014.ai

Prof. David Attwood / Basel FEL Conference August 25-29, 2014

XUV/X-ray pulses from High Harmonic Generation

Intense near-infrared femtosecond laser

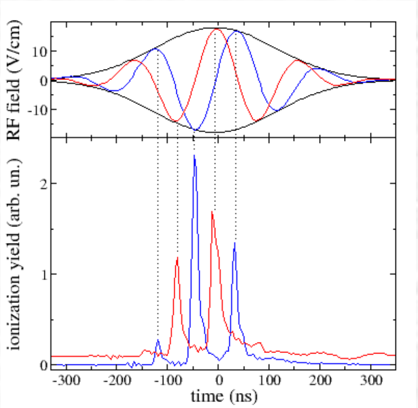


Step 1: ionization and removal of an electron from the positive ion core

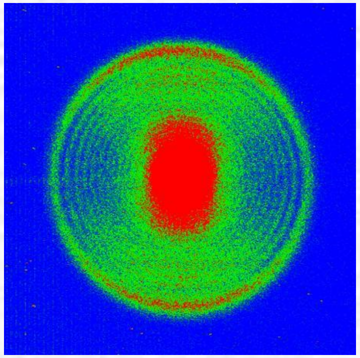
Step 2: acceleration of the electron in the oscillatory laser field

Step 3: recombination, accompanied by the emission of an XUV photon

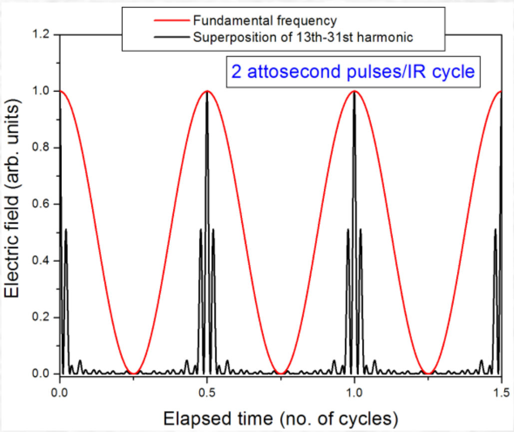
Intense near-infrared femtosecond laser + XUV radiation



Guertler et.al., Phys. Rev. Lett. 92, 063901 (2004)



Nicole et.al., Phys. Rev. Lett. 88, 133001 (2002)

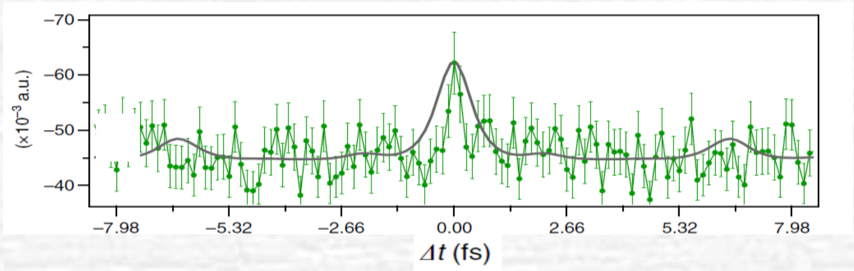


Source parameter comparison FEL vs. HHG

A sampling of what is possible using HHG

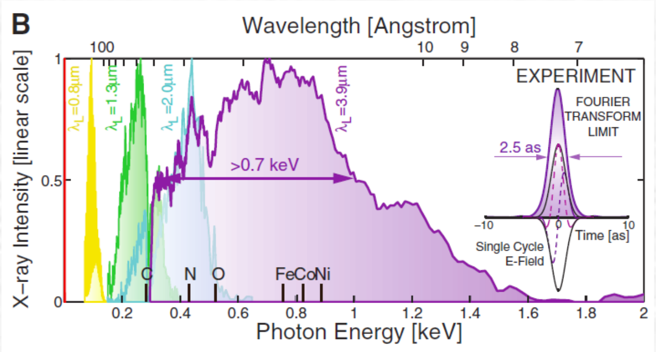
K. Midorikawa and co-workers, Nat. Comm. 4, 2691 (2013)

9 mJ, 800 nm, 30 fs + 2.5 mJ, 1300 nm, 35 fs
→ 1.3 μJ pulse in 28-35 eV, 500 as



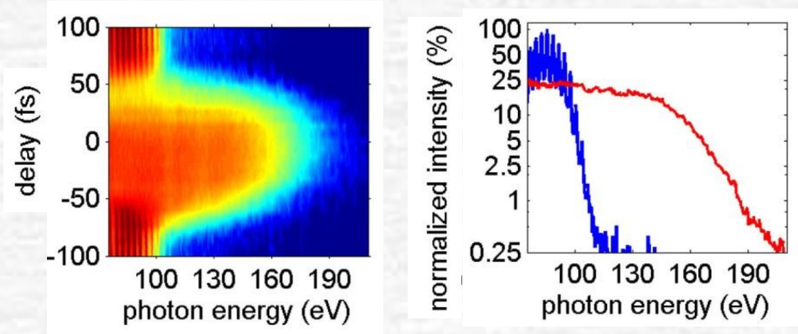
A. Baltuska and co-workers, Science 336, 6086 (2012)

10 mJ, 3.9 μm, 80 fs → 10^5 photons per shot in a fractional bandwidth of 1% at 1 keV



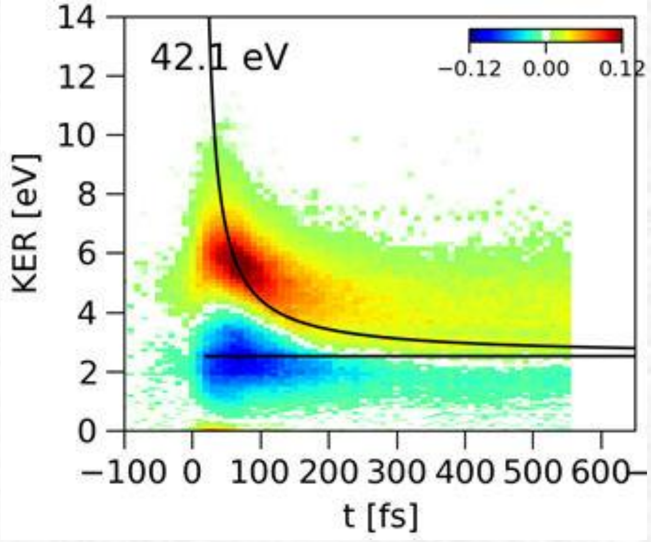
Our own work (B. Schütte et al., in preparation)

4×10^{14} *W/cm²* , 800 nm, 50 fs + 9×10^{14} *W/cm²* 1300 nm, 50 fs → 10^7 photons per shot between 70 and 160 eV

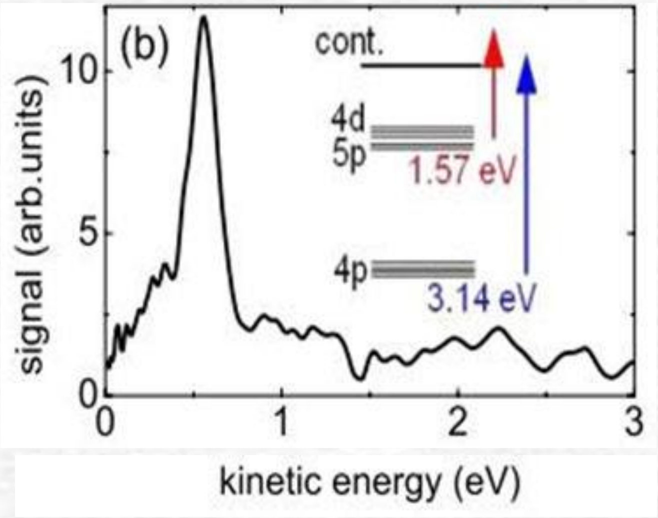


Contents of this talk

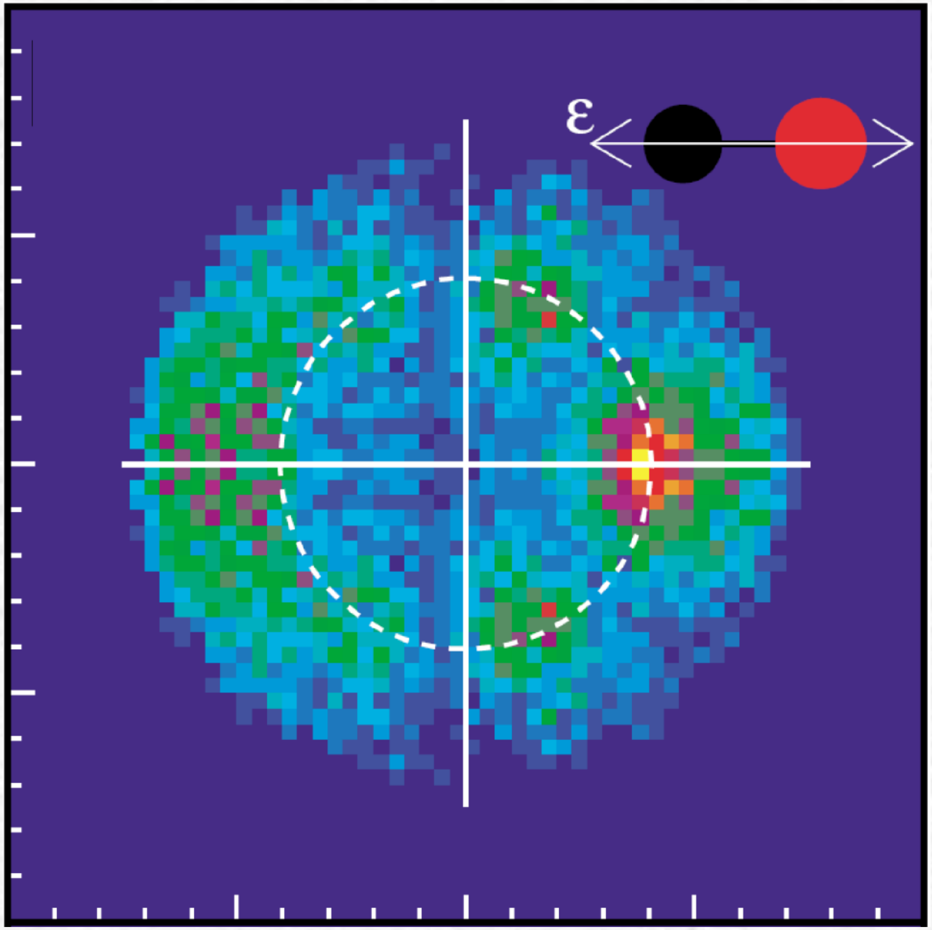
- **Pump-probe spectroscopy on molecules**
  - Using photoelectron holography for time-resolved measurements of molecular structure
  - XUV-induced molecular dynamics



- **Pump-probe spectroscopy on clusters**
  - Probing charging and dissociation of highly-excited rare gas clusters



Holography in XUV/X-ray photoionization



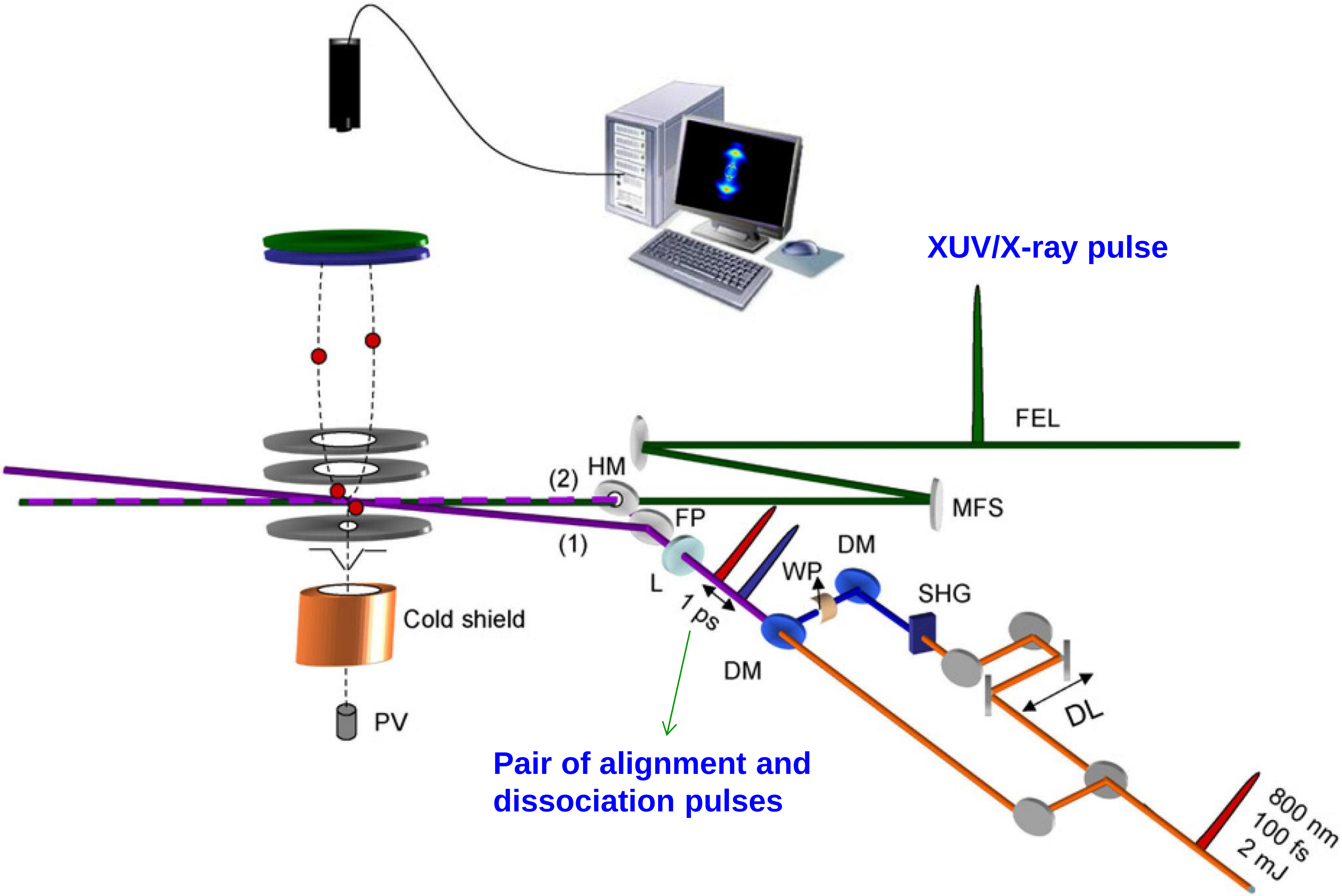
C(1s) core-shell photo-emission from CO using 294 to 326 eV radiation.

Away from the Carbon atom (black) the angular distribution is relatively unstructured.

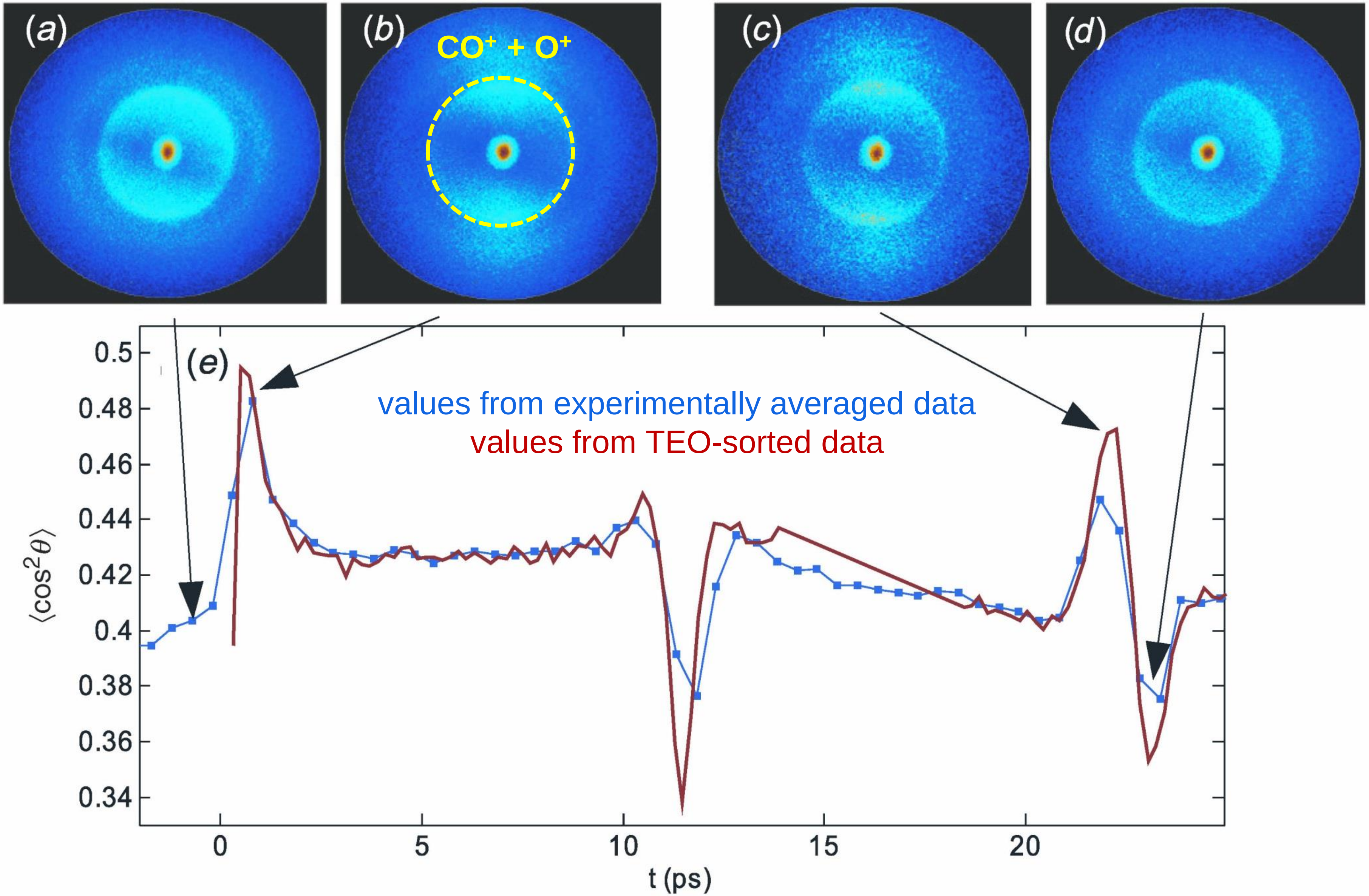
In the direction of the Oxygen atom (red) a holographic structure is observed.

Landers *et al*, Phys. Rev. Lett. 87, 013002 (2001)

Pump-probe experiments at the FLASH FEL



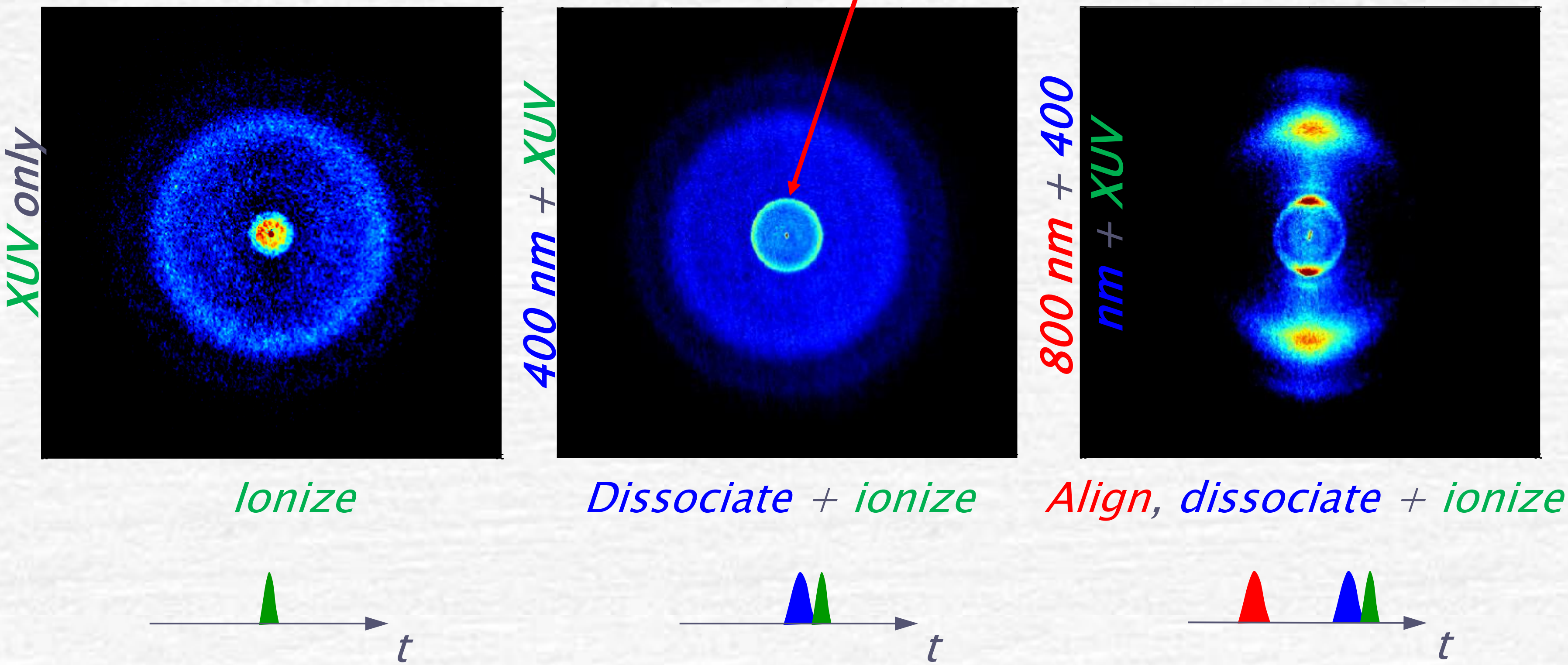
Dynamic alignment of CO_2 at the FLASH FEL



P. Johnsson et al., J. Phys B 42, 134017 (2009)

Dissociation of aligned Br_2 at the FLASH FEL

13 nm ionization **400 nm dissocation** **800 nm alignment**

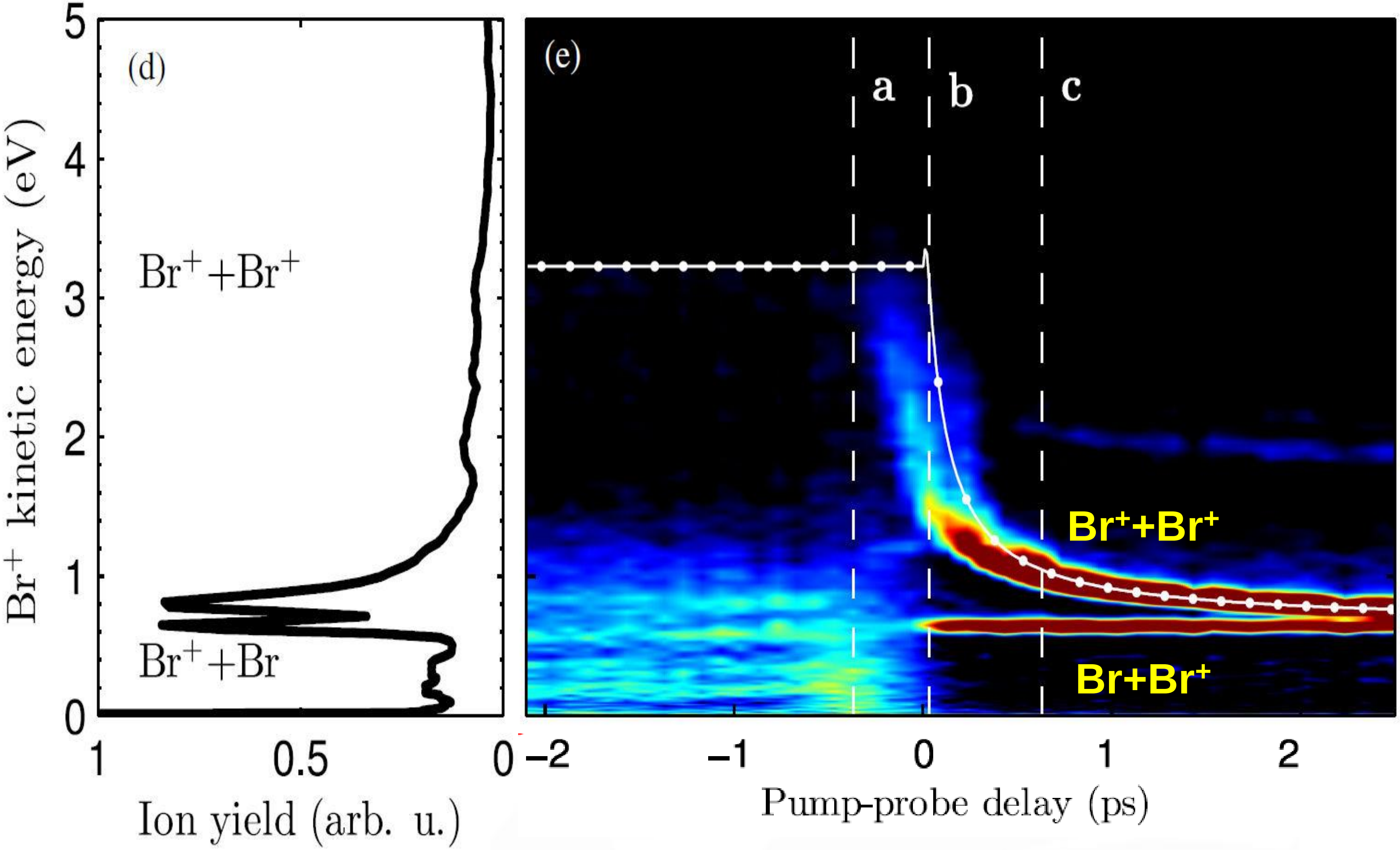


Implementation of protocol for three-color alignment-pump-probe experiments

FEL-IR/UV overlap on the basis of dissociative ionization of H_2

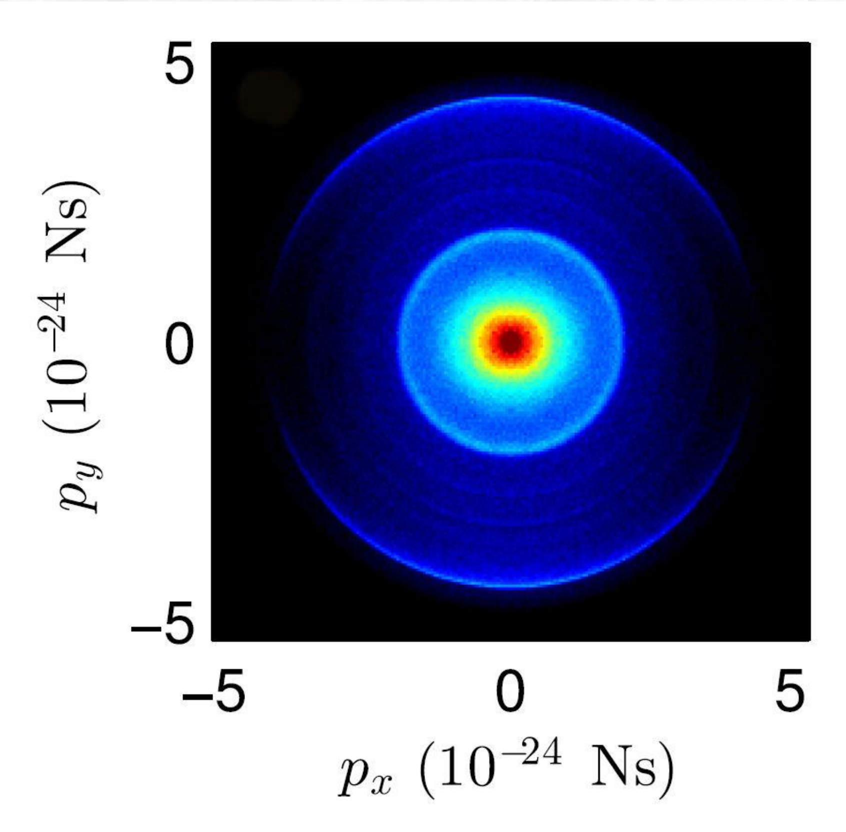
N. Berrah et al., Journal of Modern Optics 57, 1015 (2010)

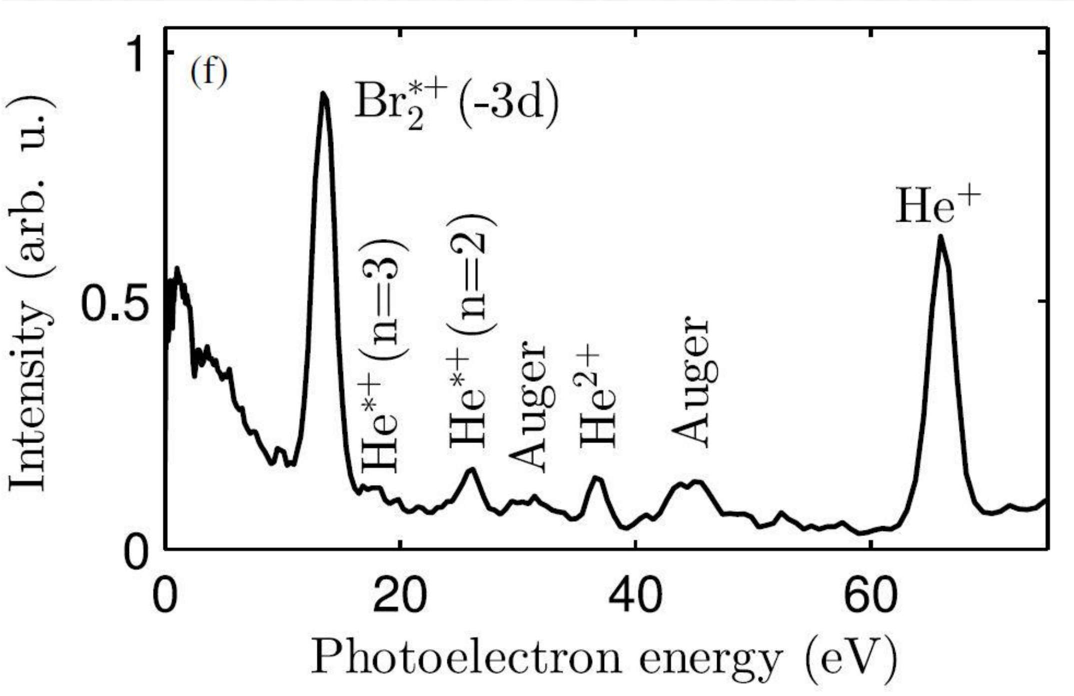
Dissociation of aligned Br_2 at the FLASH FEL



A. Rouzée al., J. Phys. B 46, 164029 (2013)

Dissociation of aligned Br_2 at the FLASH FEL



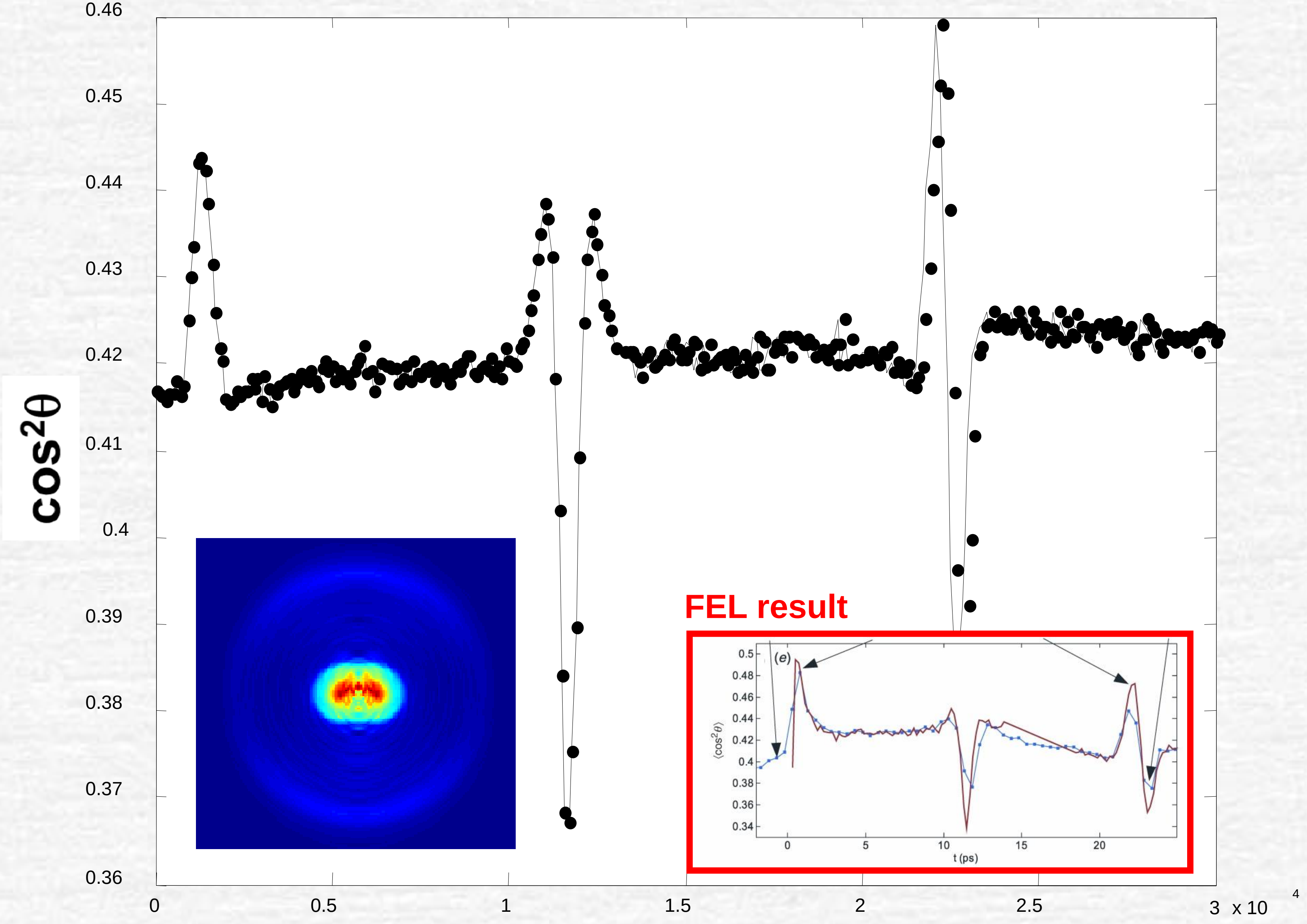


Demonstrated all the essential ingredients required for time-dependent photoelectron holography experiment: alignment, dissociation, photoelectron imaging, three-pulse pump-pump-probe scans

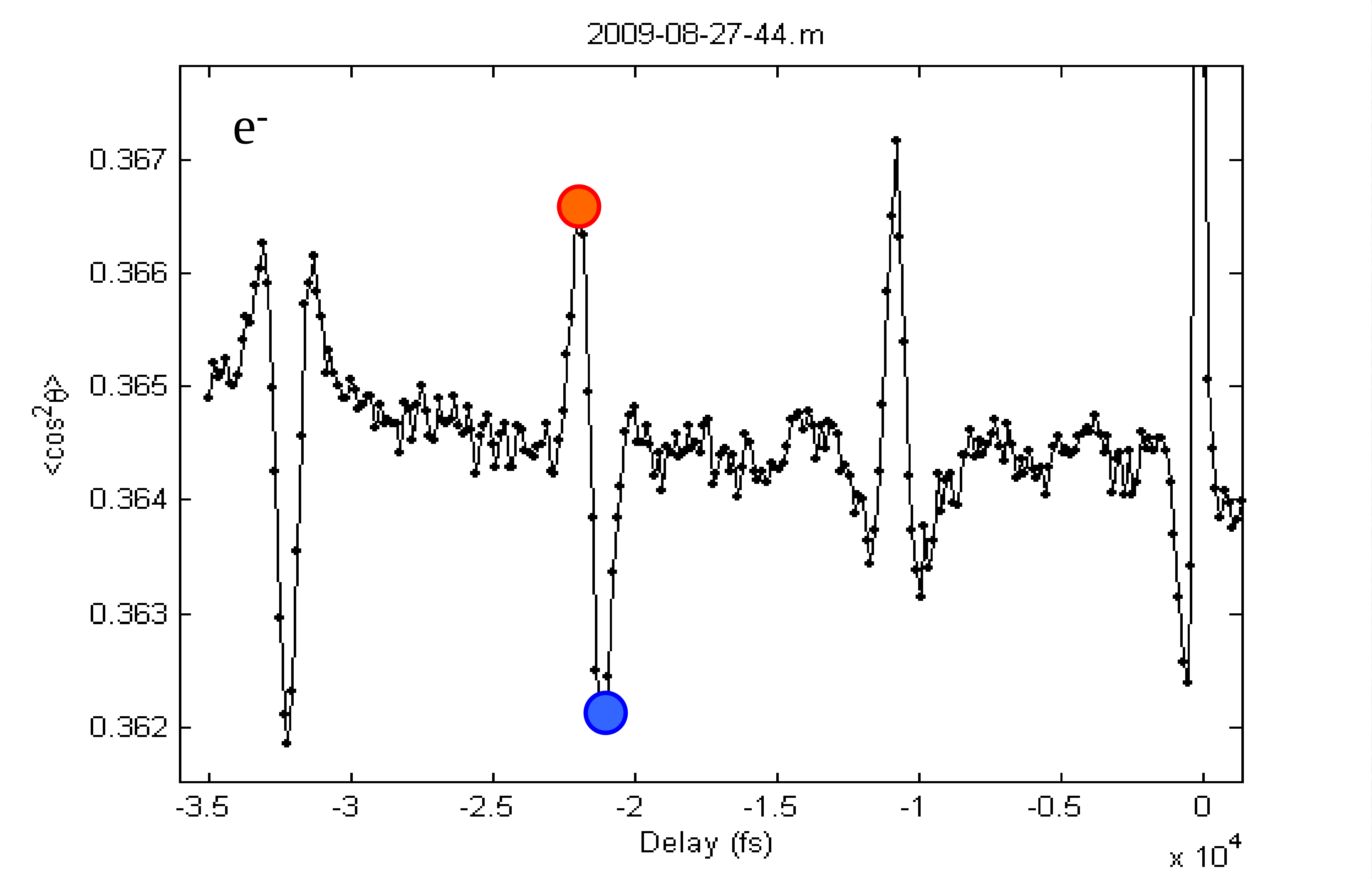
But: no successful photoelectron delay scans yet

A. Rouzée al., J. Phys. B 46, 164029 (2013)

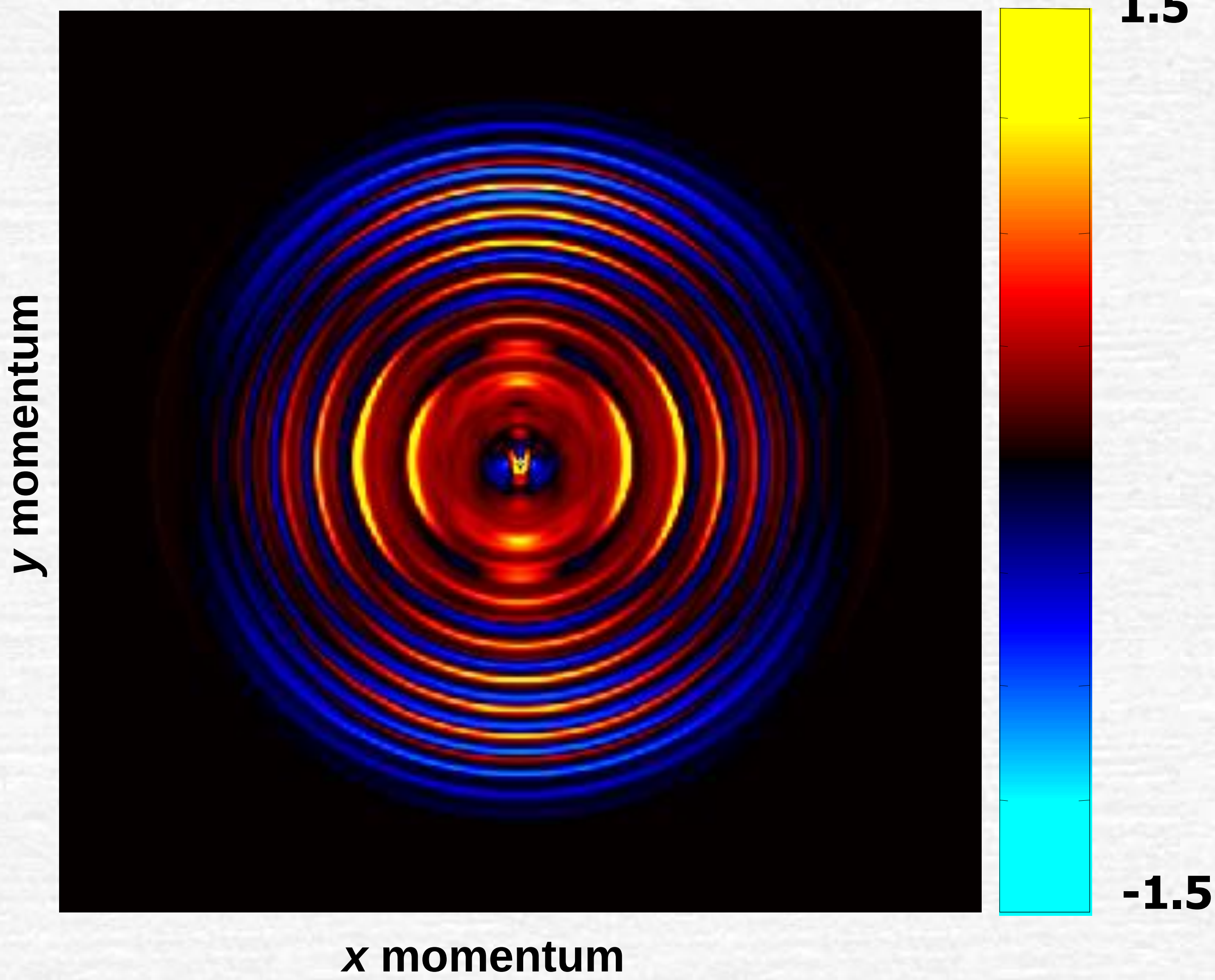
Dynamic alignment of CO_2 probed with HHG source



Photoelectron angular distributions from aligned CO_2 molecules

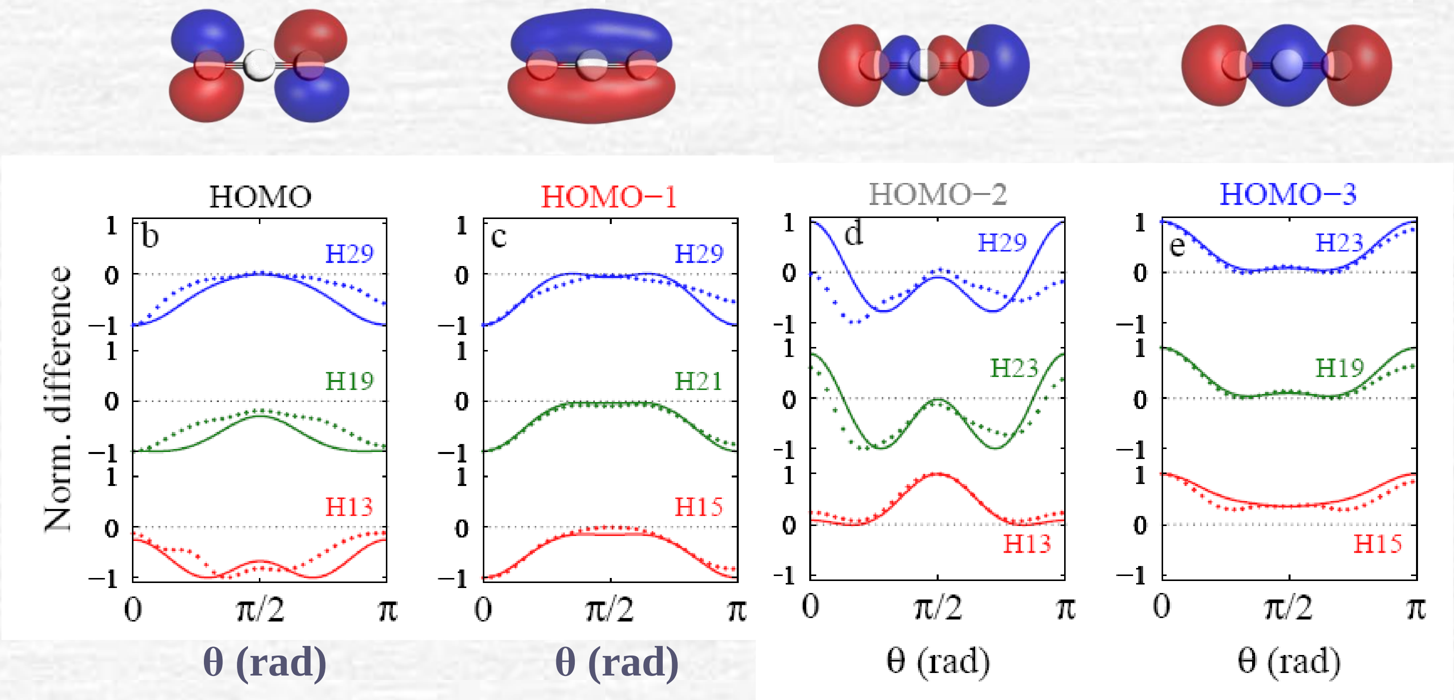


Differential PAD: alignment – anti-alignment



F. Kelkensberg et al., PRA 84, 051404 (R) (2011)

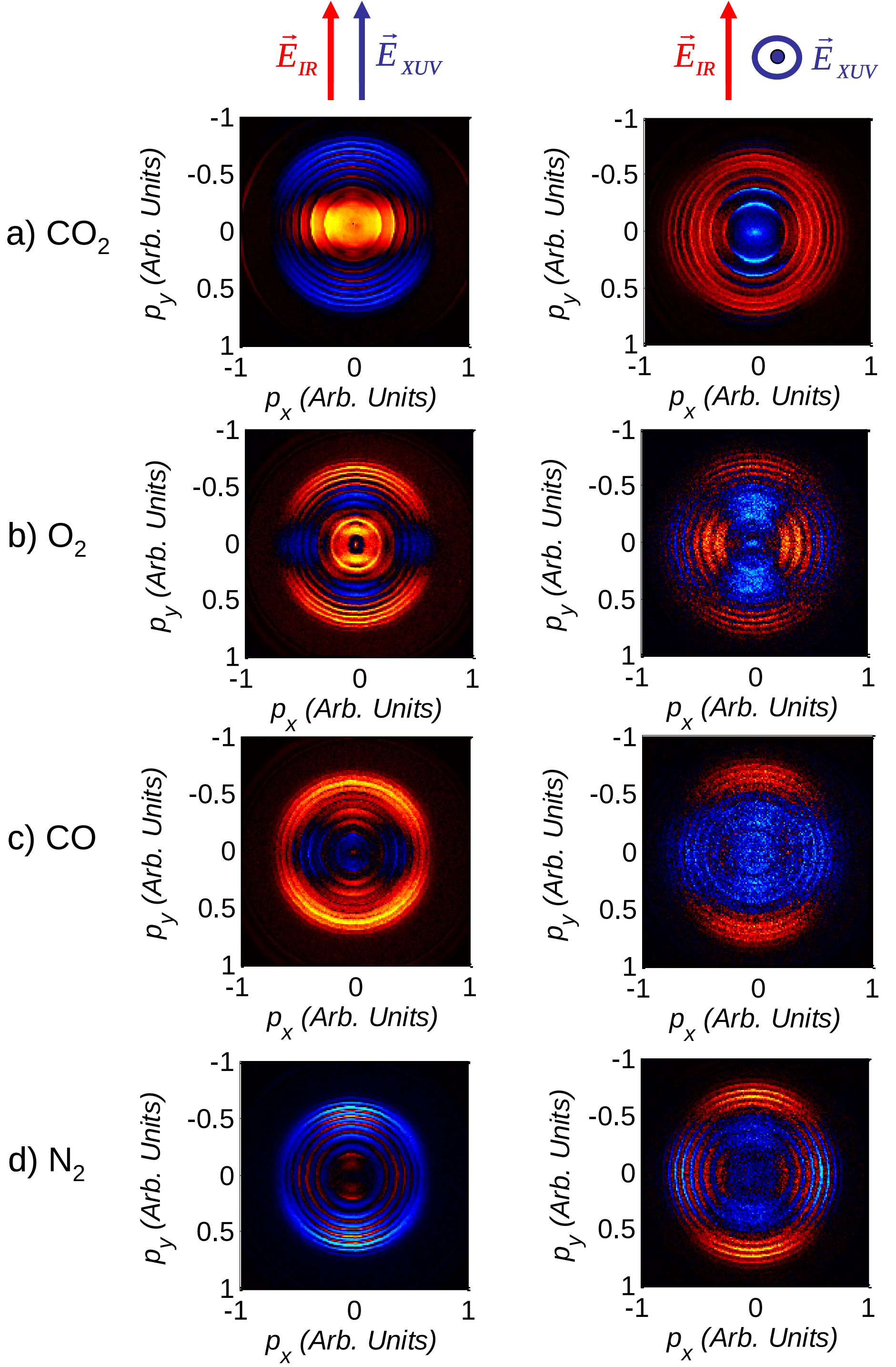
Angular distributions evolve with energy

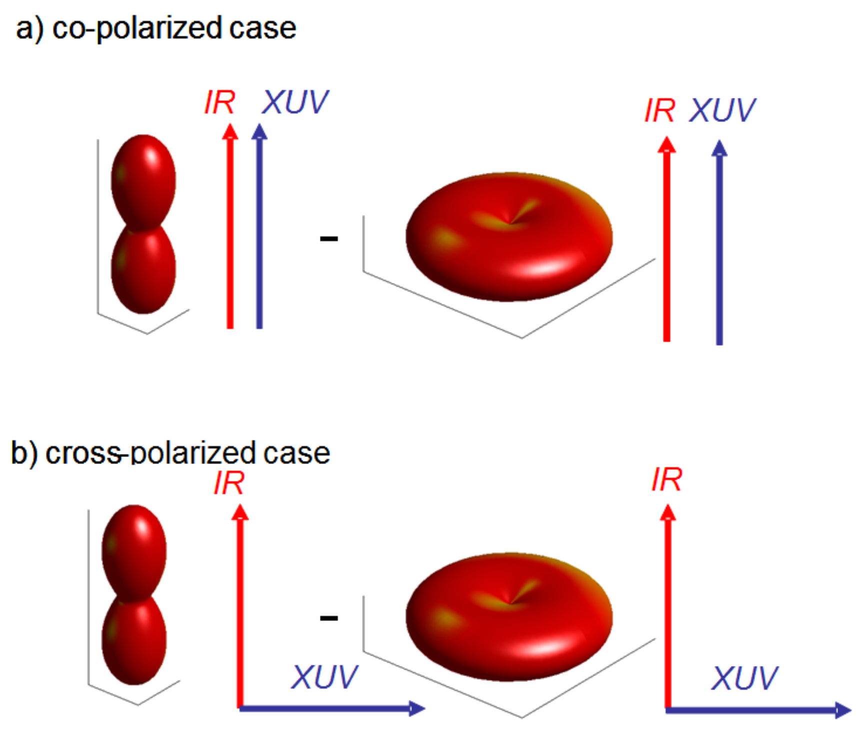


1. **Angular distribution sensitive to electronic structure**
2. **Energy dependence from interaction with molecular Coulomb field → molecular structure**

F. Kelkensberg et al., PRA 84, 051404 (R) (2011)

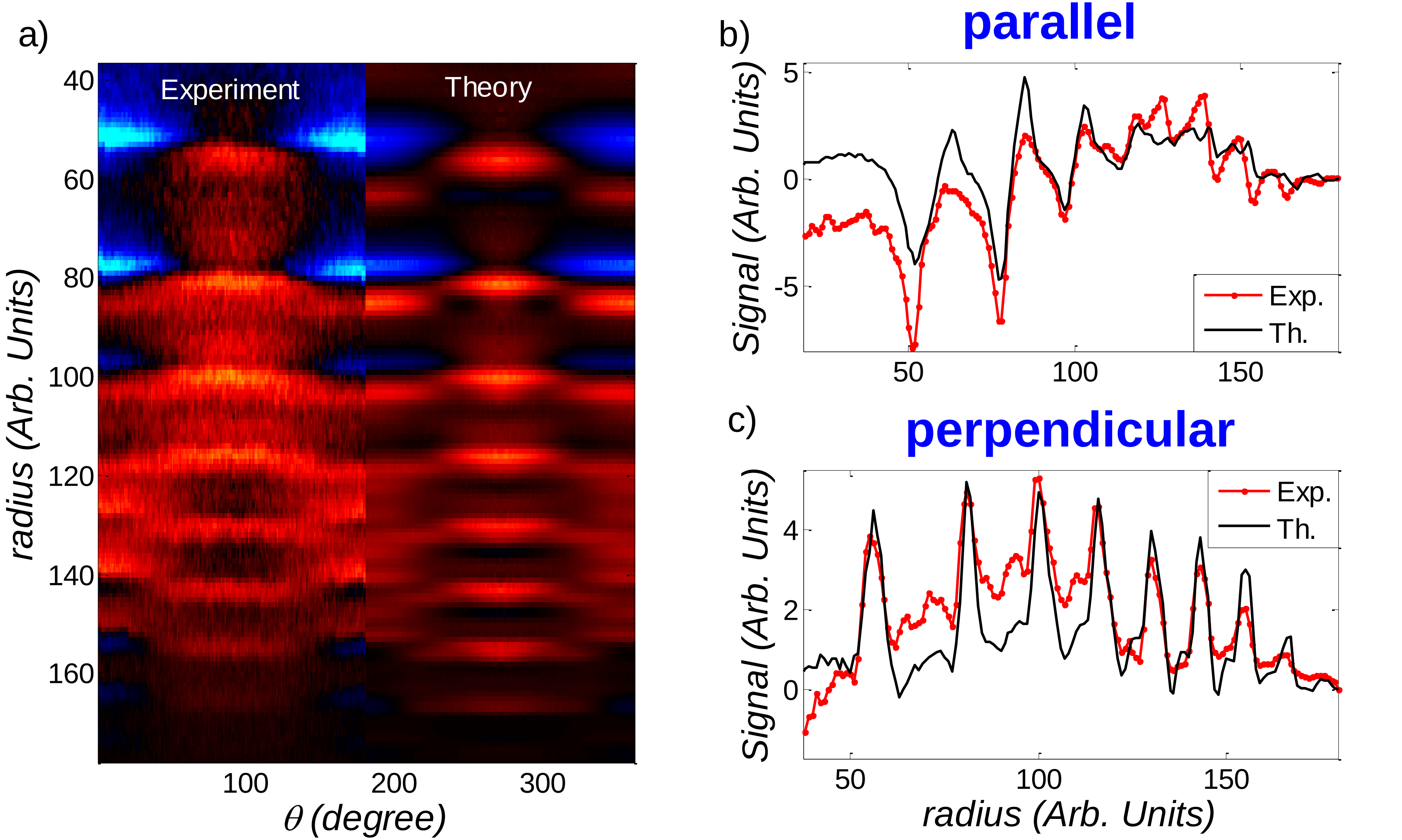
Going from co-polarized to cross-polarized pulses





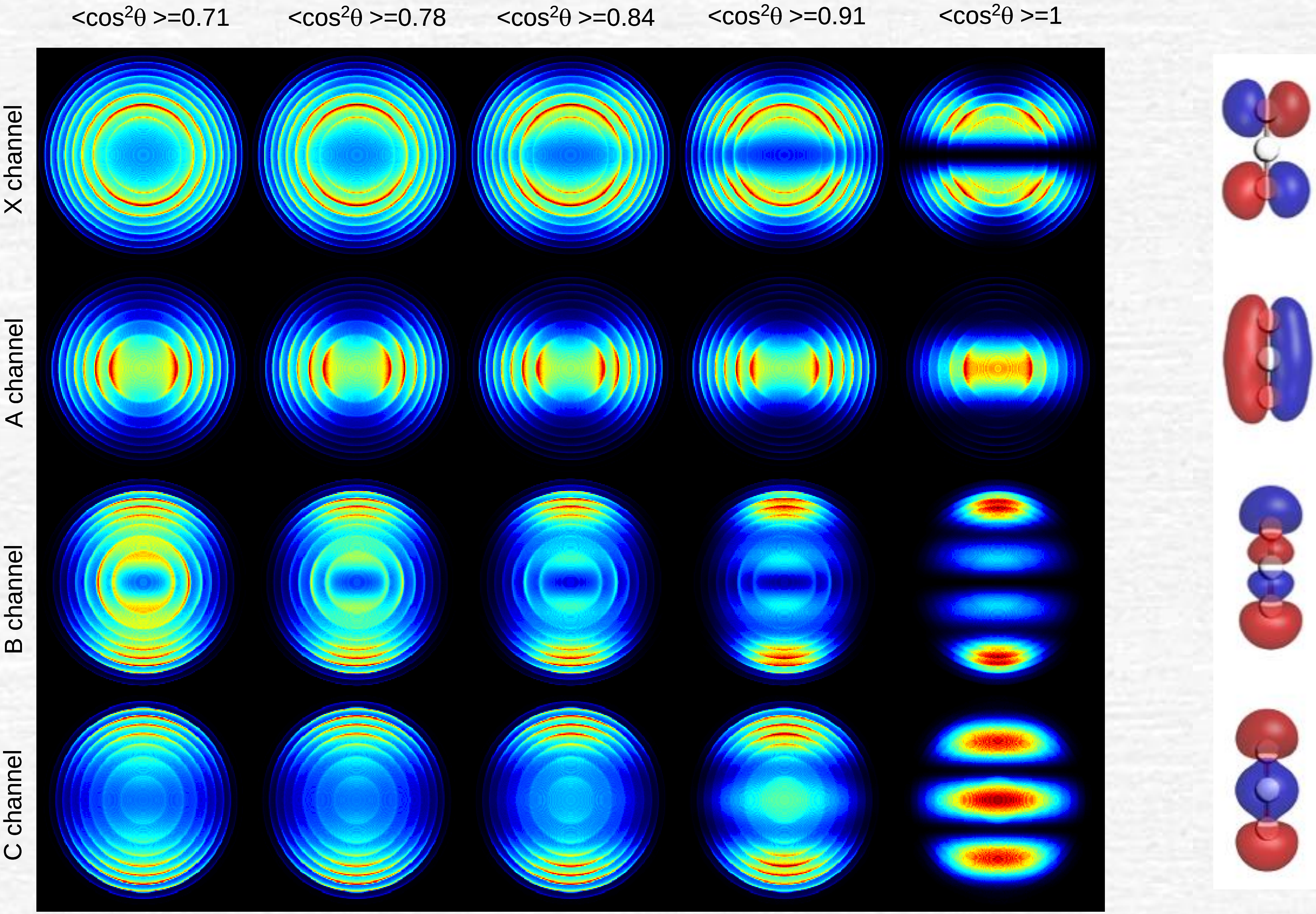
A. Rouzée et al., J. Phys. B 47, 124017 (2014)

Comparison with R-matrix calculations for CO_2



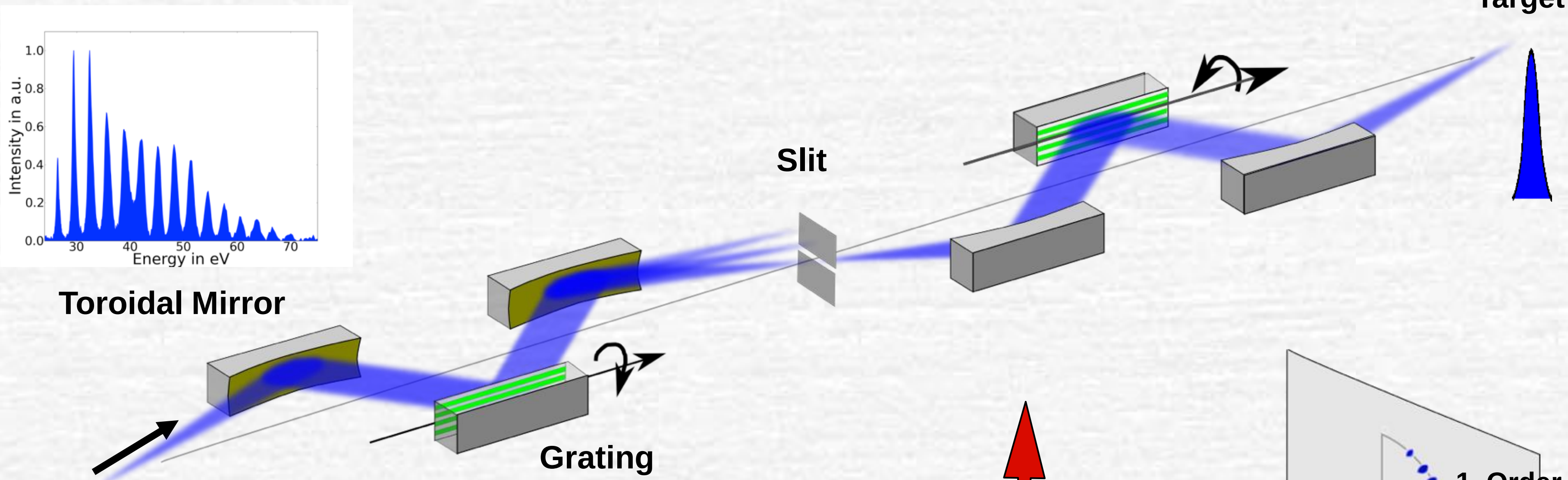
A. Rouzée et al., J. Phys. B 47, 124017 (2014)

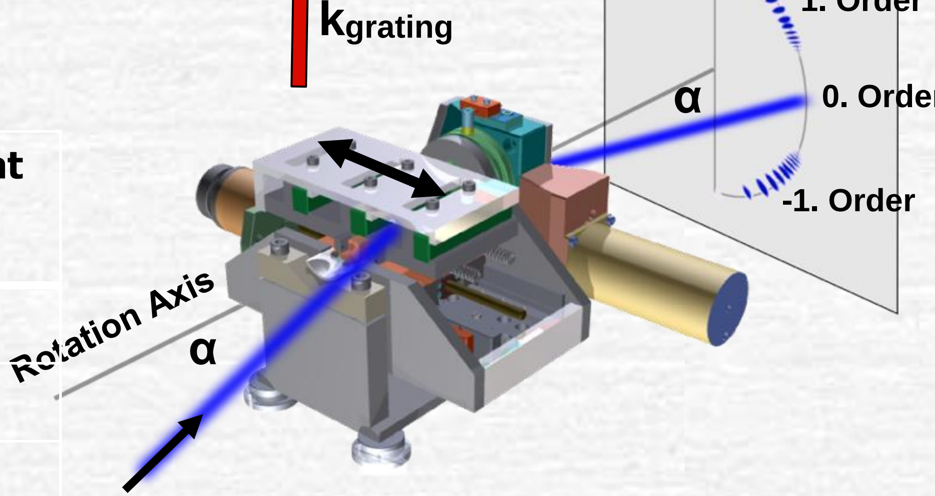
Predictions for better alignment



A. Rouzée et al., J. Phys. B 47, 124017 (2014)

Selecting single harmonics with a time-compensating monochromator

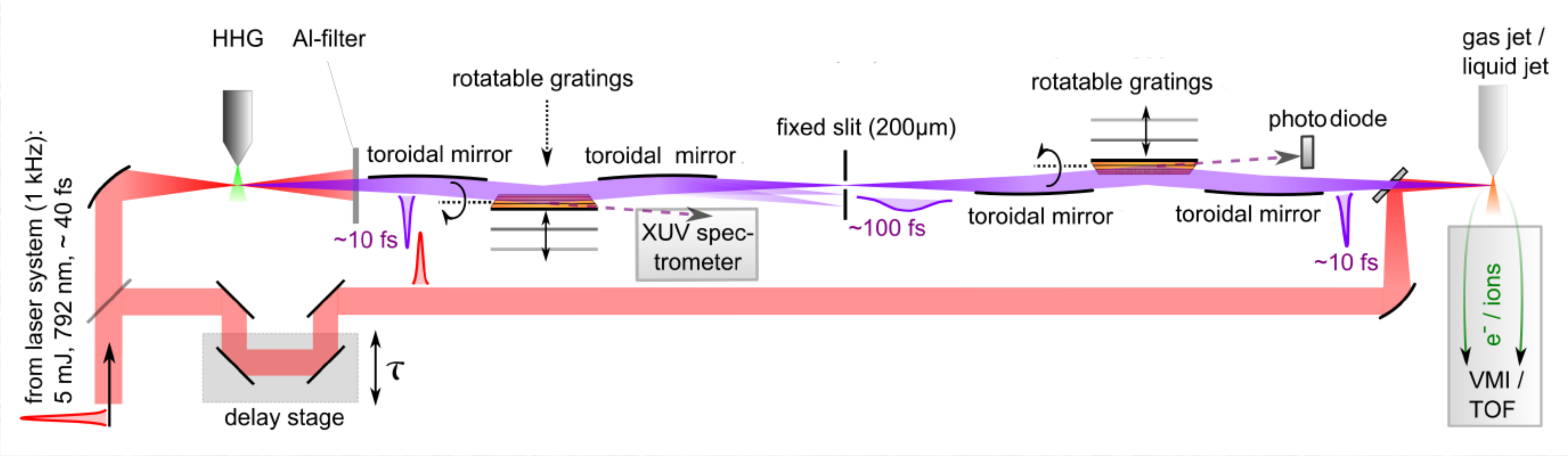


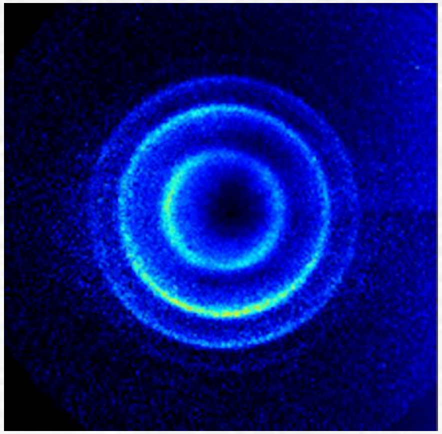


Grating 1, 600 lines/mm	Stretching at intermediate focus	Stretching at apparatus focus
H29, 45 eV	150 fs	4 fs
H25, 38.8 eV	180 fs	4 fs
H21	220 fs	5 fs

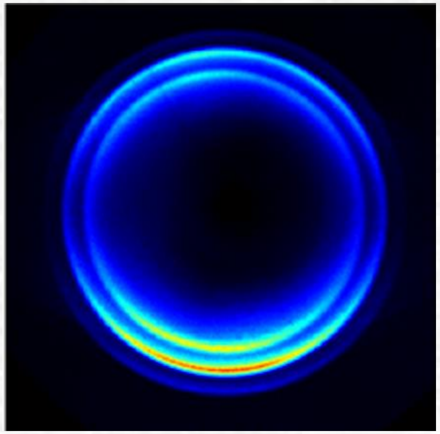
Collaboration with Luca Poletto (Padua)

Time-compensating XUV monochromator



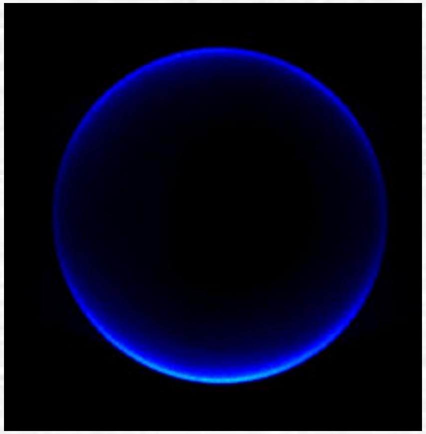


zero-th order



first order

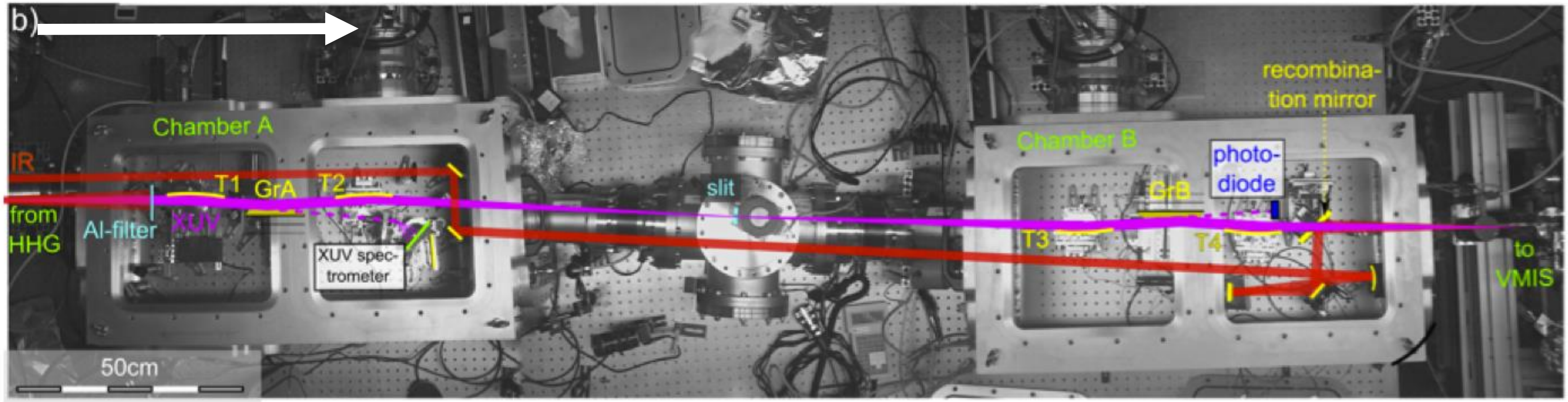
H17



H19

Martin Eckstein and Oleg Kornilov

Beamline design and parameters

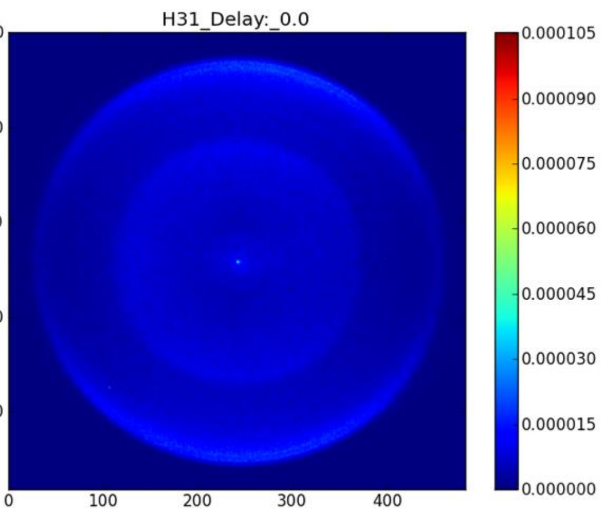


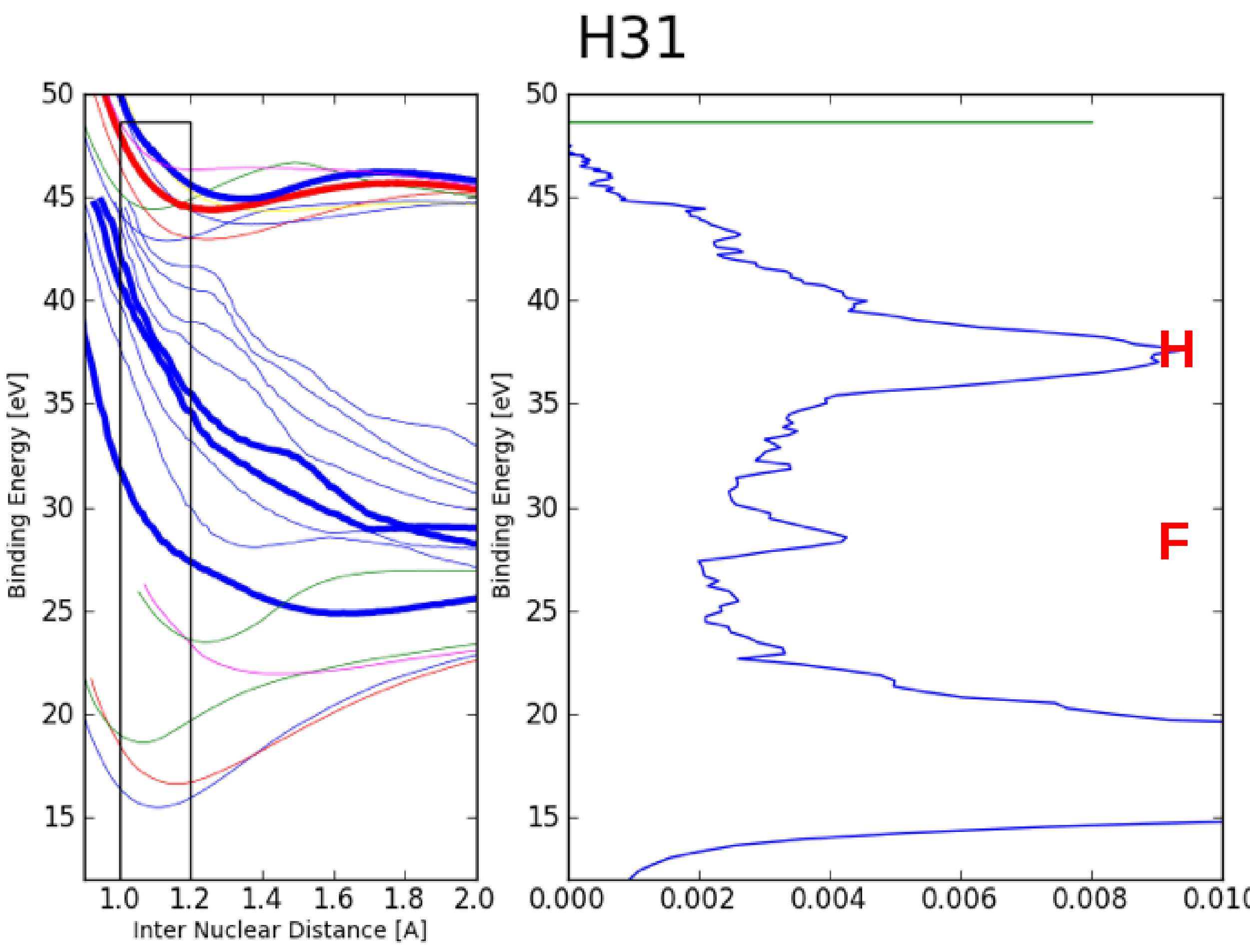
- Time resolution: <25 fs (cross-correlation between XUV and IR)
- Spectral resolution: <500meV (deduced from Ar photoelectron spectra)
- Transmission: 3%-16% for 3 eV - 50 eV (calibrated XUV photodiode)
- Continuous automated scans in the complete spectral range
- Long term stability: scan durations >36 hrs
- Up to 10^7 photons per pulse at 1kHz repetition rate

Martin Eckstein and Oleg Kornilov

Proof of concept: Photochemistry of N_2

VMI Image





XUV-only photoelectron images revealing the participation of successive ionic states

Extracting dissociation thresholds for the F- and H-state

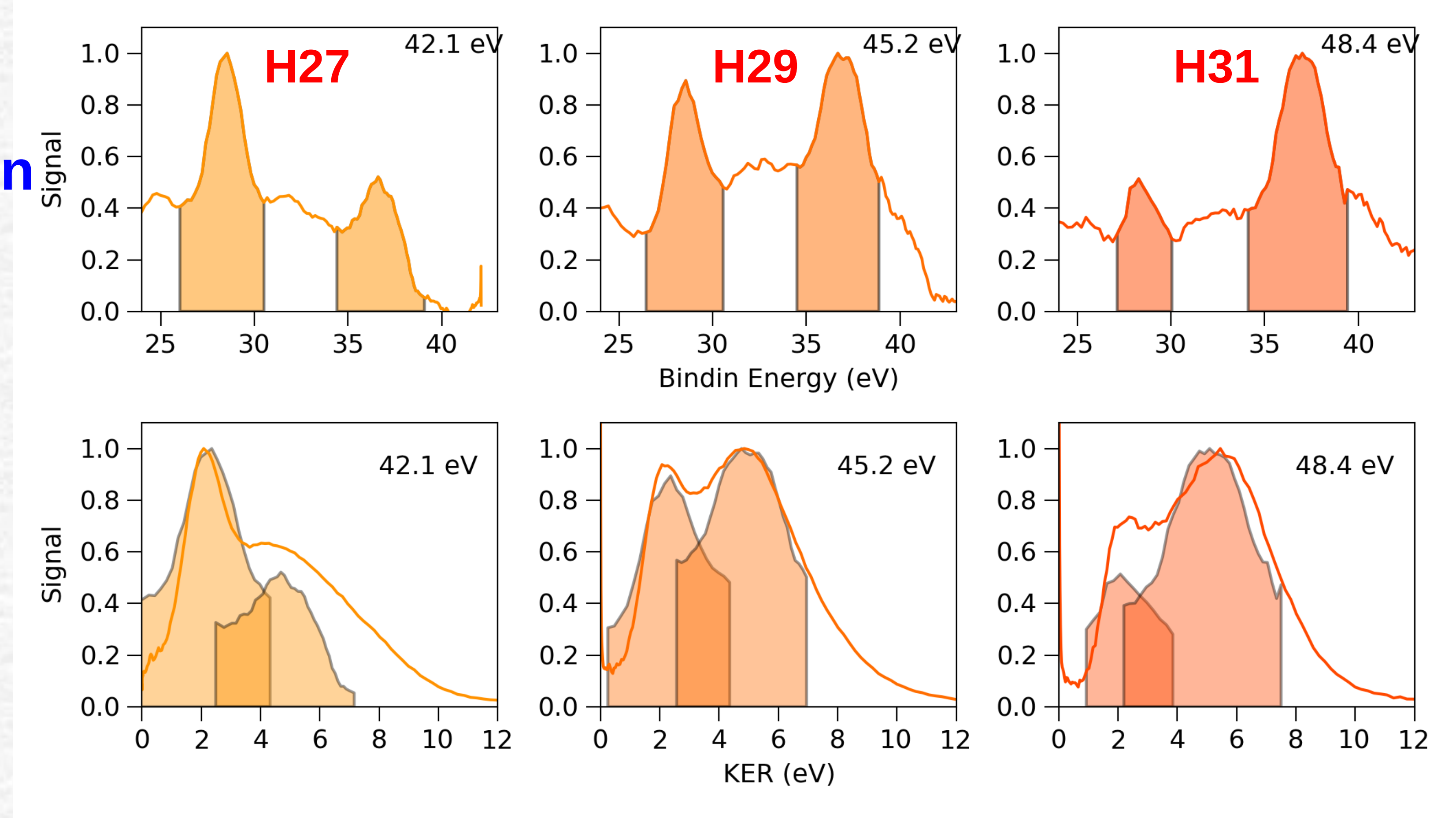
F → N^+ (^3P) + N (^2D)
E = 26.676 eV (L3)

H → N^+ (^1S) + N (^2P)
E = 31.921 eV (L9)

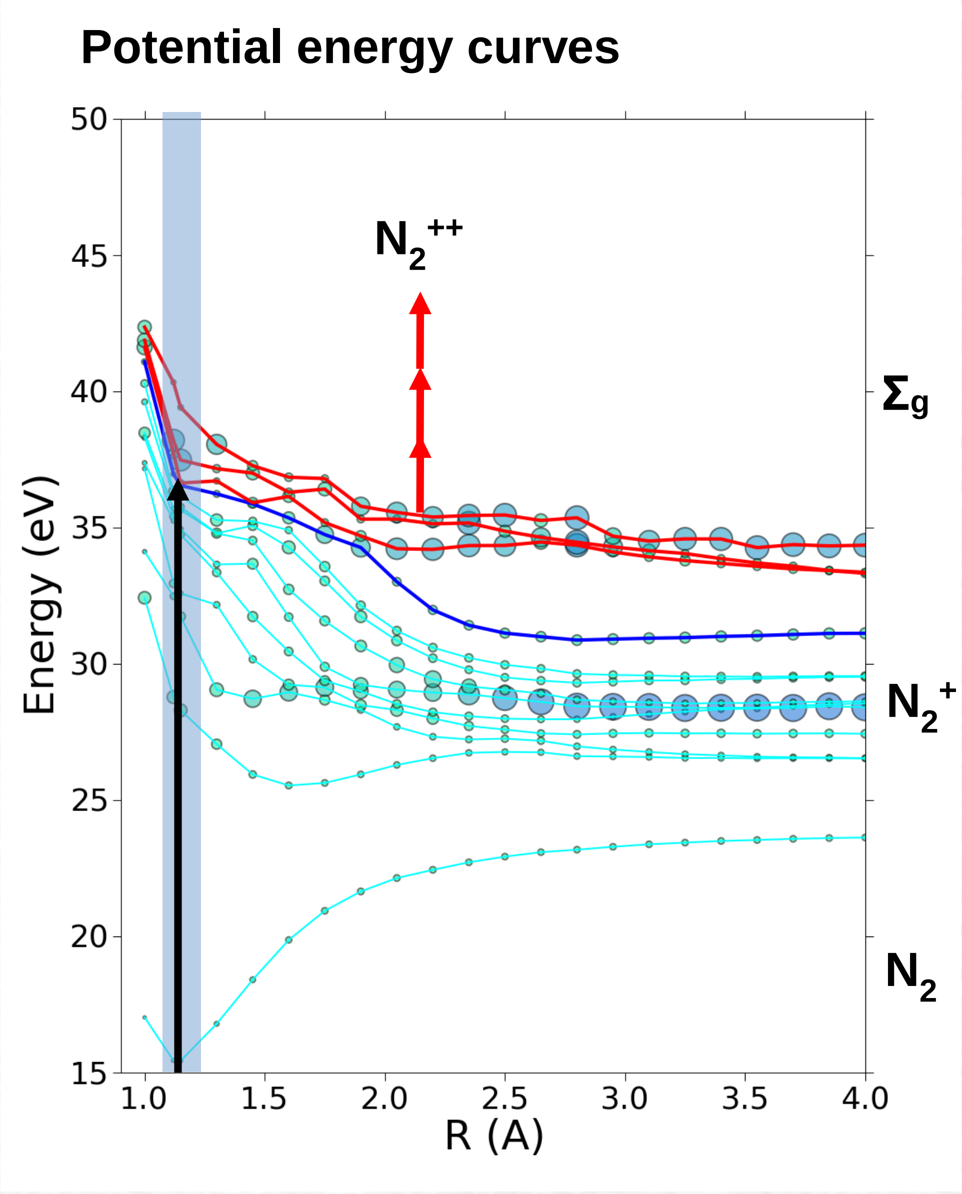
X conventional wisdom!

Electron

Ion



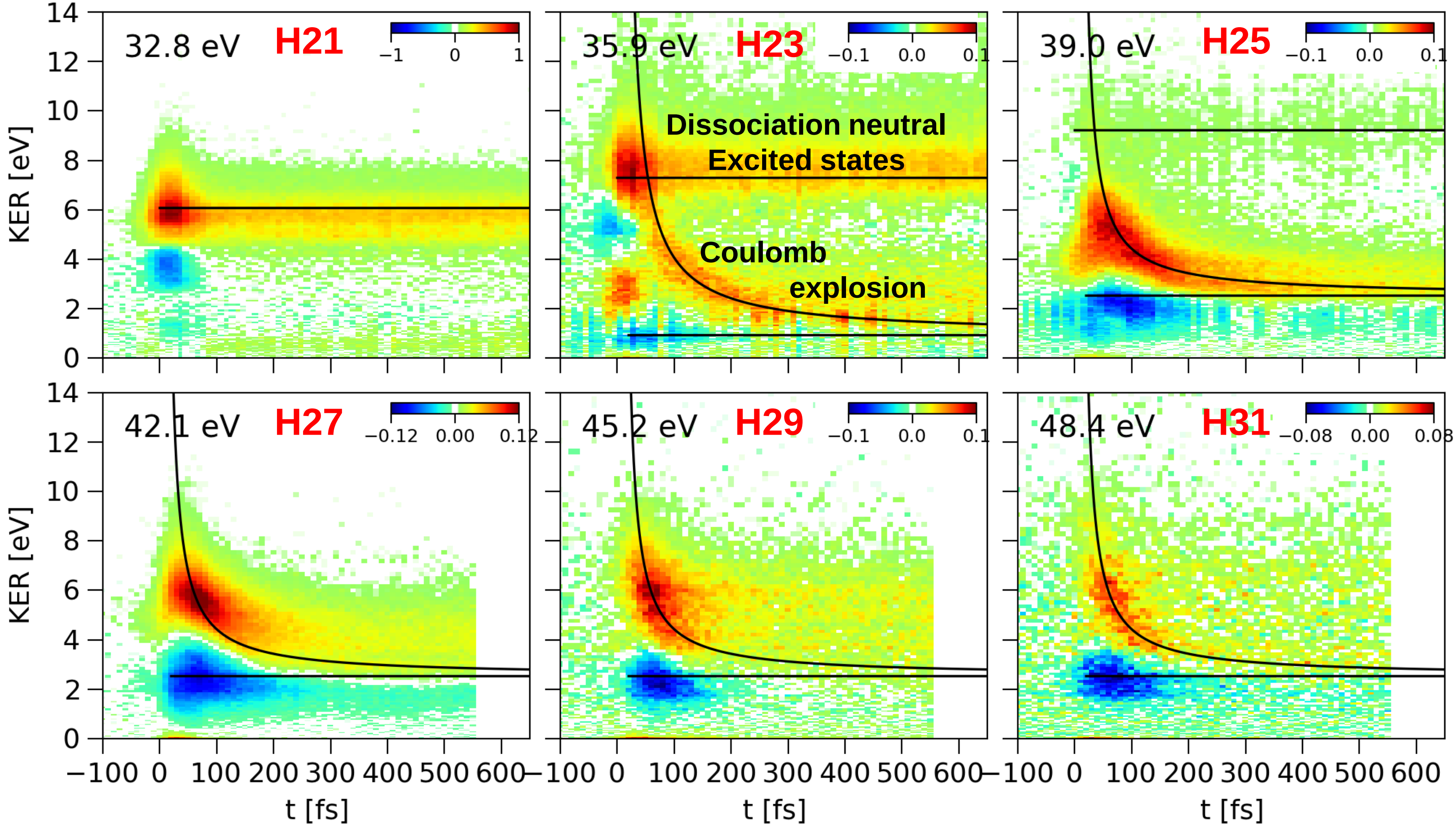
Time-dependent dynamics: XUV + IR



Removal of an electron from the $2\sigma_g$ orbital excites the N_2^+ ion in the H-band and leads to dissociation, dominantly to the L9 dissociation threshold, and with smaller contributions from higher-lying states.

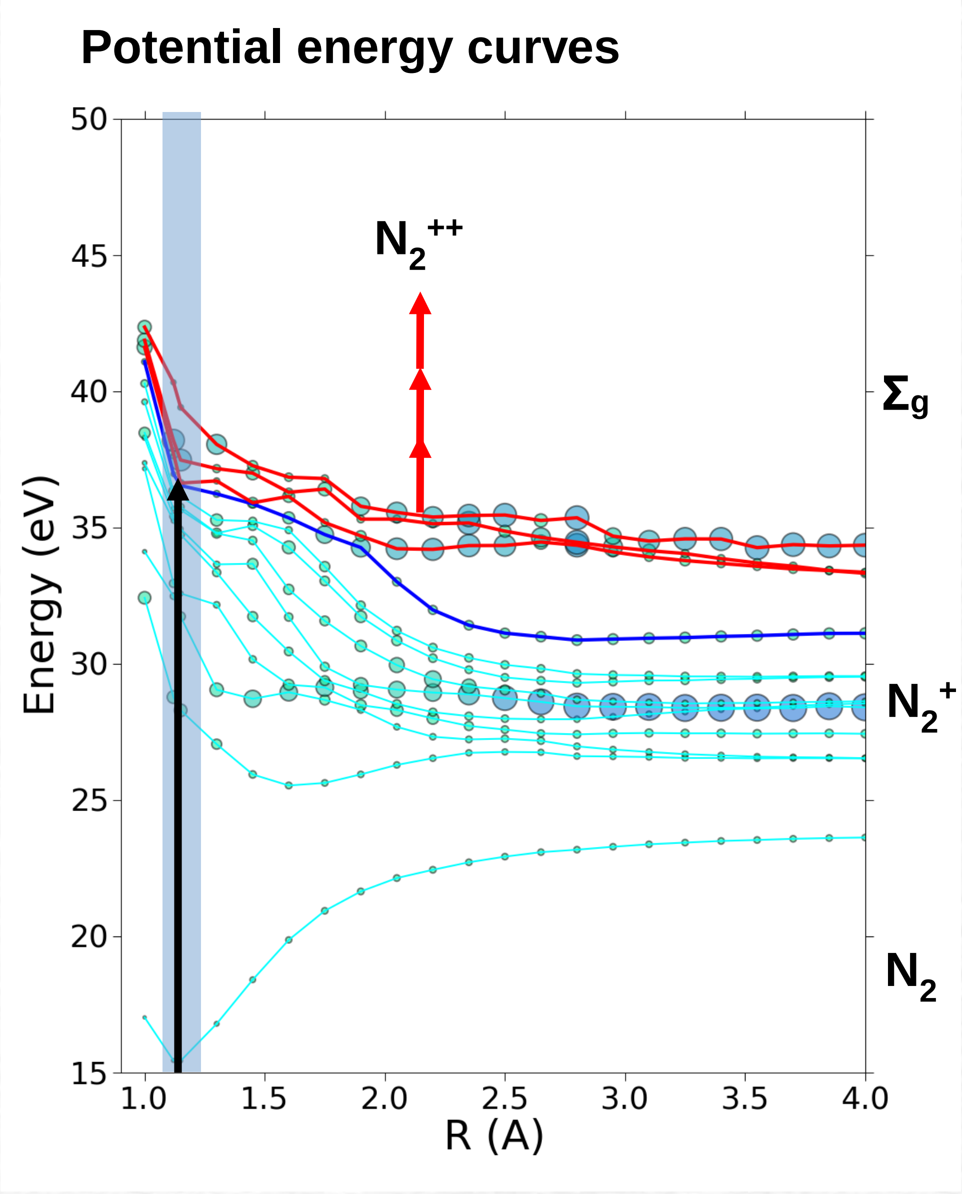
The contributions from higher-lying states can be further ionized using 3-photon excitation by the IR laser, leading to Coulomb explosion of the molecule

Time-dependent dynamics:
XUV + IR Differential Ion KER distributions



Enhancement **Depletion**

Time-dependent dynamics: XUV + IR



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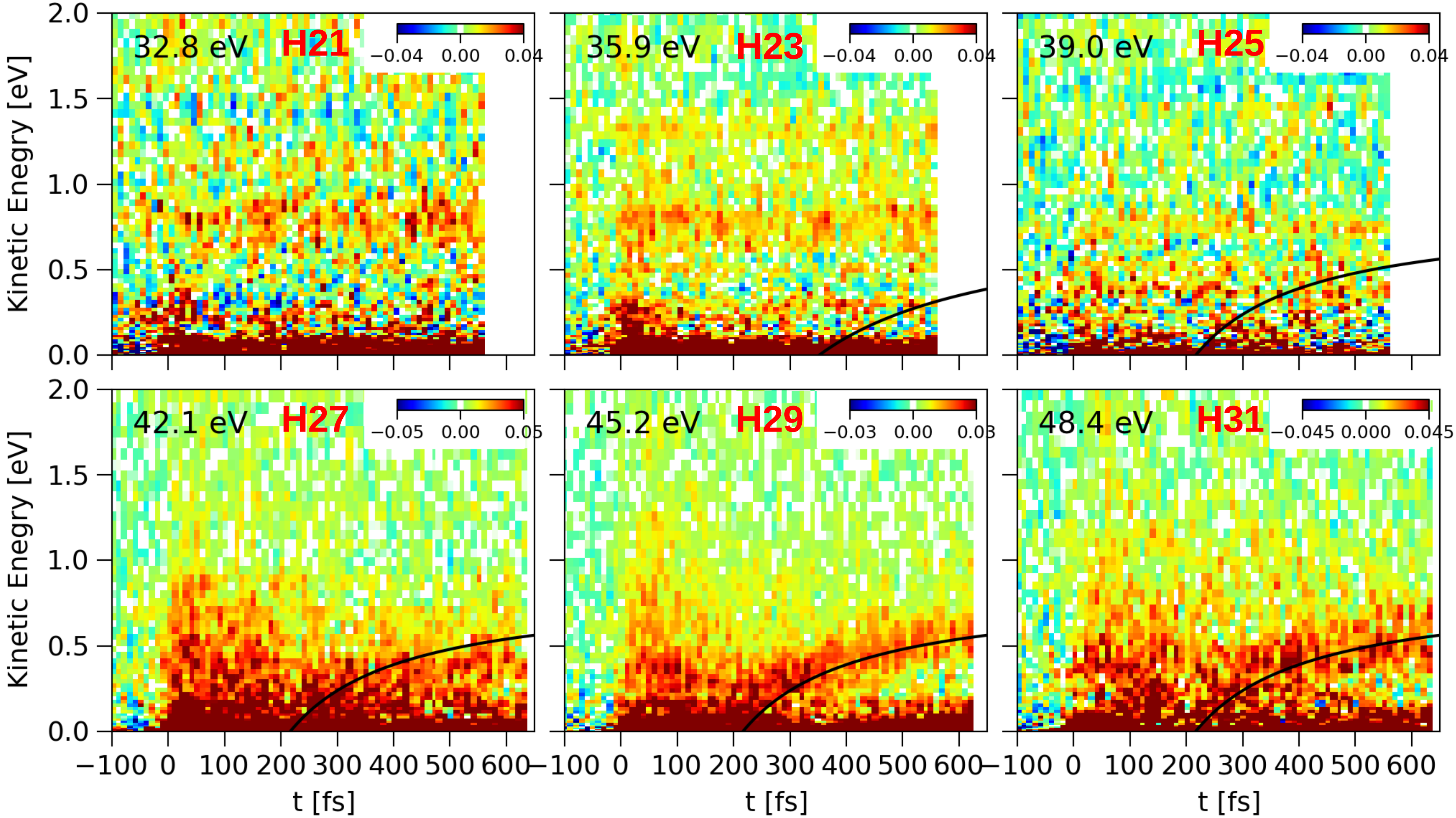
The Coulomb explosion gives a characteristic contribution in the photoelectron spectrum

Time-dependent dynamics: XUV + IR Differential Electron KE distributions



Enhancement Depletion

Conclusions and Implications

- Commissioning XUV time-compensating monochromator beamline
- Major H-band dissociation channel → L9: $N^+ (^1S) + N (^2P)$
- Main pump-probe response comes from secondary channel → L11
- Strong impact on atmospheric models: ^2P atoms are 2 orders of magnitude less reactive than ^2D atoms!!!

2.3.1. Photodissociation

The complex chemistry of Titan's atmosphere is initiated with the photodissociation/photoionization of molecular nitrogen and methane (in the current calculations the contributions of energetic particles are not included and ionization is considered only for N_2). Photons in the EUV region of the solar radiation spectrum, depending on their energy, are able to ionize and dissociate N_2 according to the following pathways (Banks and Kockarts, 1973; Nicolas et al., 2003):

$$N_2 + h\nu \rightarrow N(^4S) + N^+ + e^- (10\%) \lambda < 510\ \text{Å}$$
$$\rightarrow N(^2D) + N^+ + e^- (90\%) \lambda < 510\ \text{Å}$$
$$\rightarrow N_2^+ + e^- \quad 510\ \text{Å} < \lambda < 796\ \text{Å}$$
$$\rightarrow N(^2D) + N(^4S) 796\ \text{Å} < \lambda < 1000\ \text{Å}.$$

(25 eV)

Huygens-Cassini mission to Saturn

Lavvas et al, Planetary and Space Science 56,27 (2008)

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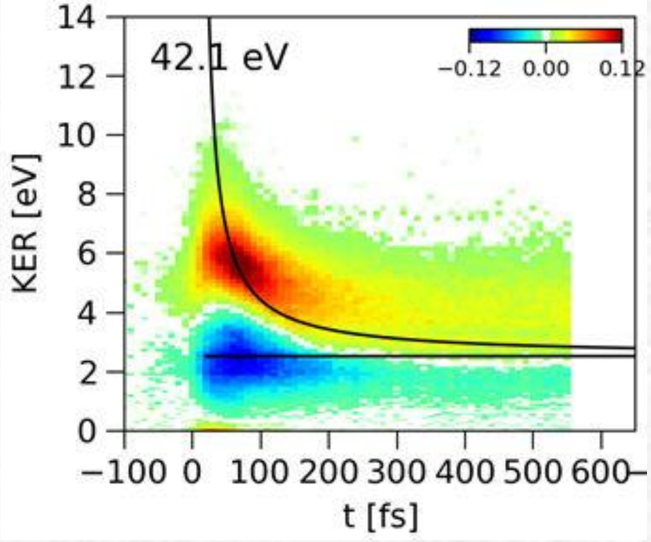
  - XUV-induced molecular dynamics



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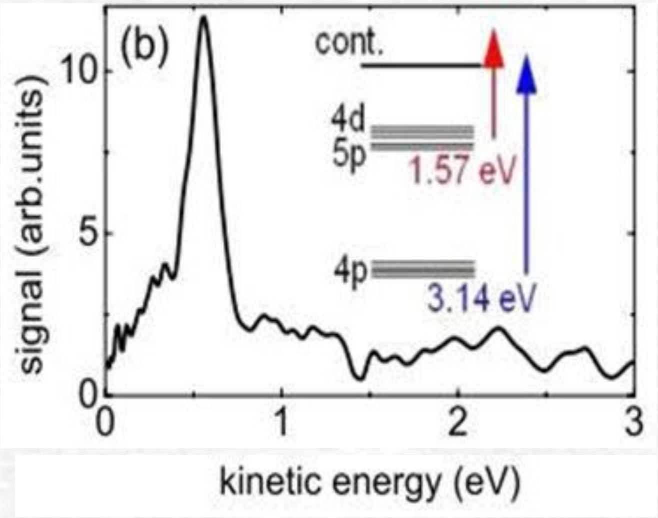
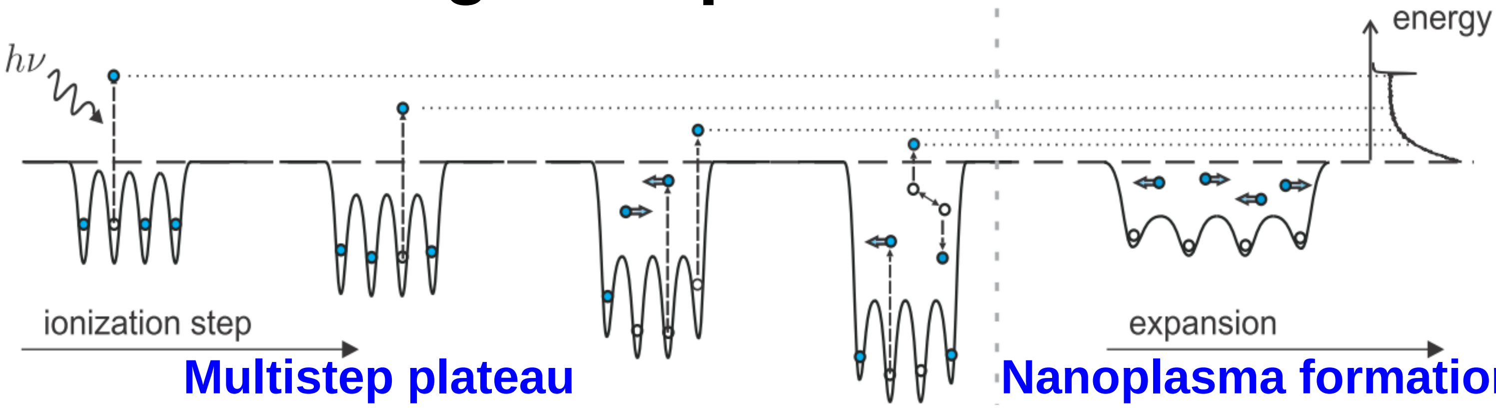
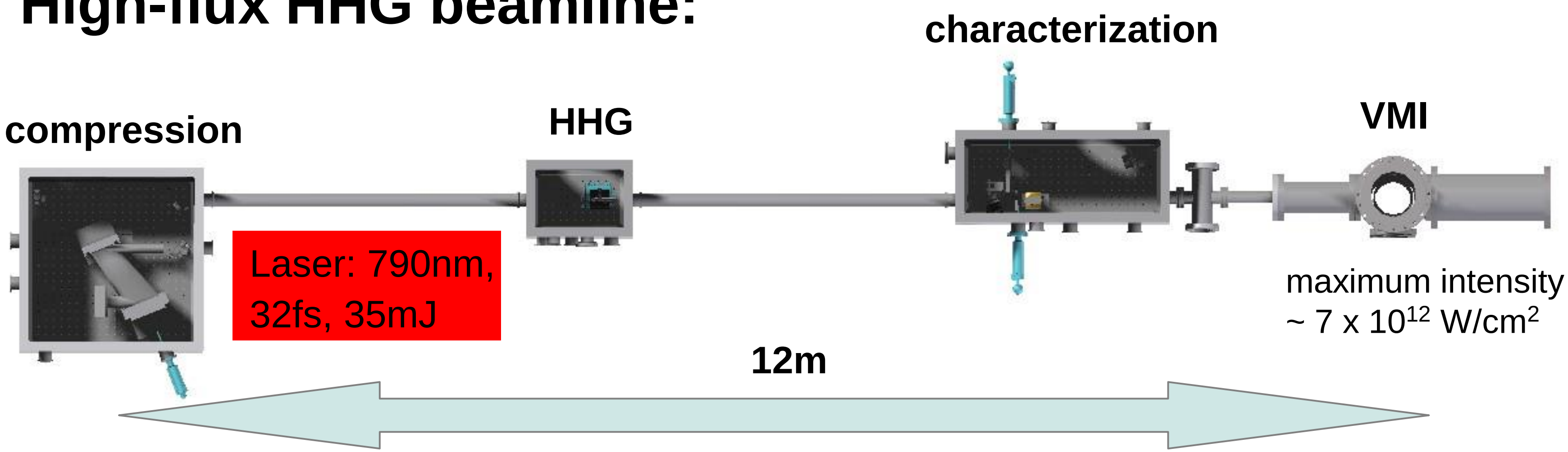
  - Probing charging and dissociation of highly-excited rare gas clusters



Understanding from previous FEL research:



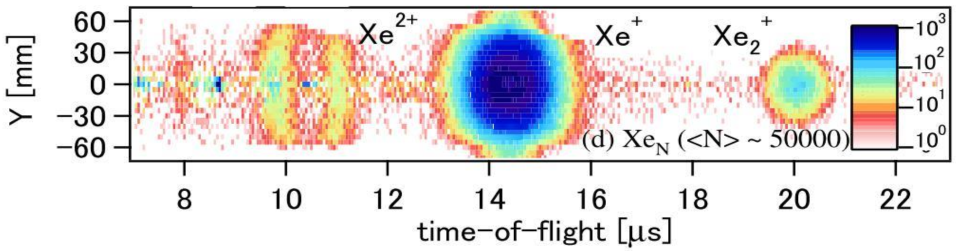
High-flux HHG beamline:



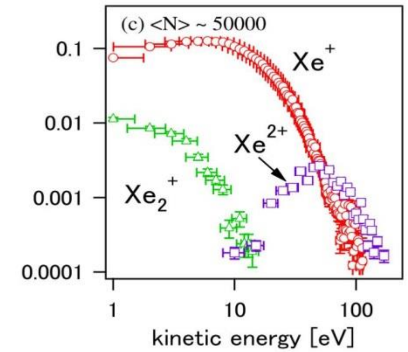
HHG very promising for improved understanding of cluster dynamics

Clusters in HHG / FEL pulses

FEL



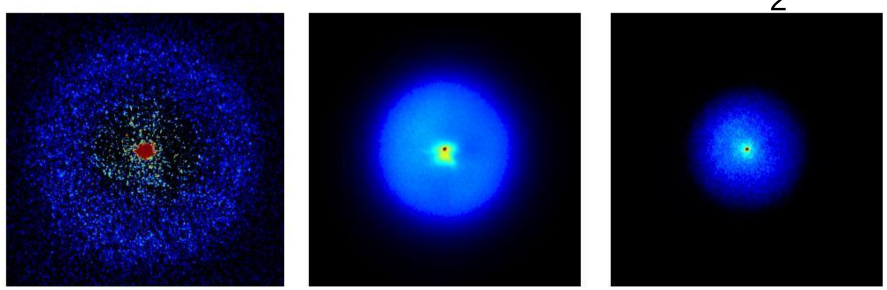
H. Iwayama *et al.*, J. Phys. B **43**, 161001 (2010).

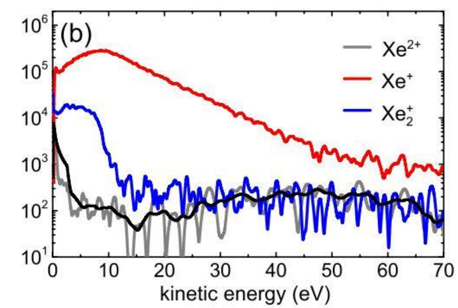


<N>=36000

HHG

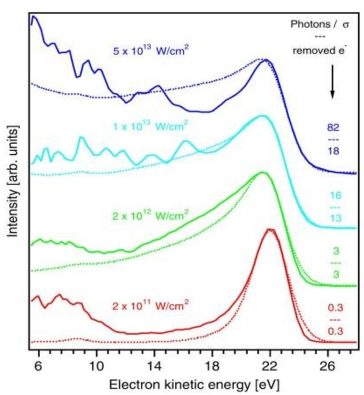
Xe^{2+} Xe^{+} Xe_2^{+}





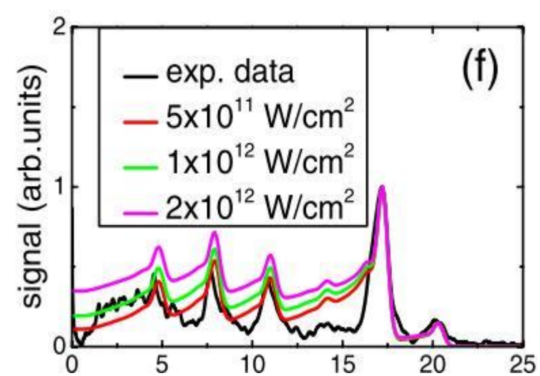
B. Schütte *et al.*, PRL 112, 073003 (2014).

FEL



C. Bostedt *et al.*, PRL **100**, 133401 (2008).

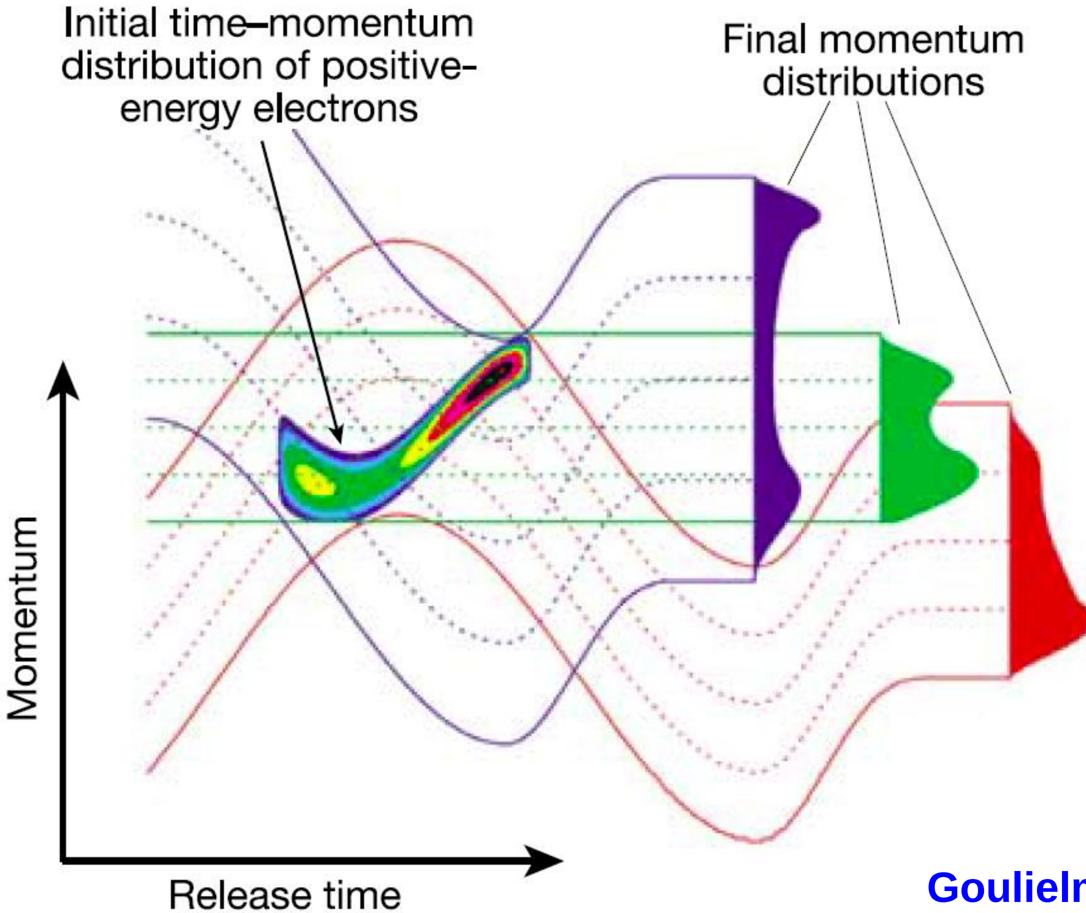
HHG



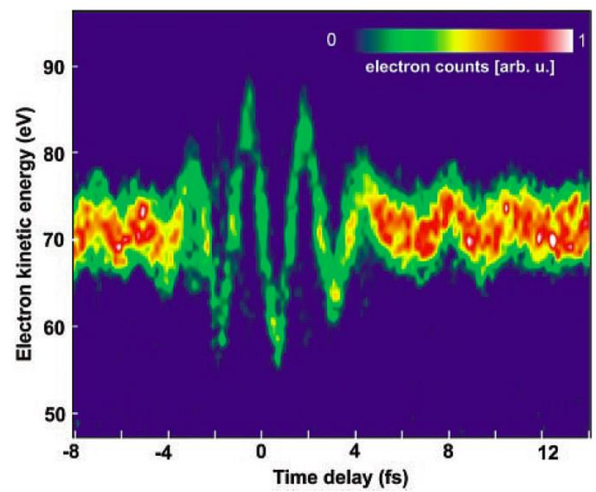
B. Schütte *et al.*, PRL 112, 073003 (2014).

FEL results can be qualitatively reproduced!

Look at charging dynamics using THz streaking



Blue and red:
Lines of constant canonical momentum p=k(t)+qA(t) give different tomographic projections



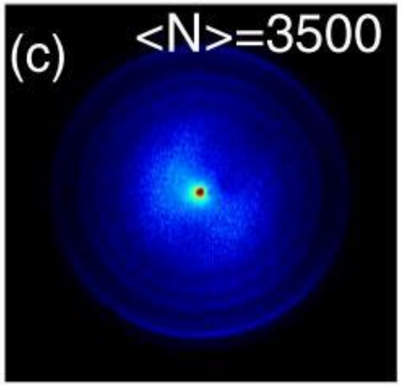
Goulielmakis et al., Science 305, 1267 (2004)

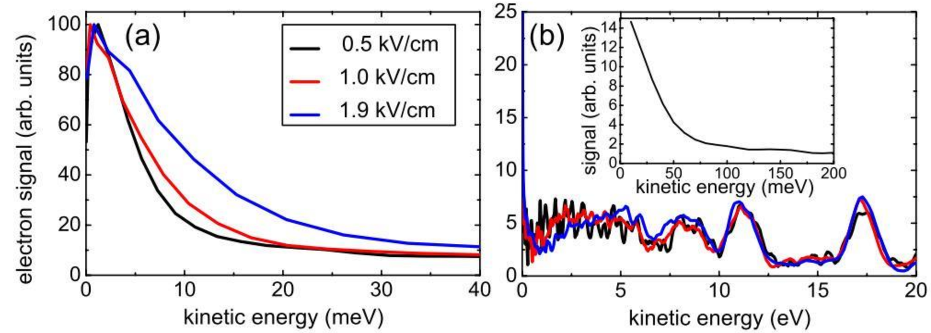
Lesson from attosecond science: time-dependent photoionization can be revealed by a streaking measurement

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

Fragmentation and Recombination dynamics

Frustrated Recombination:

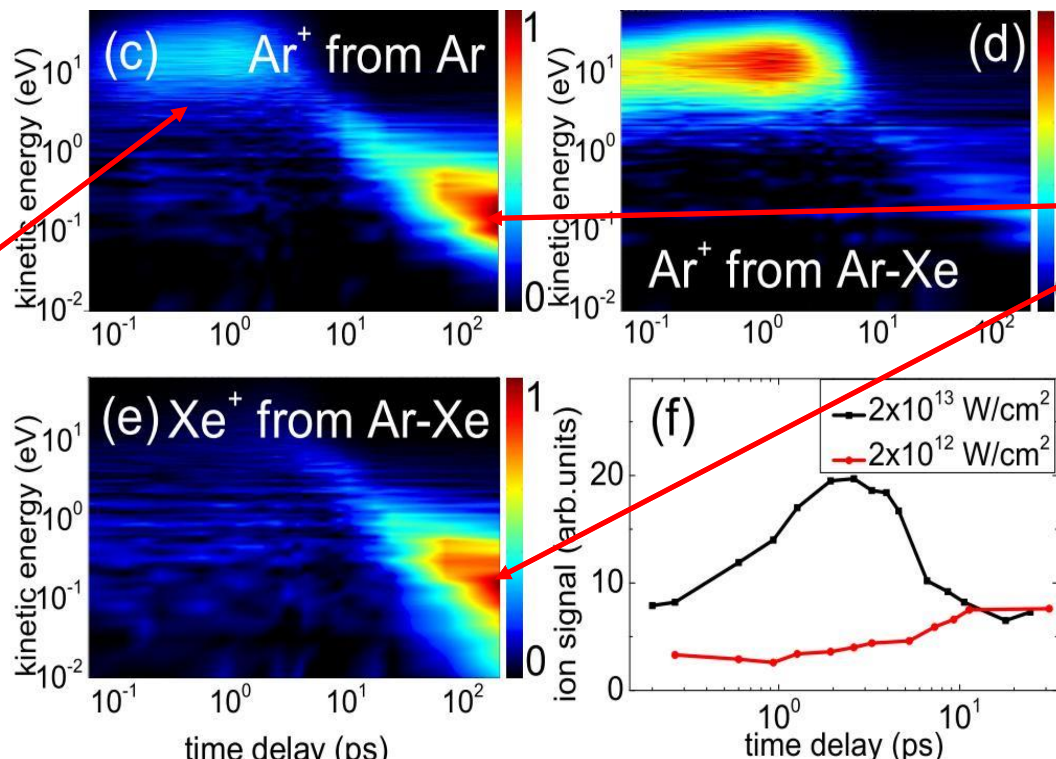




B. Schütte *et al.*, PRL 112, 073003 (2014)

Reionization of Excited Atoms from Recombination (REAR)

Short time delays: Resonance effects



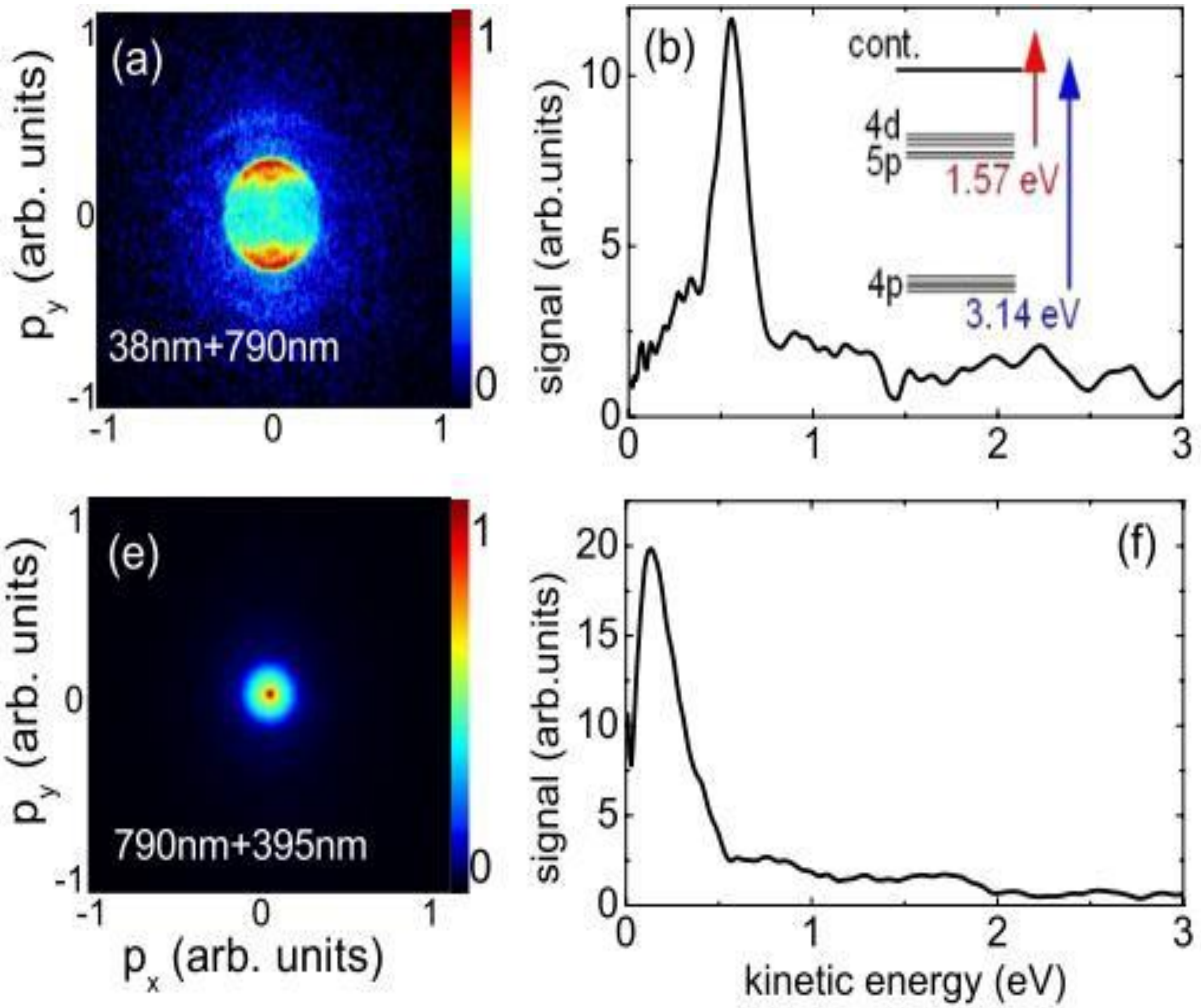
Long time delays: Recombination

NIR induced signals

B. Schütte *et al.*, PRL 112, 253401 (2014)

Fragmentation and Recombination dynamics

Reionization of Excited Atoms from Recombination (REAR)



B. Schütte *et al.*, PRL 112, 253401 (2014)

Summary and outlook

Free electron lasers offer unparalleled performance when very high photon energies (> 1 keV) or very high XUV/X-ray non-linearities are required

High harmonic generation (HHG) can serve as a complementary source under conditions where the required photon energies are somewhat lower (< 1 keV) or under conditions where the dependence on the XUV/X-ray intensity is linear or of modest order
The easy (full-time) access and the ready availability of synchronized secondary sources (IR, UV, THz) is a major advantage

Anticipated technical improvements:

- Broadband continua up to and possibly beyond the O K-edge
- Monochromatized XUV-IR/UV pump-probe spectroscopy with ≤ 10 fs resolution
- Focusibility of single harmonics to ≥5x10^{13} W/cm^{2}
- XUV pump-XUV probe spectroscopy with attosecond time resolution

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+ many others….