Connecting Lab-Based Attosecond Science with FEL research

Marc Vrakking

Max-Born-Institut (MBI), Berlin, DE

FOM Institute for Atomic and Molecular Science (AMOLF), Amsterdam, NL

CERN Colloquium 27-1-2011

Collaborators

Freek Kelkensberg
Wing Kiu Siu
Georg Gademann
Ymkje Huismans
Aneta Smolkowska
Arnaud Rouzee
Arjan Gijsbertsen

Max-Born-Institut

Olga Smirnova
Alex Harvey

Misha Ivanov

Dieter Bauer

Giuseppe Sansone
Federico Ferrari
Enrico Benedetti
Mauro Nisoli

Ken Schafer

Robert Lucchese

Franck Lépine
Chrisian Bordas

Sergei Prophuzhenko

Stefan Düsterer
Nikola Stojanovic
Franz Tavella
Armin Azima
Michael Gensch
Rolf Treusch
+ the machine group

FOM Institute
for
Plasma Physics
Rijnhuizen

Joost Bakker
Britta Redlich
Lex van der Meer
Giel Berden

Tatiana Marchenko

Kiyoshi Ueda
Hironobu Fukozawa
Ayako Yamada

Irina Znakovskaya
Sergey Zherebtsov
Matthias Kling

Frank Filsinger
Jochen Küpper
+ ASG

Michael Spanner

Sebastian Zamith

Johan Mauritsson
Per Johnsson
Marko Swoboda
Kathrin Kluender
Anne L'Huillier

Thomas Schlatholter
Ronnie Hoekstra
Ina Blank

David Holland

Willem Vermin

Novel XUV/x-ray sources (HHG and FELs) allow to push atomic & molecular science beyond the present state-of-the-art

❖Temporal properties → Electron dynamics

Example:

Attosecond time-resolved pump-probe spectroscopy in H_2

Wavepacket interferometry experiments give access to attosecond electron dynamics

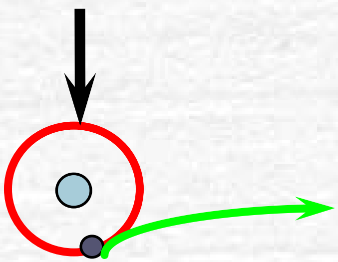
❖Wavelength properties → Nuclear dynamics

Example:

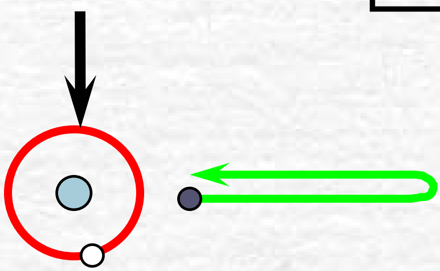
Molecular frame photo-emission gives access to (time-dependent) orbital and structural information – pump-probe spectroscopy at FLASH/LCLS and using an HHG source

Making Attosecond Pulses: High Harmonic Generation

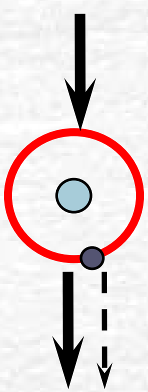
Intense near-infrared femtosecond laser



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

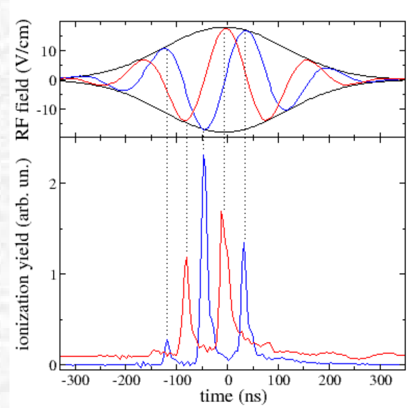


Step 2: acceleration of the electron in the oscillatory laser field

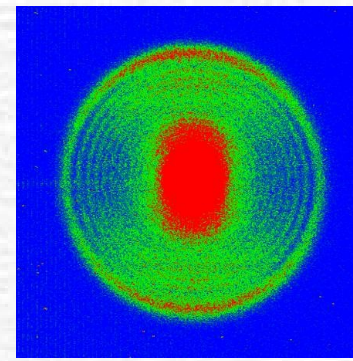


Step 3: recombination, accompanied by the emission of an XUV photon

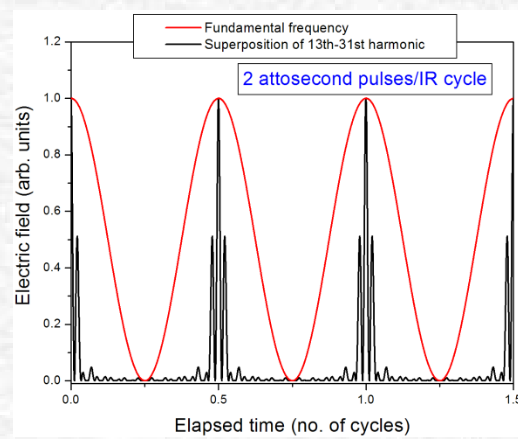
Intense near-infrared femtosecond laser + XUV radiation

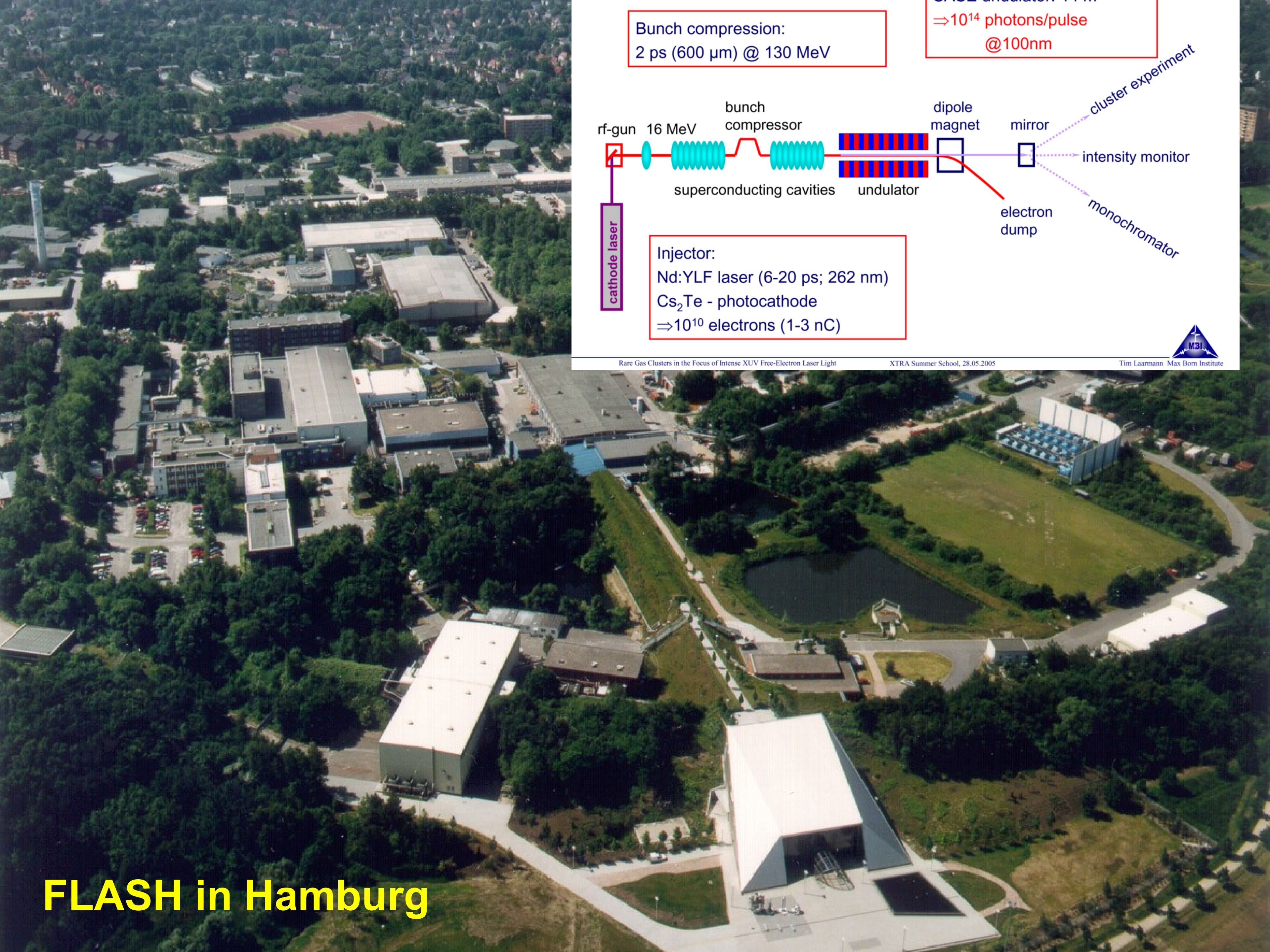


Guertler et.al., Phys. Rev. Lett. 92, 063901 (2004)



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





FLASH in Hamburg

1. Application of Attosecond Pulses to Molecular Science

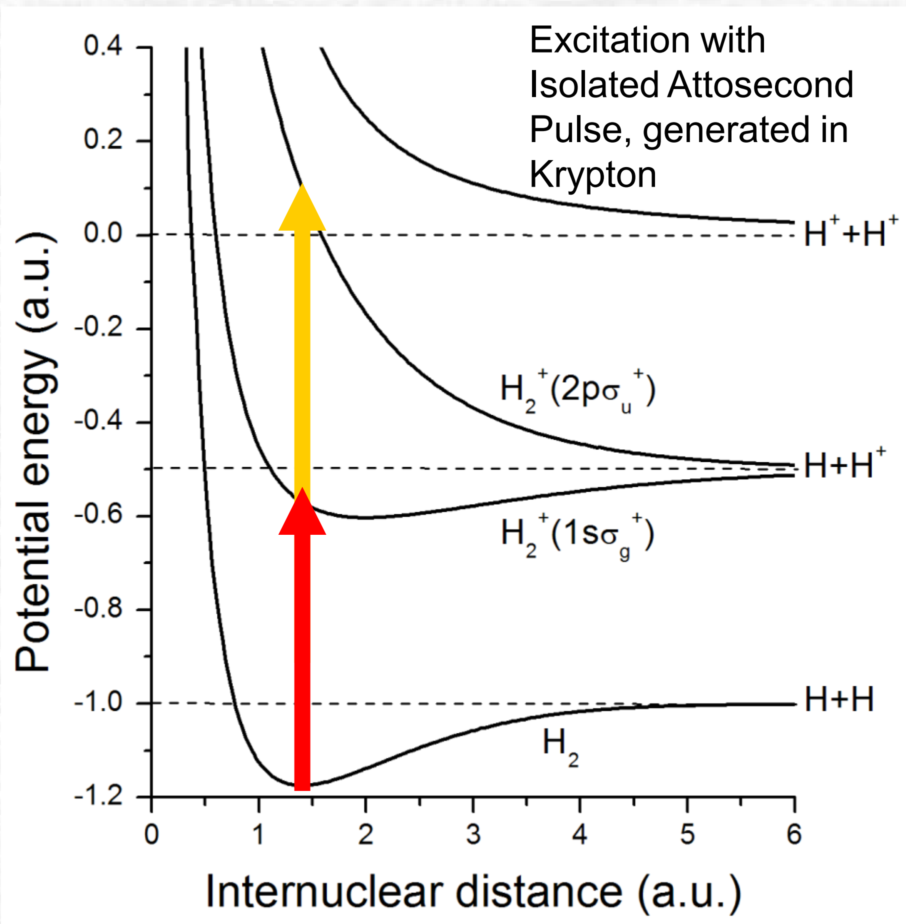
Question: why should we care?

Answer:

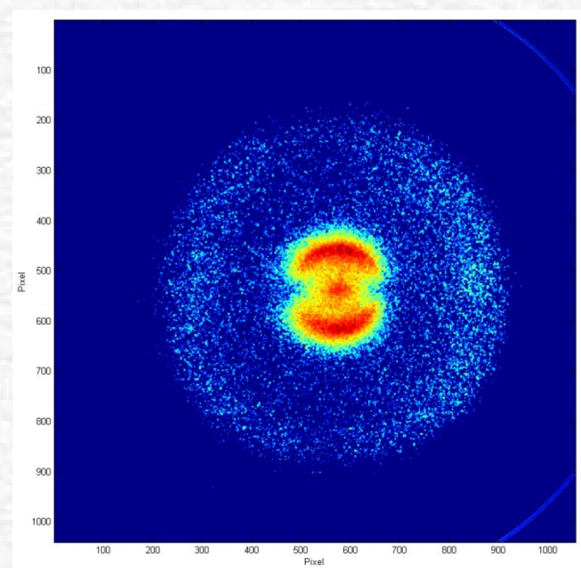
- **Any light-induced photo-chemical process starts with an electronic interaction; atomic motion sets in after coupling of electronic and nuclear degrees of freedom**

- **Many light-induced interactions involve multiple electrons – when is coupling of electronic degrees of freedom important?**

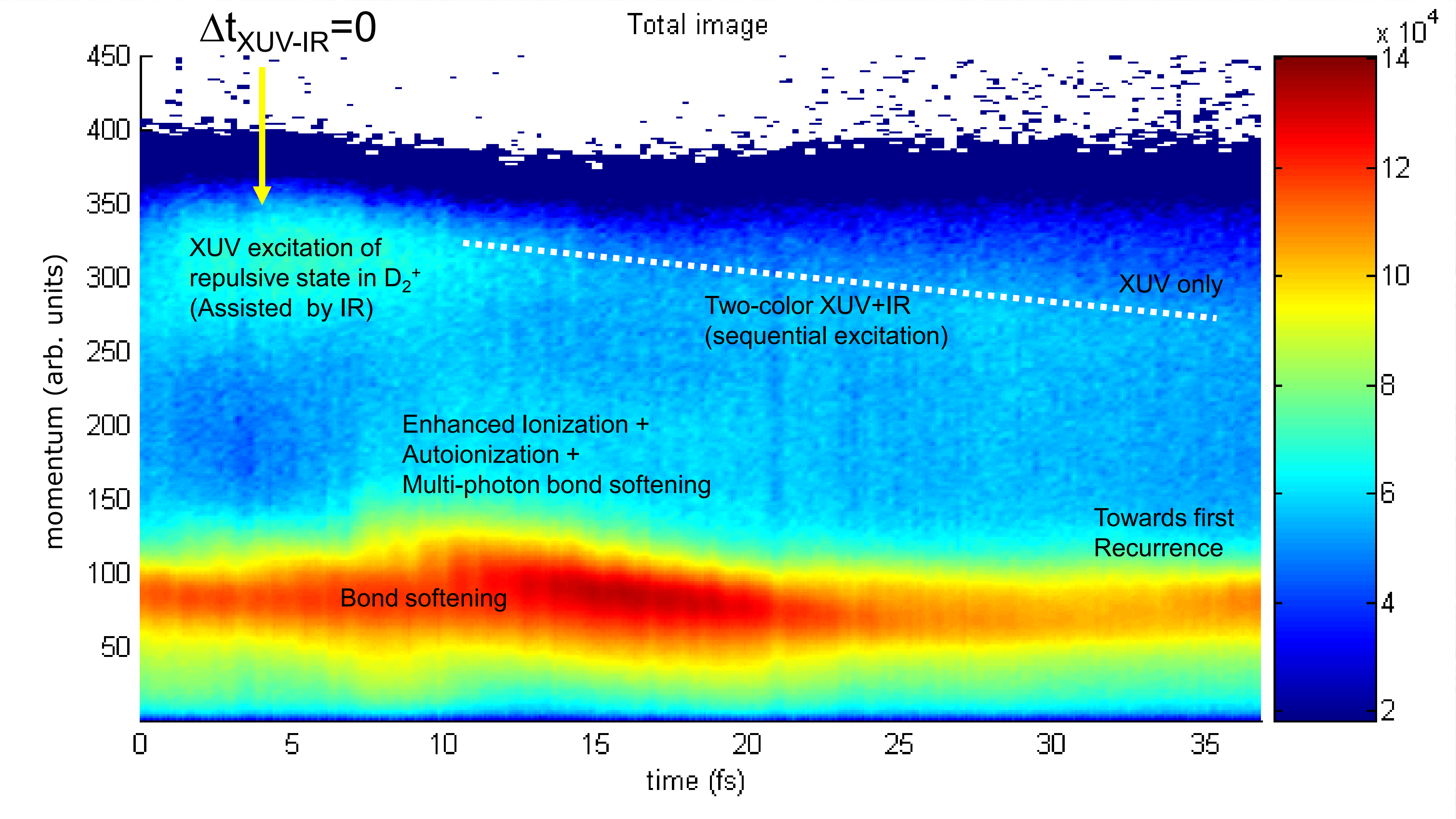
XUV-IR Pump-probe experiments on H_2 and D_2



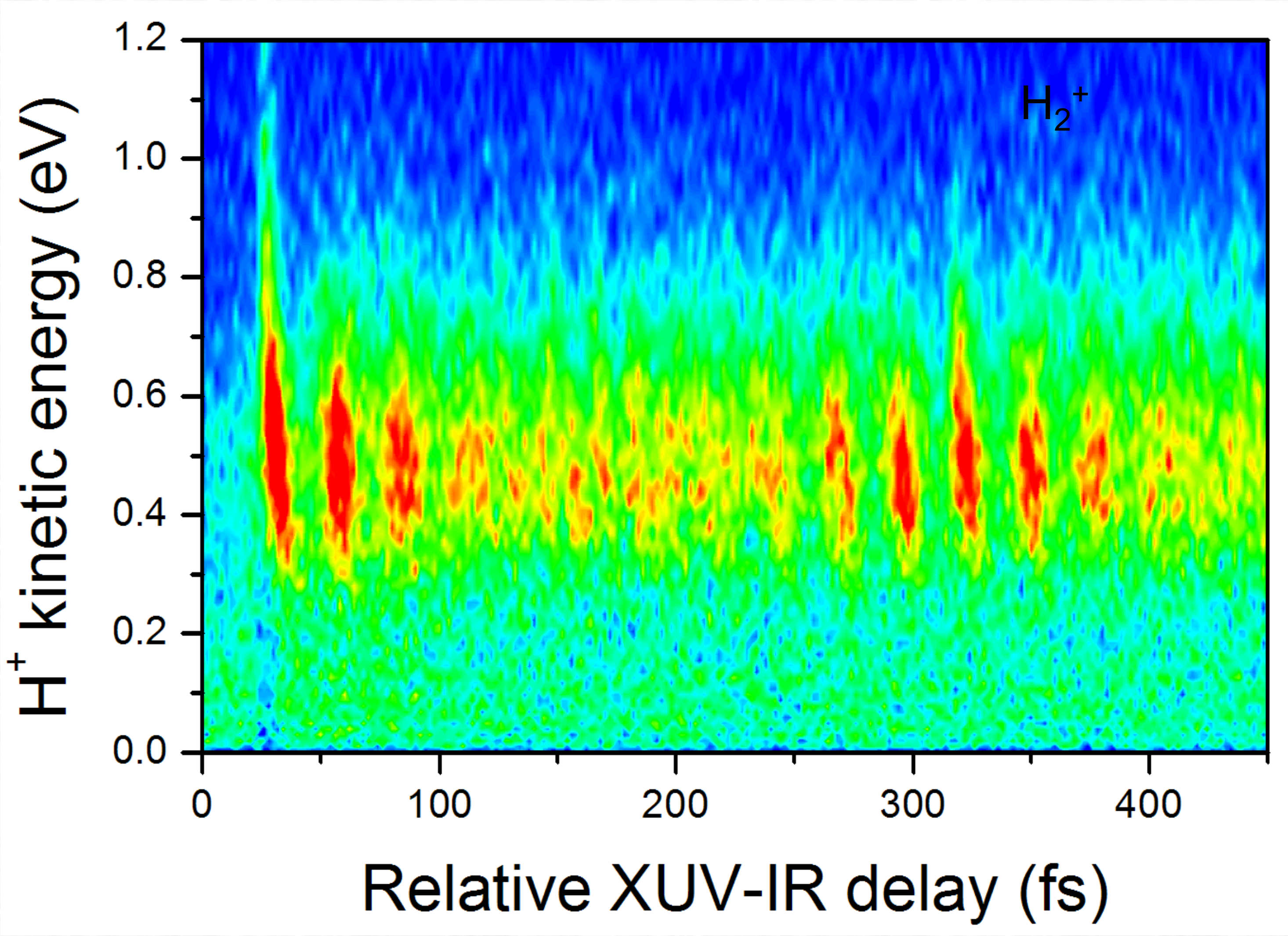
Use isolated attosecond pulse generated in Krypton to launch a wavepacket on the $2p\sigma_u^+$ state or the $1s\sigma_g^+$ state and investigate the subsequent IR interaction



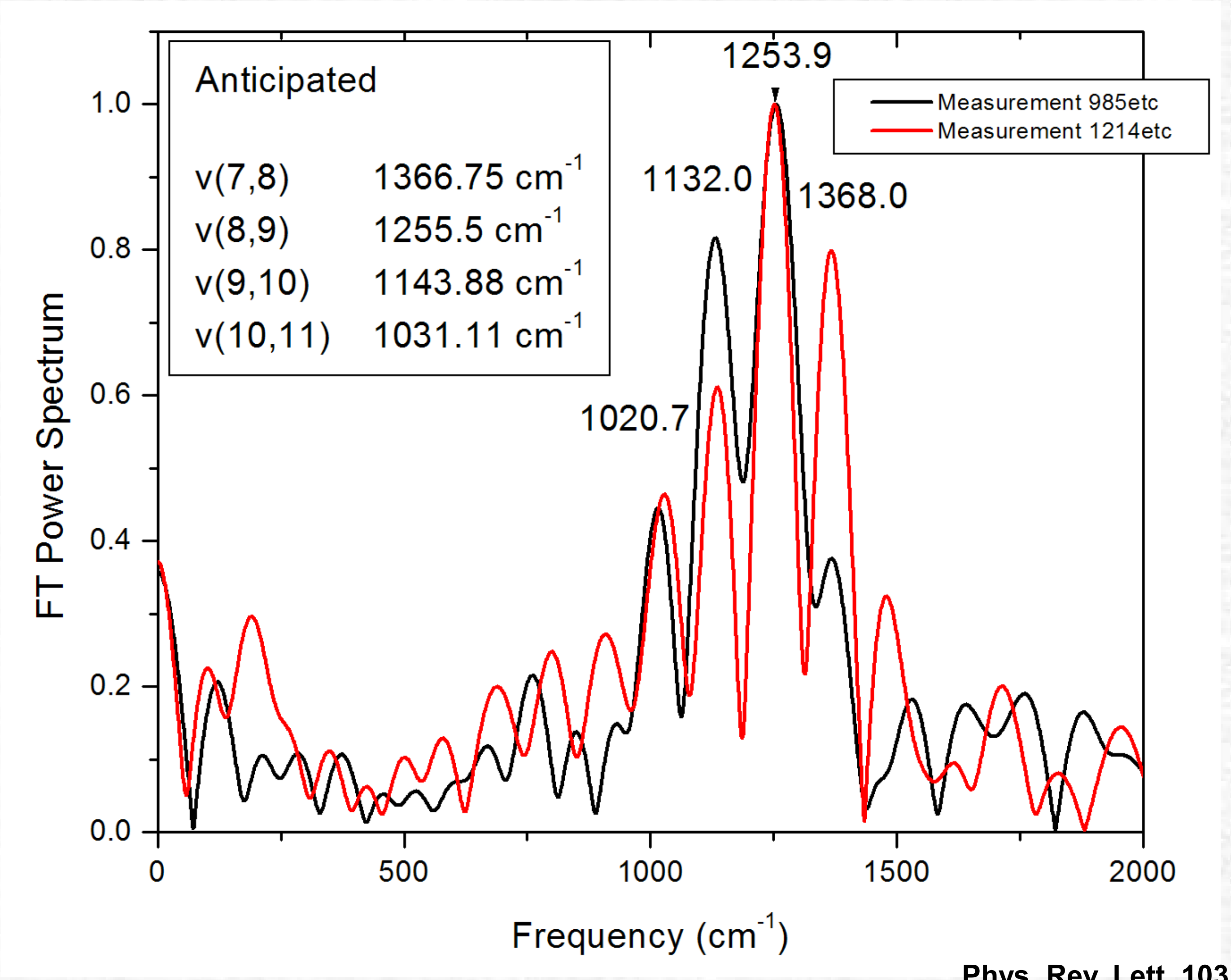
Momentum distribution as a function of XUV-IR time-delay



Vibrational wave packet measured by R-dependence of bond softening

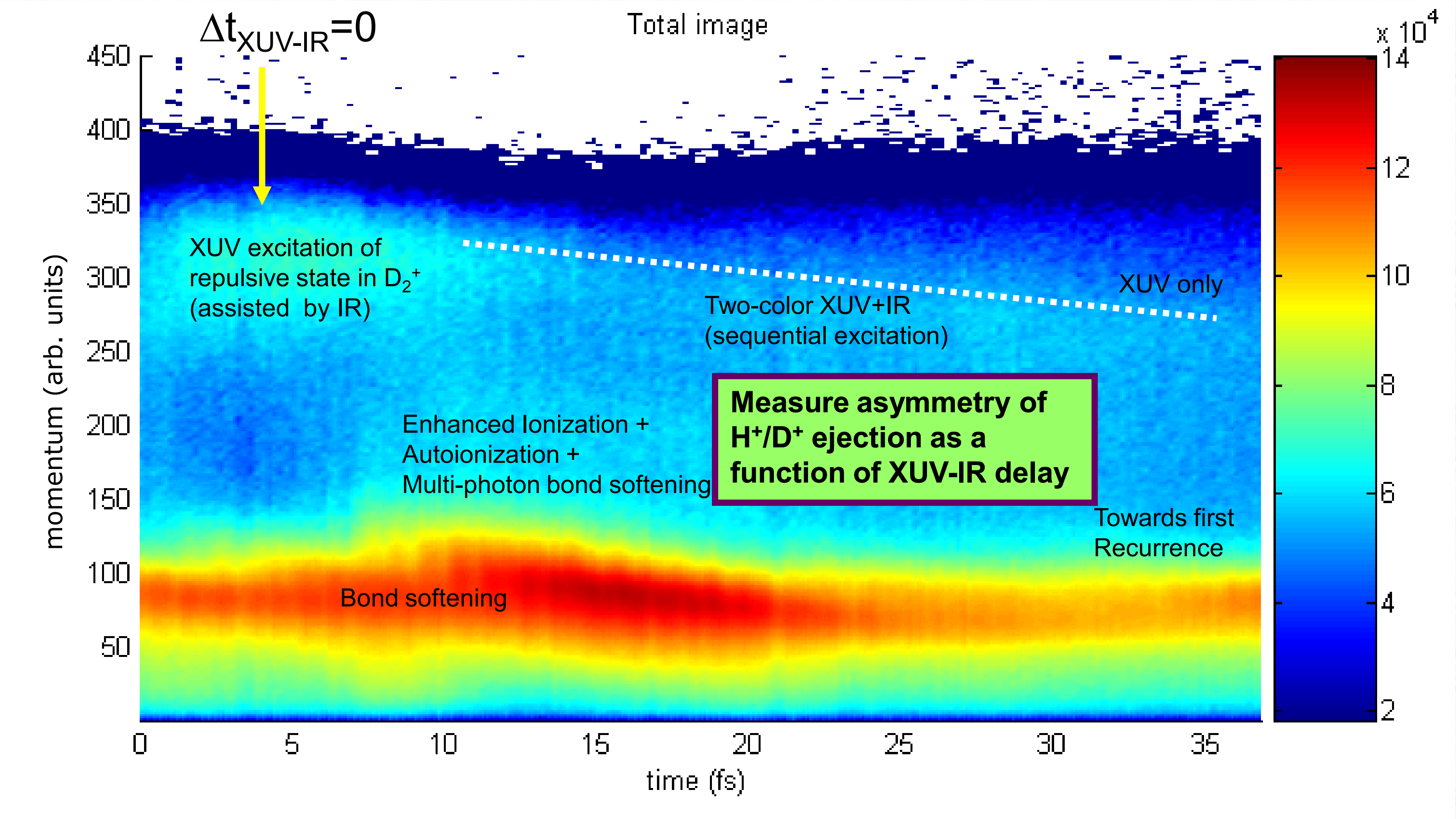


Assignment of observed quantum beats in bond softening signal



Phys. Rev. Lett. 103, 123005 (2009)

Momentum distribution as a function of XUV-IR time-delay



Localized States of H_2^+

The $2p\sigma_u^+$ and $1s\sigma_g^+$ states can be viewed as bonding and anti-bonding combinations of 1s atomic orbitals

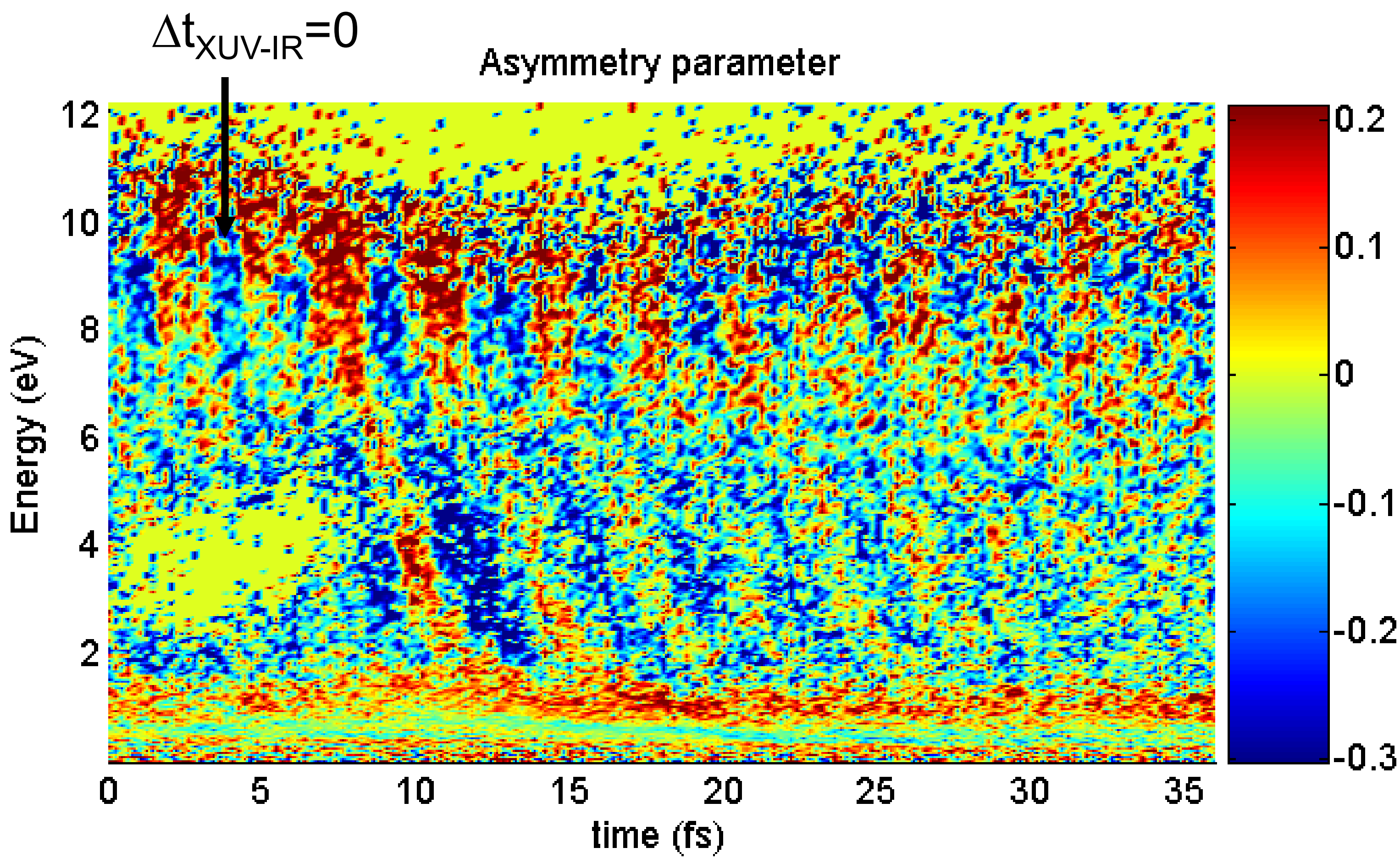
$$|g\rangle = \frac{1}{\sqrt{2}}\left(|1s_{left}\rangle + |1s_{right}\rangle\right)$$
$$|u\rangle = \frac{1}{\sqrt{2}}\left(|1s_{left}\rangle - |1s_{right}\rangle\right)$$

Therefore, the nuclear wave function can also be expressed on a basis of localized states

$$|l\rangle = \frac{1}{\sqrt{2}}\left(|g\rangle + |u\rangle\right)$$
$$|r\rangle = \frac{1}{\sqrt{2}}\left(|g\rangle - |u\rangle\right)$$

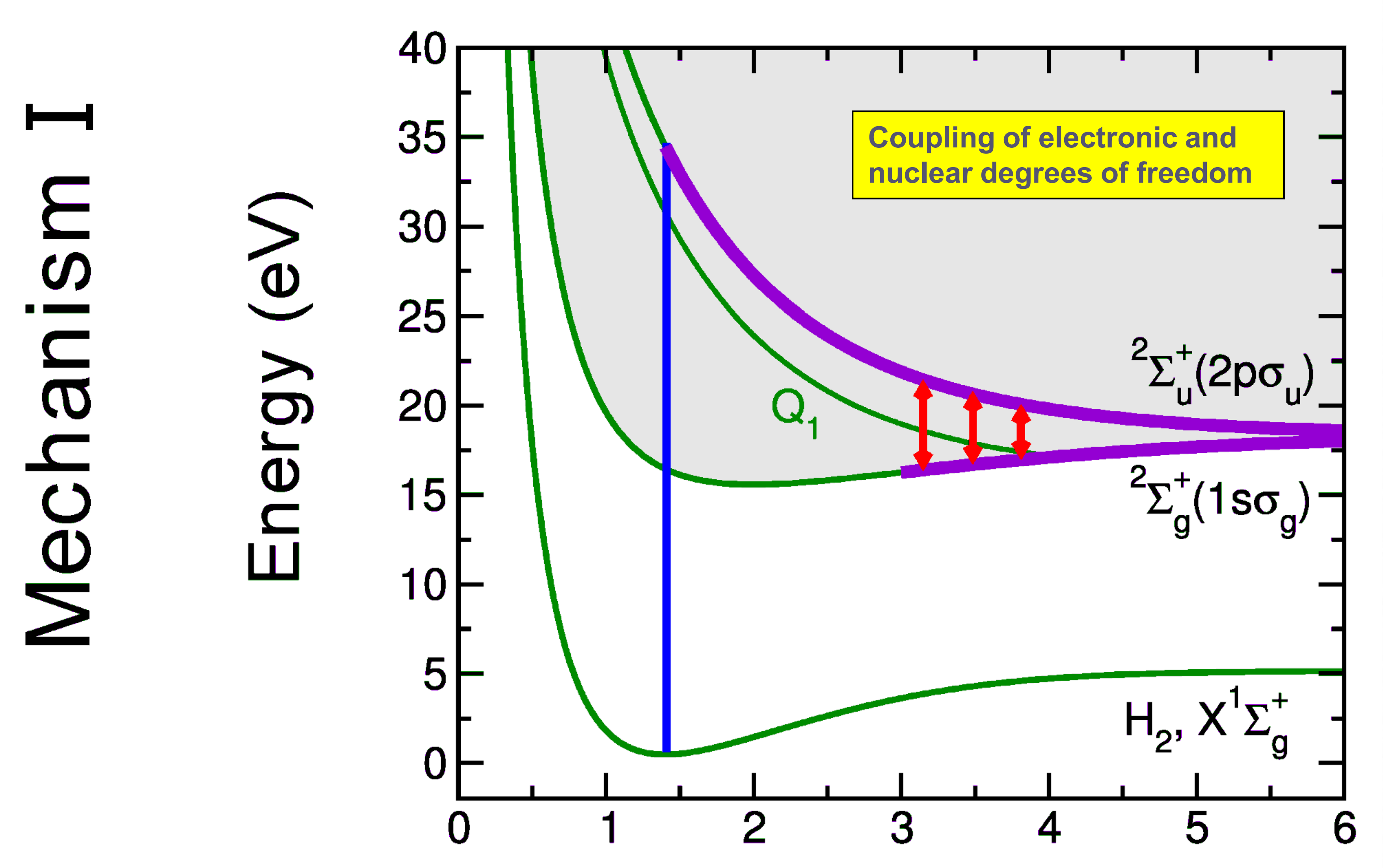
By projecting onto these states the fraction of the wave function that is on the left or right side of the molecule can be determined

Electron localization in XUV-IR dissociative ionization of H_2 and D_2



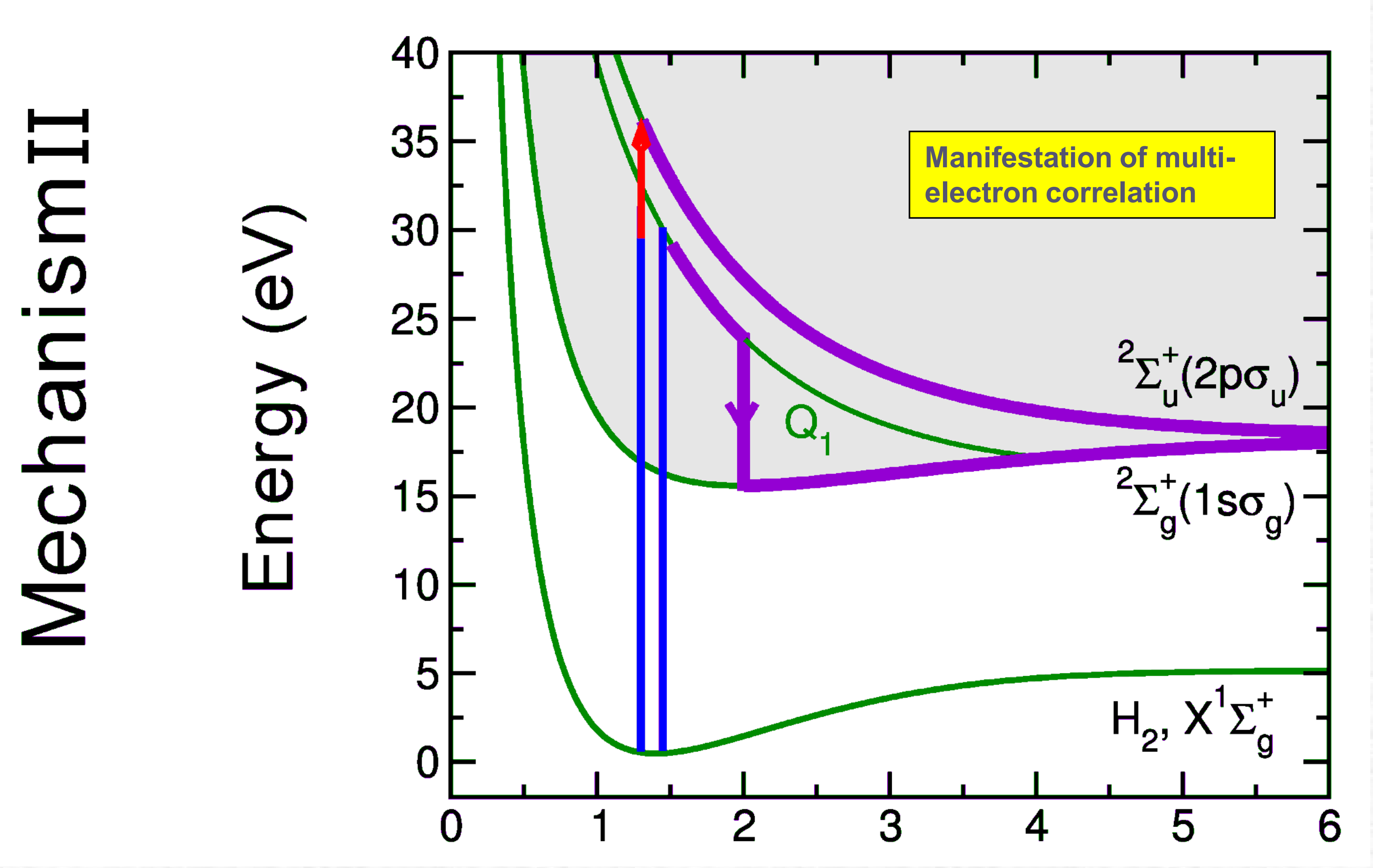
Nature 465, 763 (2010)

Mechanism I:



Mechanism I
Energy (eV)
Coupling of electronic and nuclear degrees of freedom
Q1
$^2\Sigma_u^+(2p\sigma_u)$
$^2\Sigma_g^+(1s\sigma_g)$
$H_2, X^1\Sigma_g^+$
0
5
10
15
20
25
30
35
40
0
1
2
3
4
5
6

Mechanism II:



Conclusion

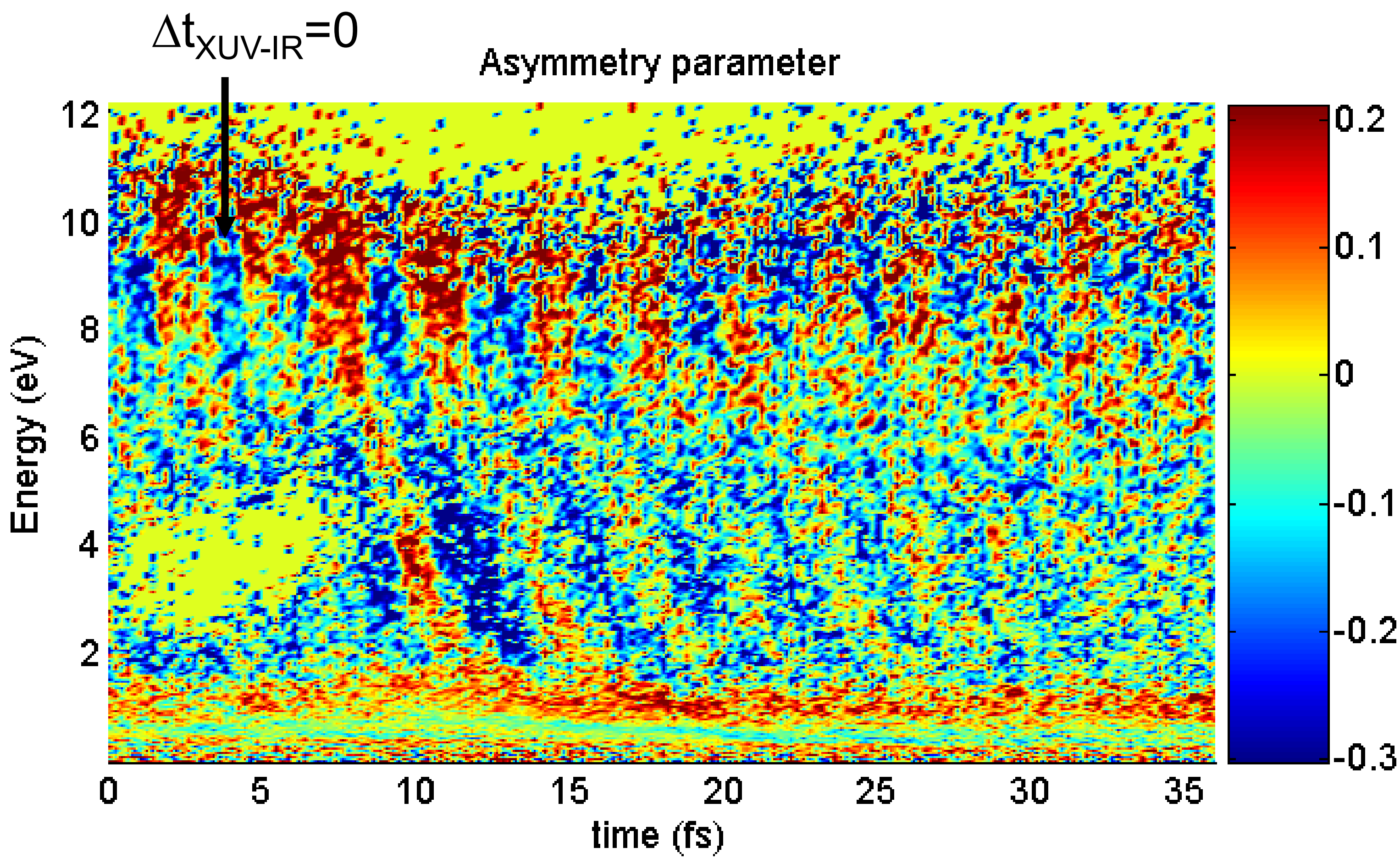
A localization of the electronic charge distribution in H_2 has been measured that depends – with attosecond time-resolution – on the delay between an isolated attosecond XUV “pump” pulse and an IR “probe” pulse

➔Mechanism I: laser-driven population transfer between the $1s\sigma_g$ and the $2p\sigma_u$ electronic states (transition from adiabatic to diabatic dynamics)
coupling of electronic and nuclear degrees of freedom

➔Mechanism II: interference between a dissociation channel involving the Q_1 auto-ionizing states and excitation of the $2p\sigma_u$ dissociative state accompanied by IR interaction with the continuum electron
electron correlation

In addition evidence has been obtained for modifcation of XUV photo-excitation as a result of an IR-laser induced polarization in the ground or ionic state → relevant to the use of x-ray probing of electron localization in molecules

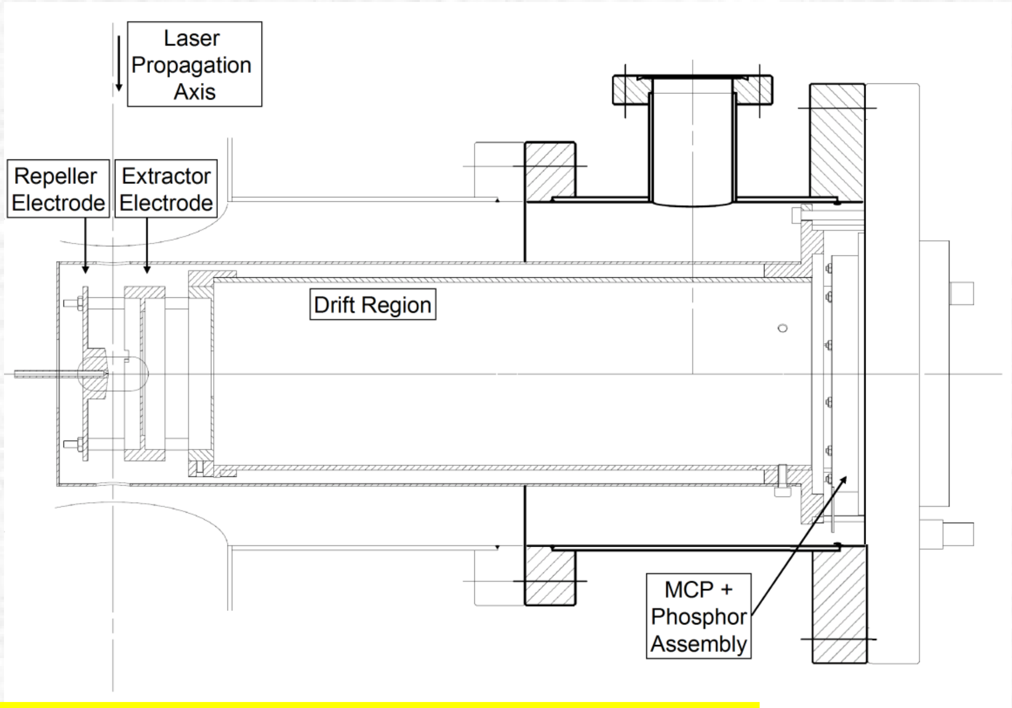
Electron localization in XUV-IR dissociative ionization of H_2 and D_2

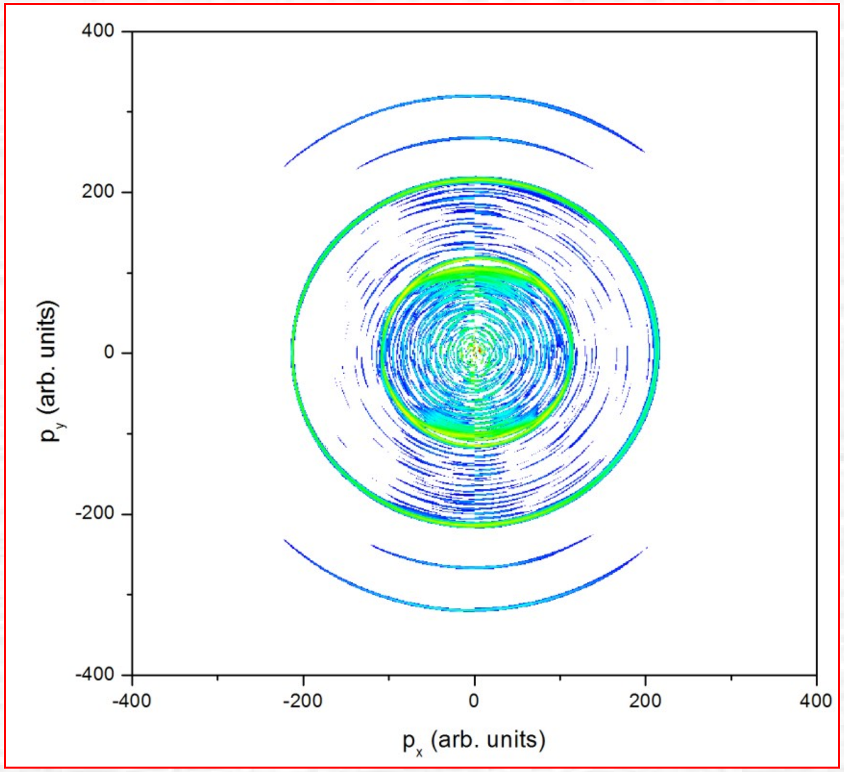


Nature 465, 763 (2010)

Improving VMI Technology:

An imaging spectrometer with integrated gas injection





Est. 1000-fold signal increase over conventional velocity map imaging spectrometers

Multi-photon ionization of Xe at 532 nm; ΔE/E ~ 2%
O.Ghafur et al, Rev. Sci. Instrum. 2009

Where can I find detectors that I can use in bad vacuum? - Google Search - Microsoft Internet Explorer

File Edit View Favorites Tools Help

Back Search Favorites

Address http://www.google.com/search?sourceid=navclient&ie=UTF-8&rlz=1T4ADBF_enNL302NL302&q=Where+can+I+find+detectors+that+I+can+use+in+bad+vacuum%3f

Google Where can I find detectors that I can use Search Bookmarks Find Check AutoFill

Web Images Maps News Shopping Gmail more

Google Where can I find detectors that I can use in bad vacuum? Search Advanced Search Preferences

Web Results **1** - **10** of about **12,000,000** for **Where can I find detectors**

Did you mean: Where can I find ***doctors*** that I can use in bad vacuum?

MS Tip - Techniques used to evaluate **vacuum** leaks in the MS
19 - What techniques or methods do you **use** to detect **vacuum** leaks in your .... A leak **detector** with a sniffer probe **can** also be used to look for leaks in GC ...
www.sisweb.com/index/referenc/tip19.htm - 19k - Cached - Similar pages

Vacuum detector leak On GlobalSpec
By using our SpecSearch product, you **can** search the **vacuum detector** leak related ... A mobile helium leak **detector** which is equally well suited for **use** in ...
sensors-transducers.globalspec.com/Industrial-Directory/**vacuum_detector**_leak - 85k - Cached - Similar pages

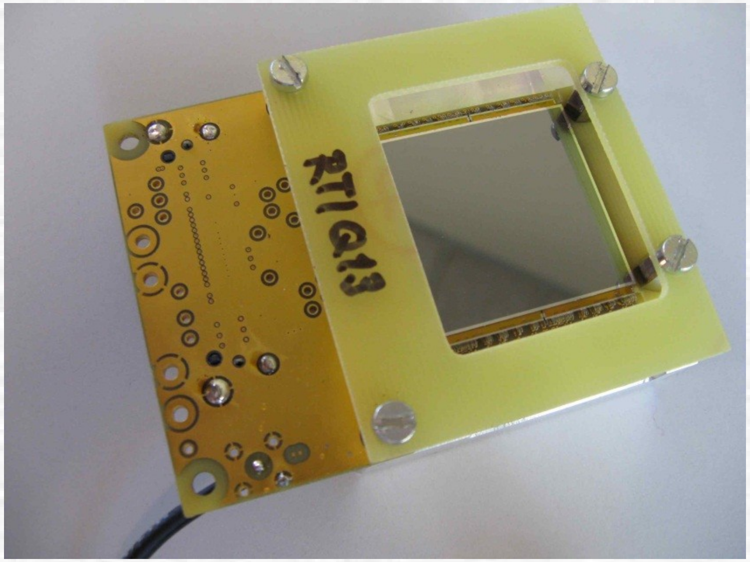
Testing the **Detectors** For CHARM - My Solar-Called Life: Solar Week ...
Also, the microchannel plates in our **detectors can** be damaged if we power up the **detectors** at ... To pump out our **vacuum** chamber we **use** two kinds of pumps. ...
www.solarweek.org/CS/blogs/solar_week_scientist_blog/archive/2007/02/27/892.aspx - 23k - Cached - Similar pages

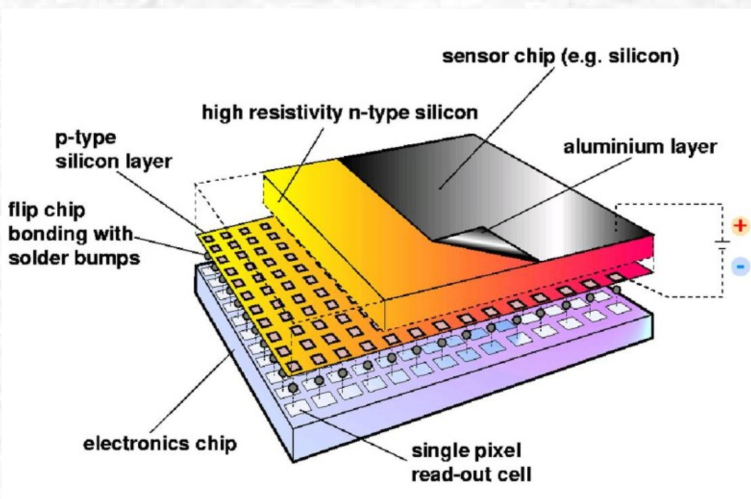
Leak **detector** - **vacuum** pump | leak detection | a leak detection ...
Mounted on the end of a flexible steel pipe, the ultrasonic sensor **can** access ... The simple to **use** Model 21-070 Gas Leak **Detector** is engineered for easy, ...
www.directindustry.com/industrial-manufacturer/leak-**detector**-66297.html - 154k - Cached - Similar pages

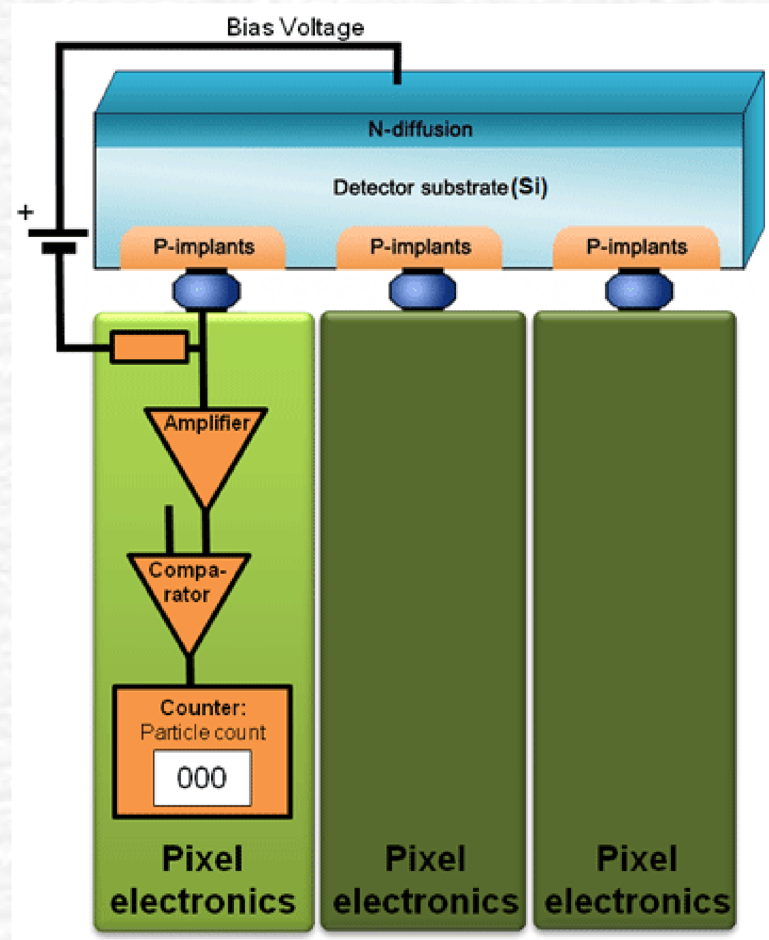
[PDF] The SEM examination of geological samples with a semiconductor ...
File Format: PDF/Adobe Acrobat - View as HTML

Doctors
Find doct
Addresses
www.Doct

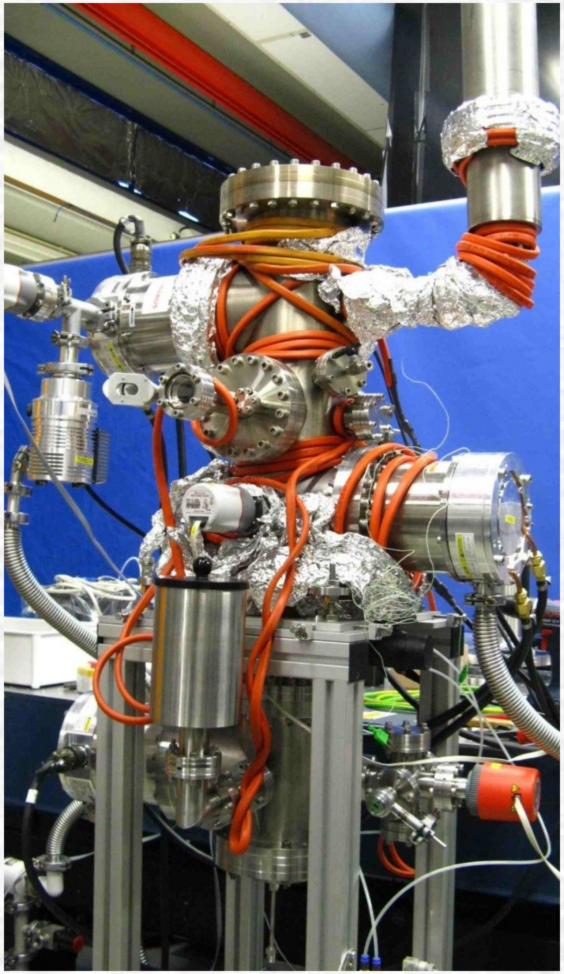
Improving VMI Technology – 2
Use of the Medipix detectors (with NIKHEF)

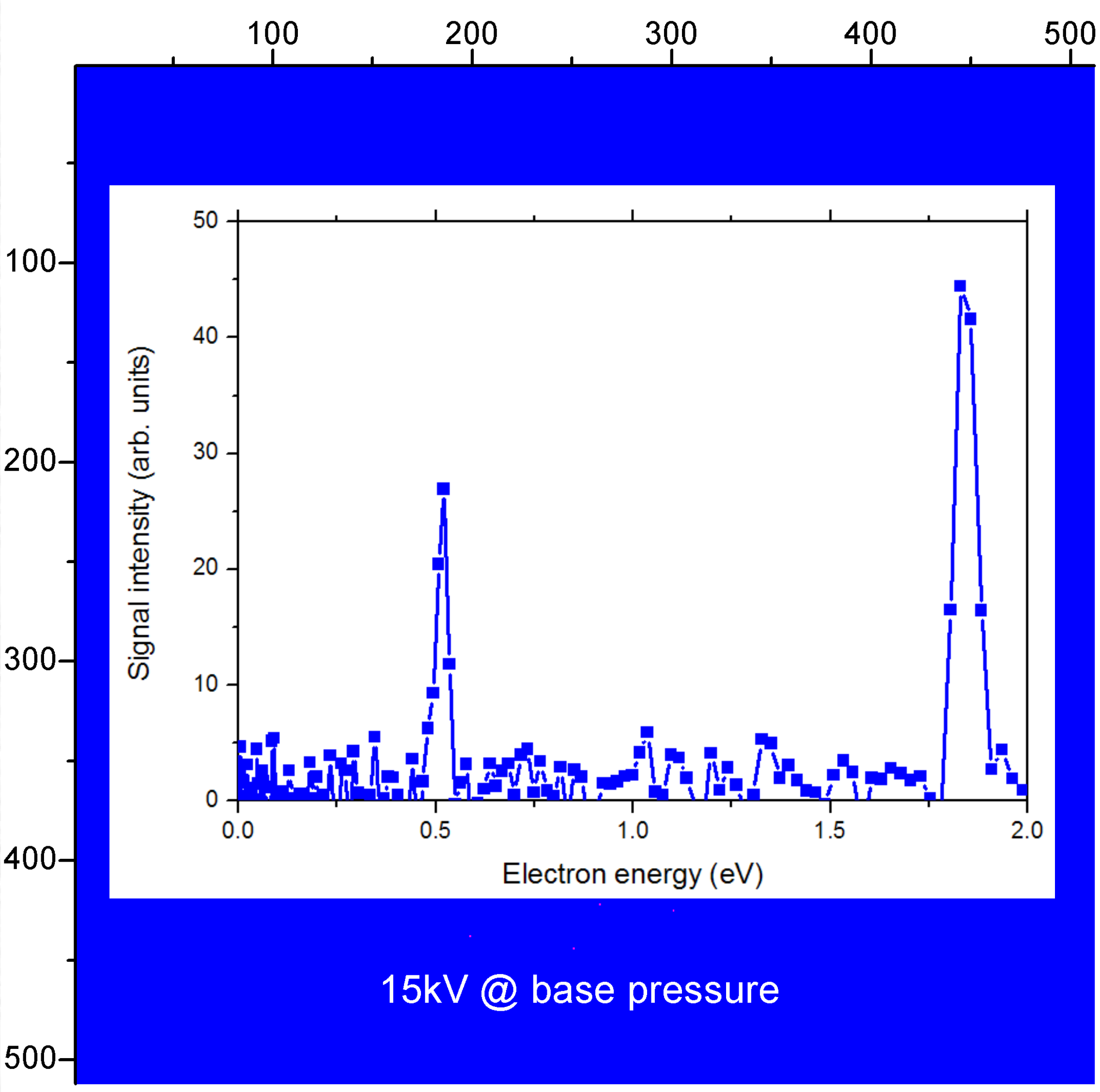






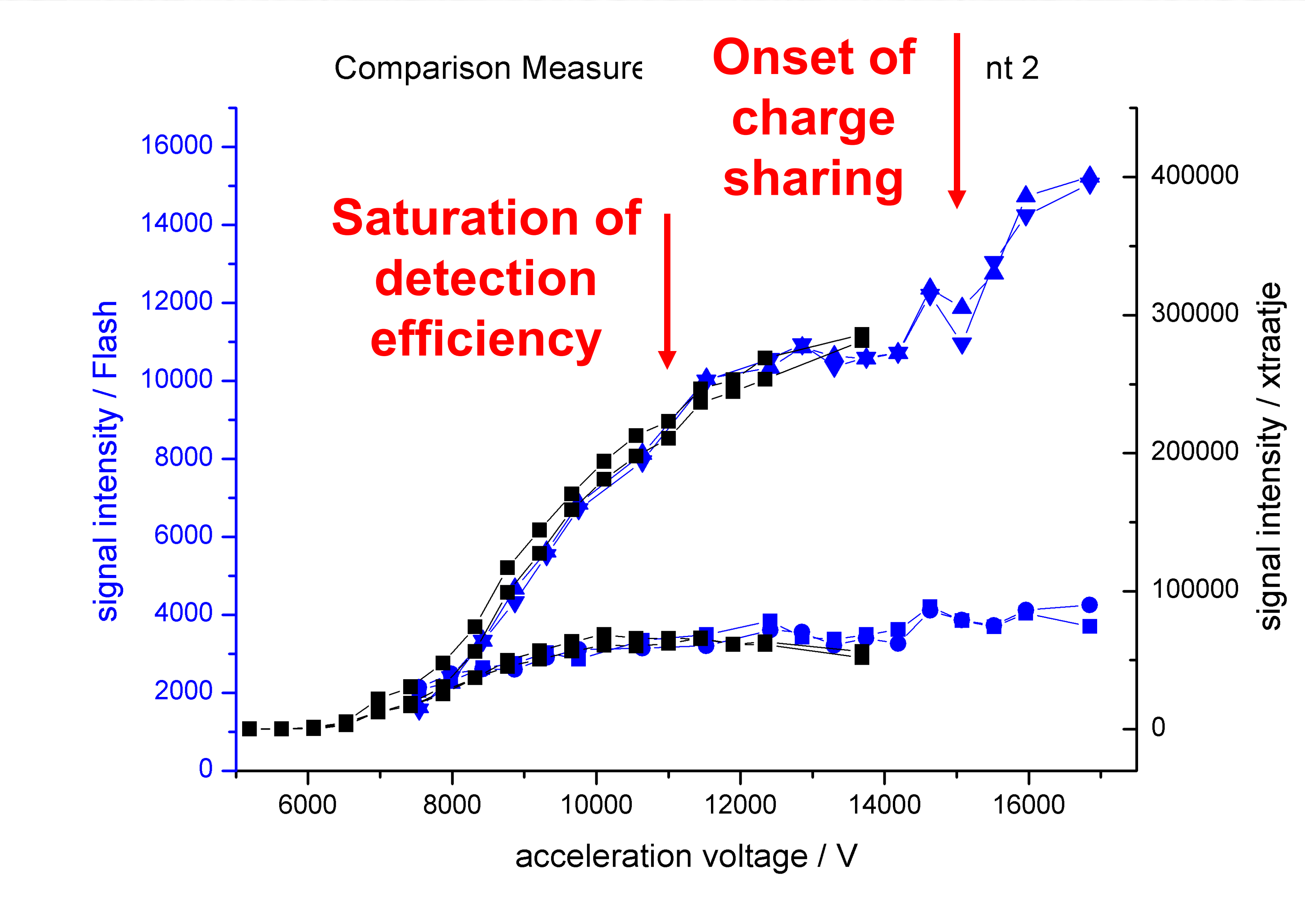
Velocity map imaging using the Medipix Detector



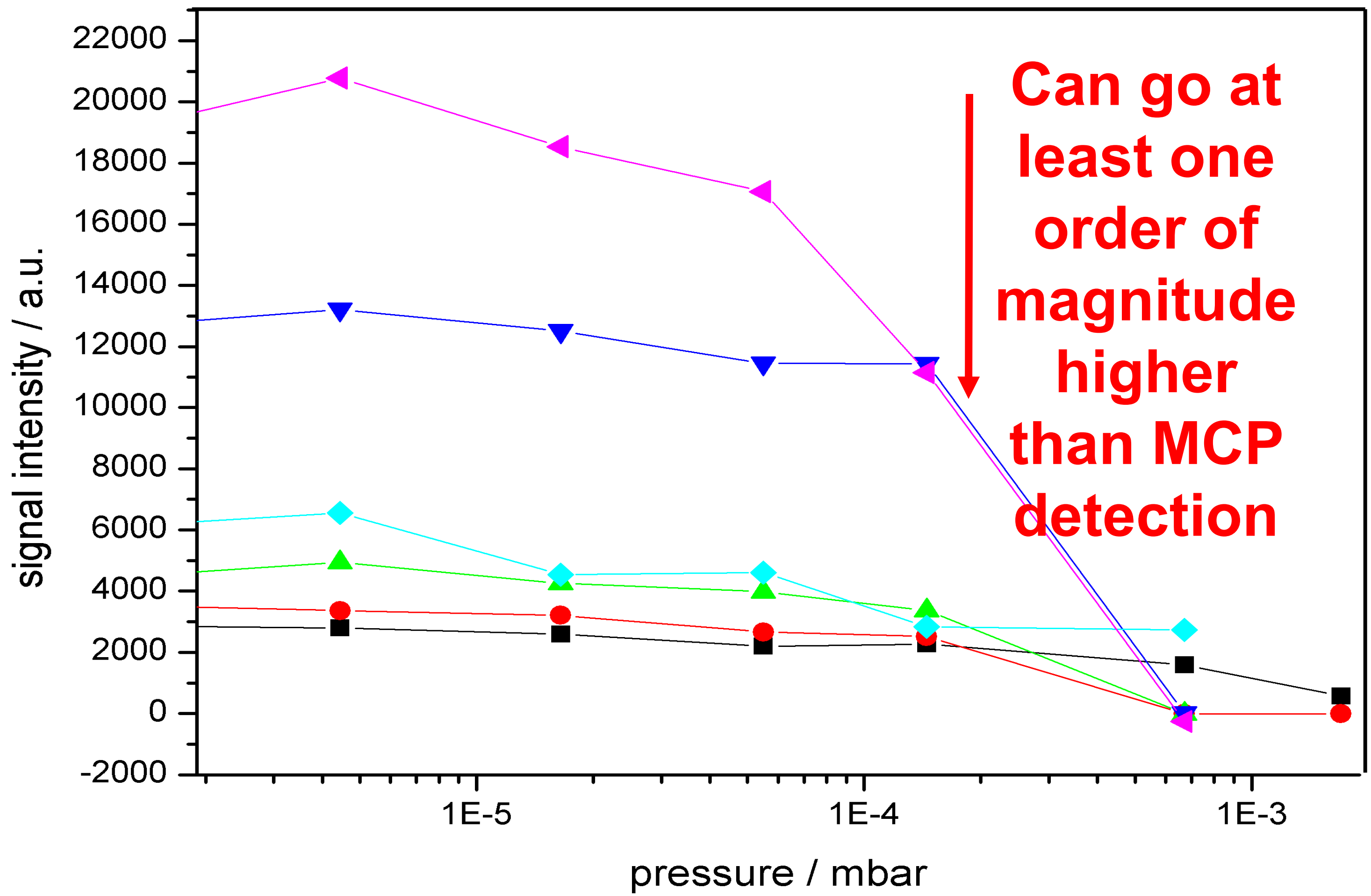


Test-experiment: 4-photon ionization of Xe, producing electrons 0.53 and 1.83 eV kinetic energy (before acceleration)

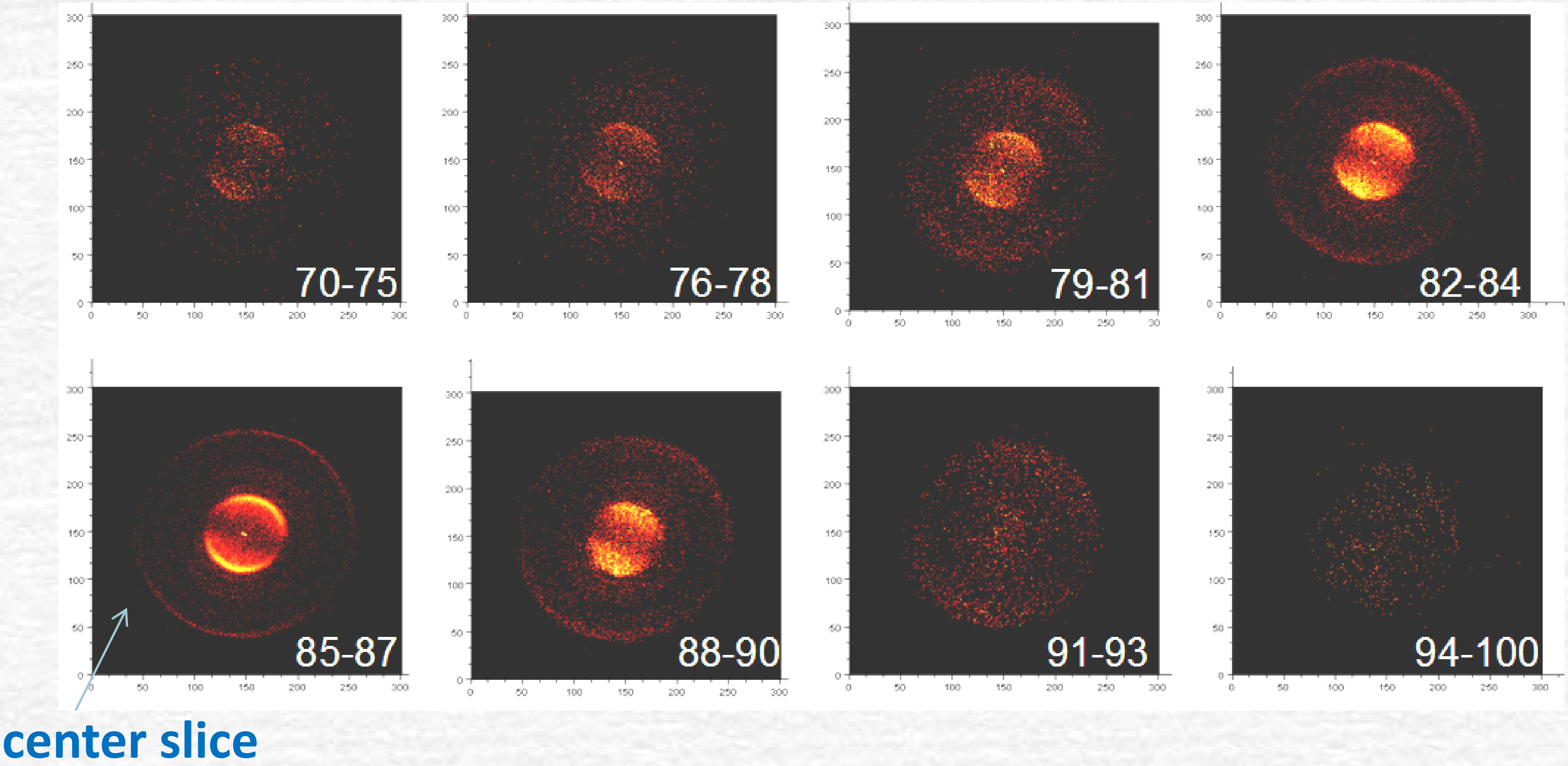
Influence of electron kinetic energy



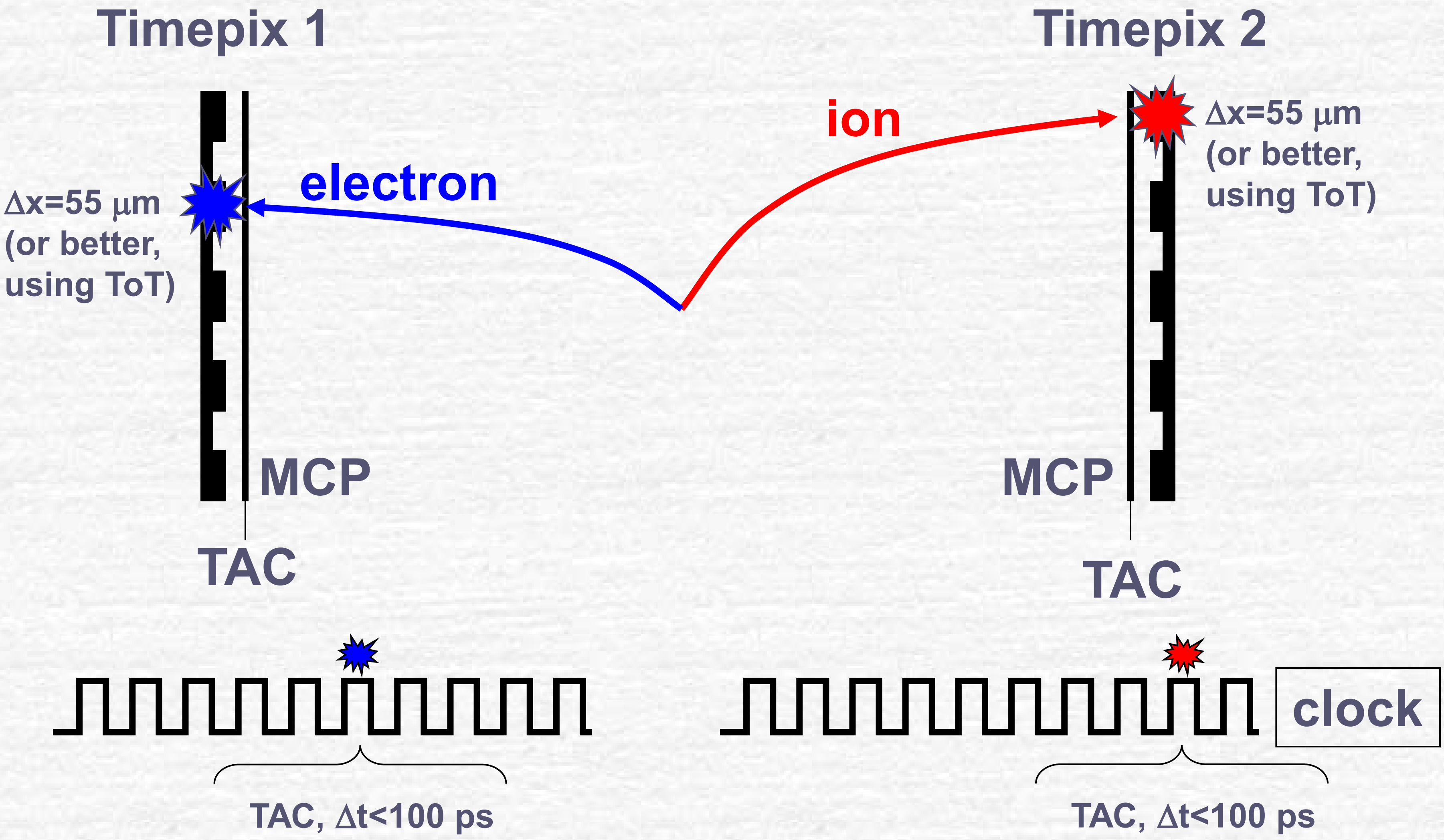
Pressure dependence (He)



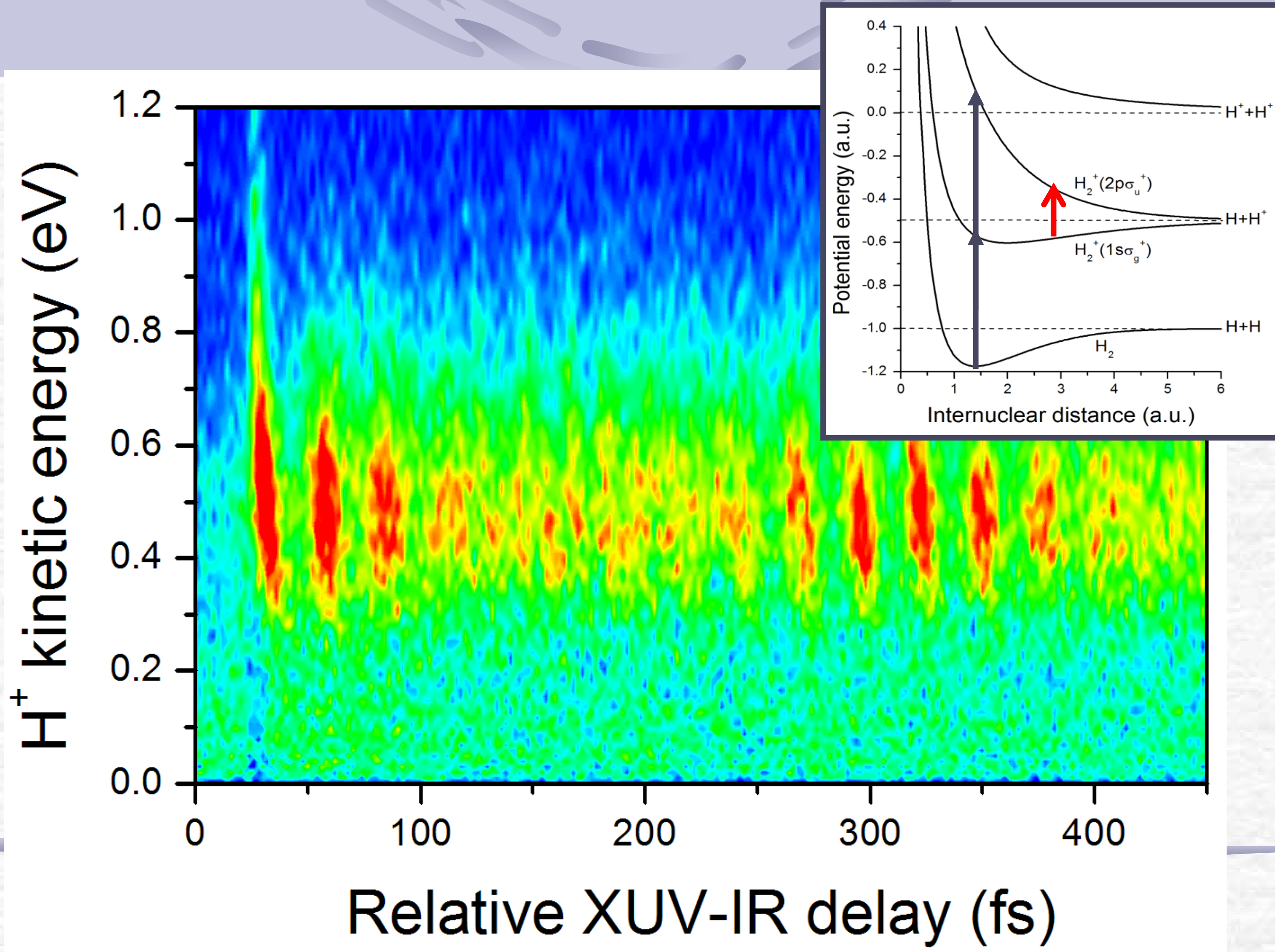
Exploiting the timing capabilities of the Timepix detector : slicing



COLTRIMS using Medipix



Vibrational wave packet measured by R-dependence of bond softening



Novel XUV/x-ray sources (HHG and FELs) allow to push atomic & molecular science beyond the present state-of-the-art

❖ Temporal properties → Electron dynamics

Example:

Attosecond time-resolved pump-probe spectroscopy in H_2

Wavepacket interferometry experiments give access to attosecond electron dynamics

❖ Wavelength properties → Nuclear dynamics

Example:

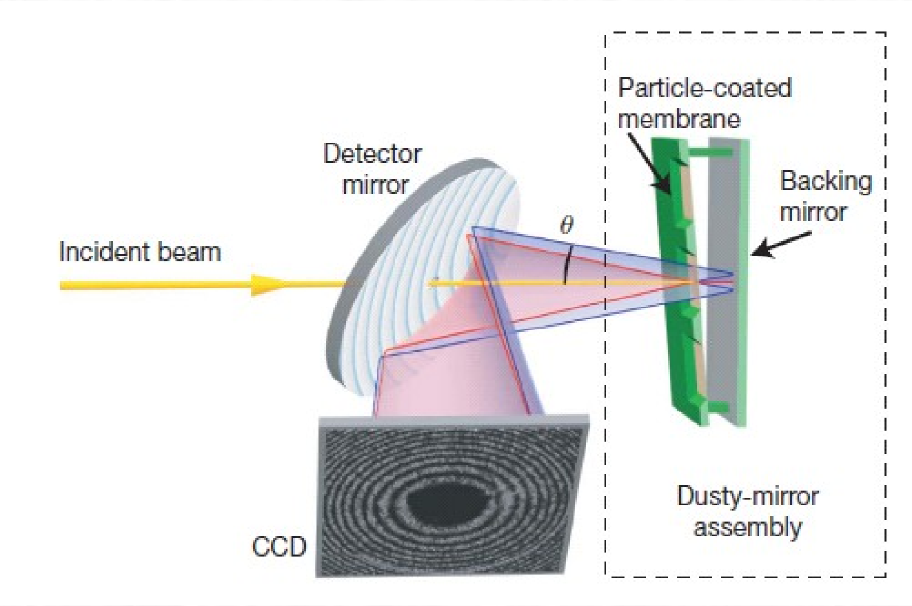
Molecular frame photo-emission gives access to (time-dependent) orbital and structural information – pump-probe spectroscopy at FLASH and using an HHG source

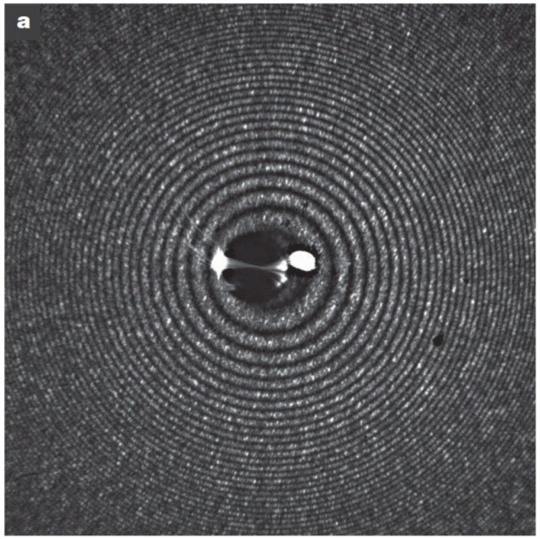
2. Using measurements of MF-PADs to reveal time-dependent structural changes

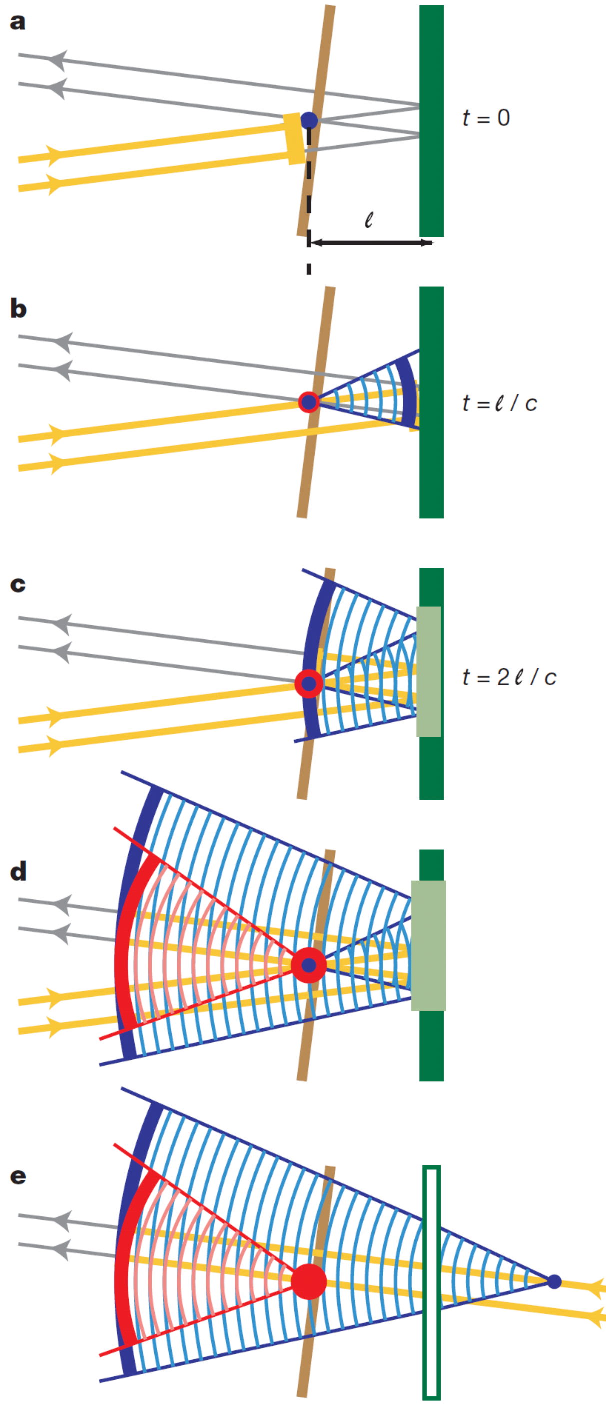
- **Pump-pump-probe experiments at FLASH and LCLS: XUV ionization of dissociating, laser-aligned molecules**

- **Pump-probe experiments using a HHG source: MF-PADs for XUV ionization of laser-aligned molecules**

Femtosecond time-delay holography at FLASH (Chapman et al.)



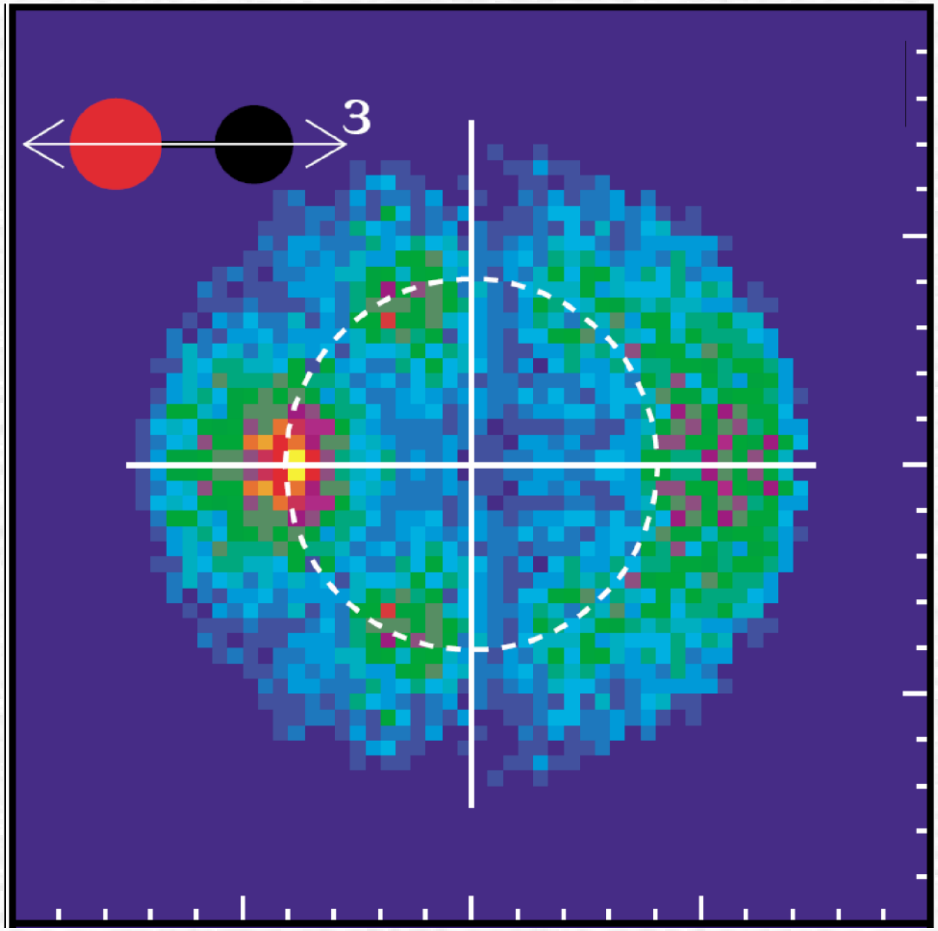




Time-delay x-ray hologram of 140 nm polystyrene spheres performed with 32 nm FLASH radiation

H. Chapman, Nature 448, 676 (2007)

Using intra-molecular electron diffraction and interference to measure (time-resolved) molecular structure

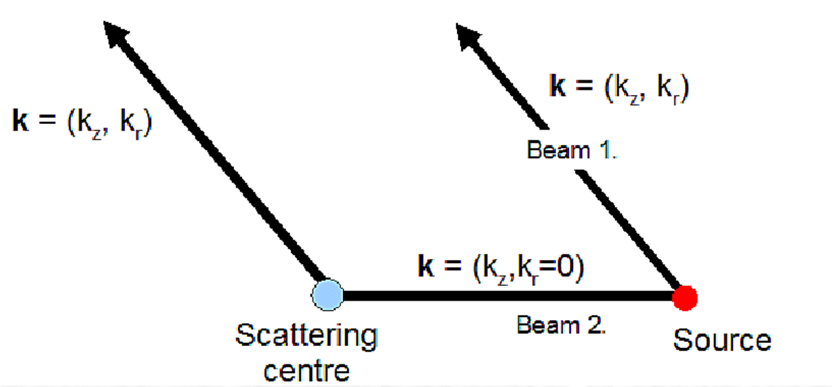


Landers *et al*, Phys. Rev. Lett. 87, 013002 (2001)

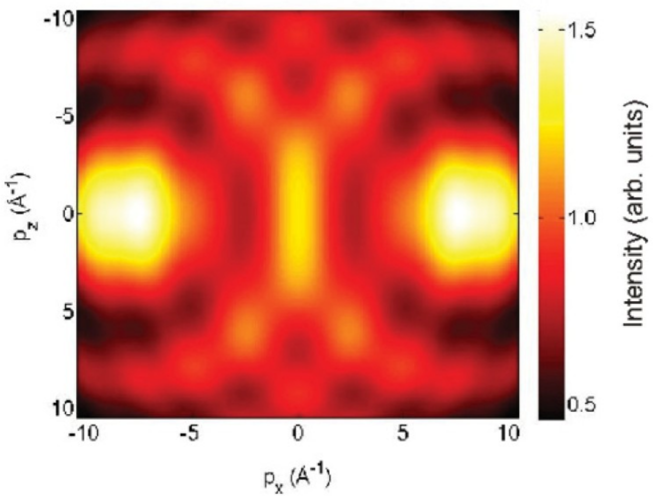
C(1s) core-shell photo-emission from CO using 294 to 326 eV radiation.

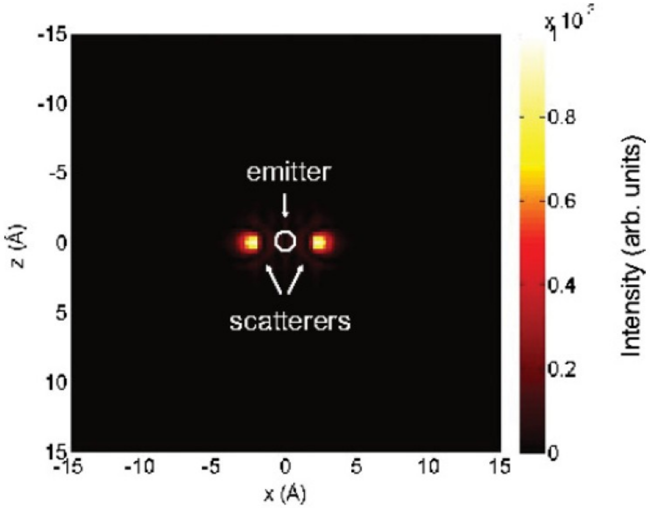
Away from the Carbon atom (black) the angular distribution is relatively unstructured.

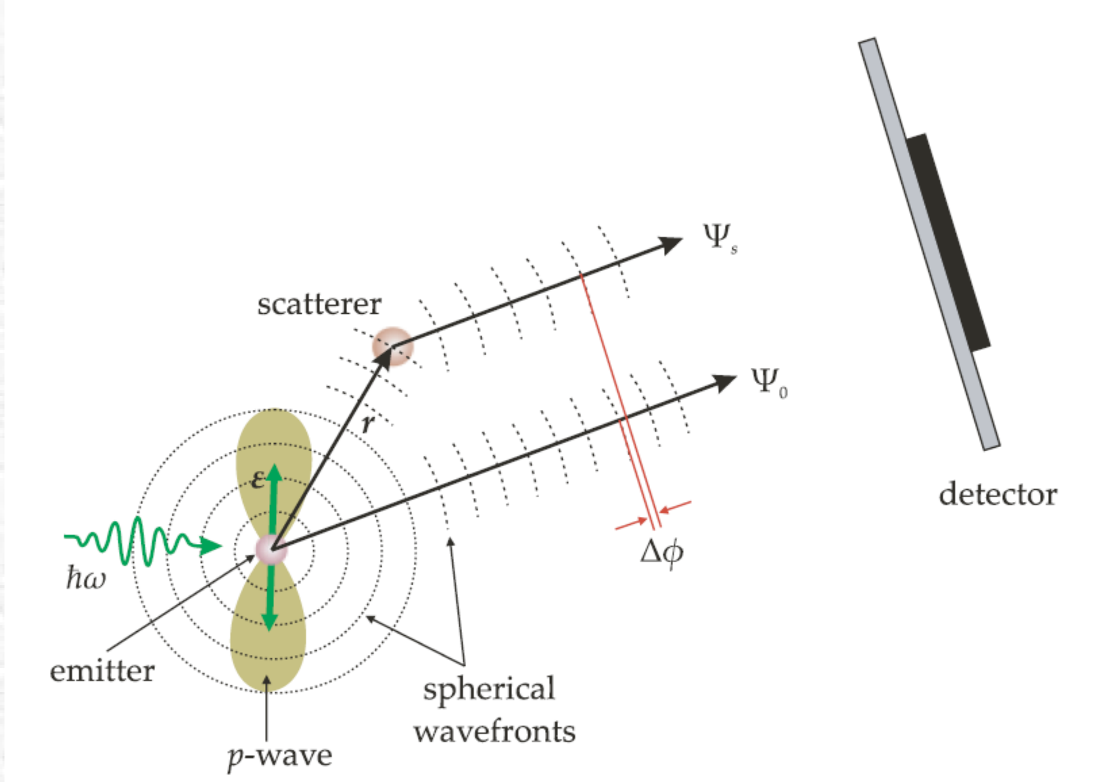
In the direction of the Oxygen atom (red) a diffraction structure is observed.



Using intra-molecular electron diffraction and interference to measure (time-resolved) molecular structure

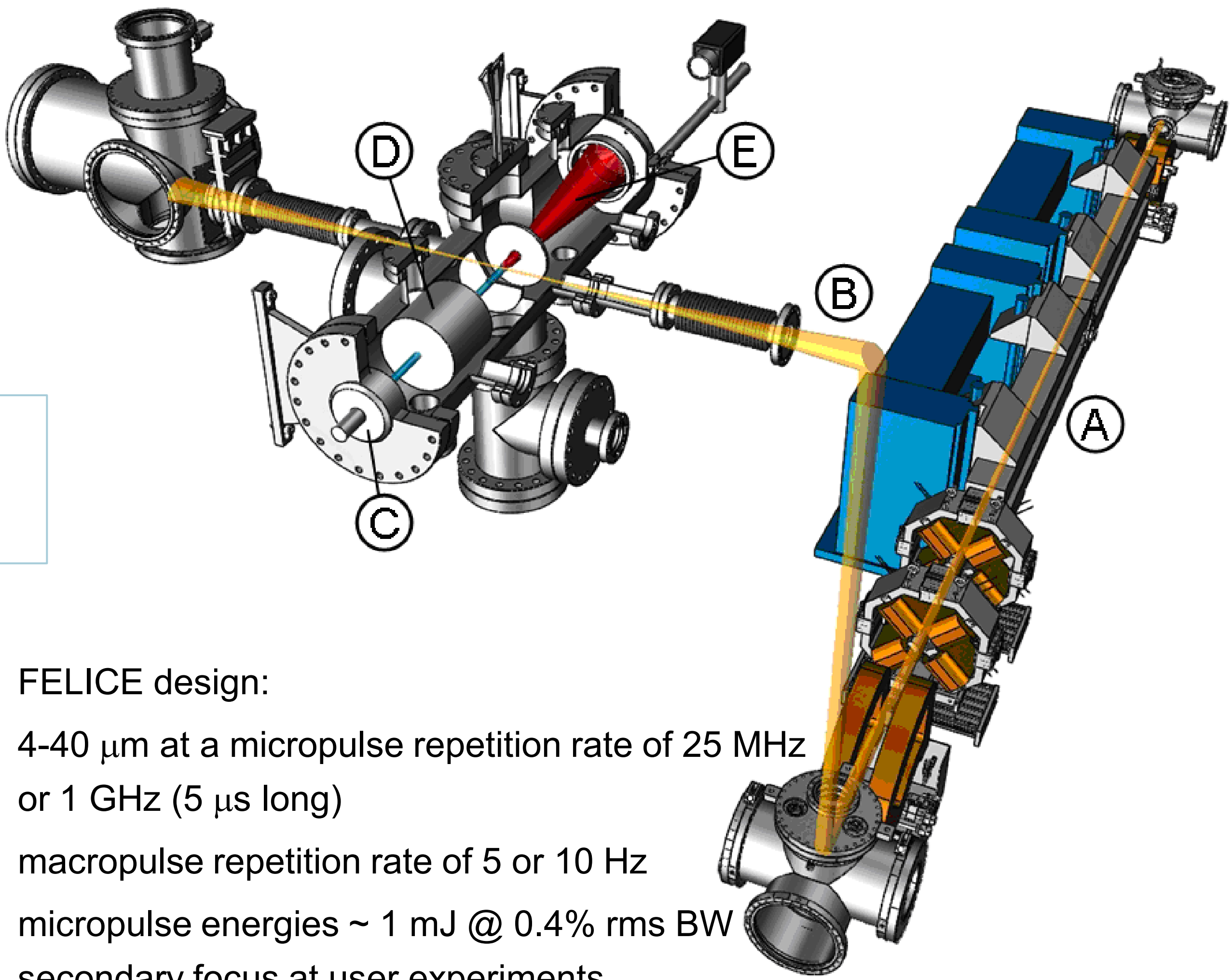






F. Krasniqi et al., Phys Rev. A 81, 033411 (2010)

Proof of concept:
Shoot electron at its own atom



N.B. Macroscopically movable along laser focus

FELICE design:

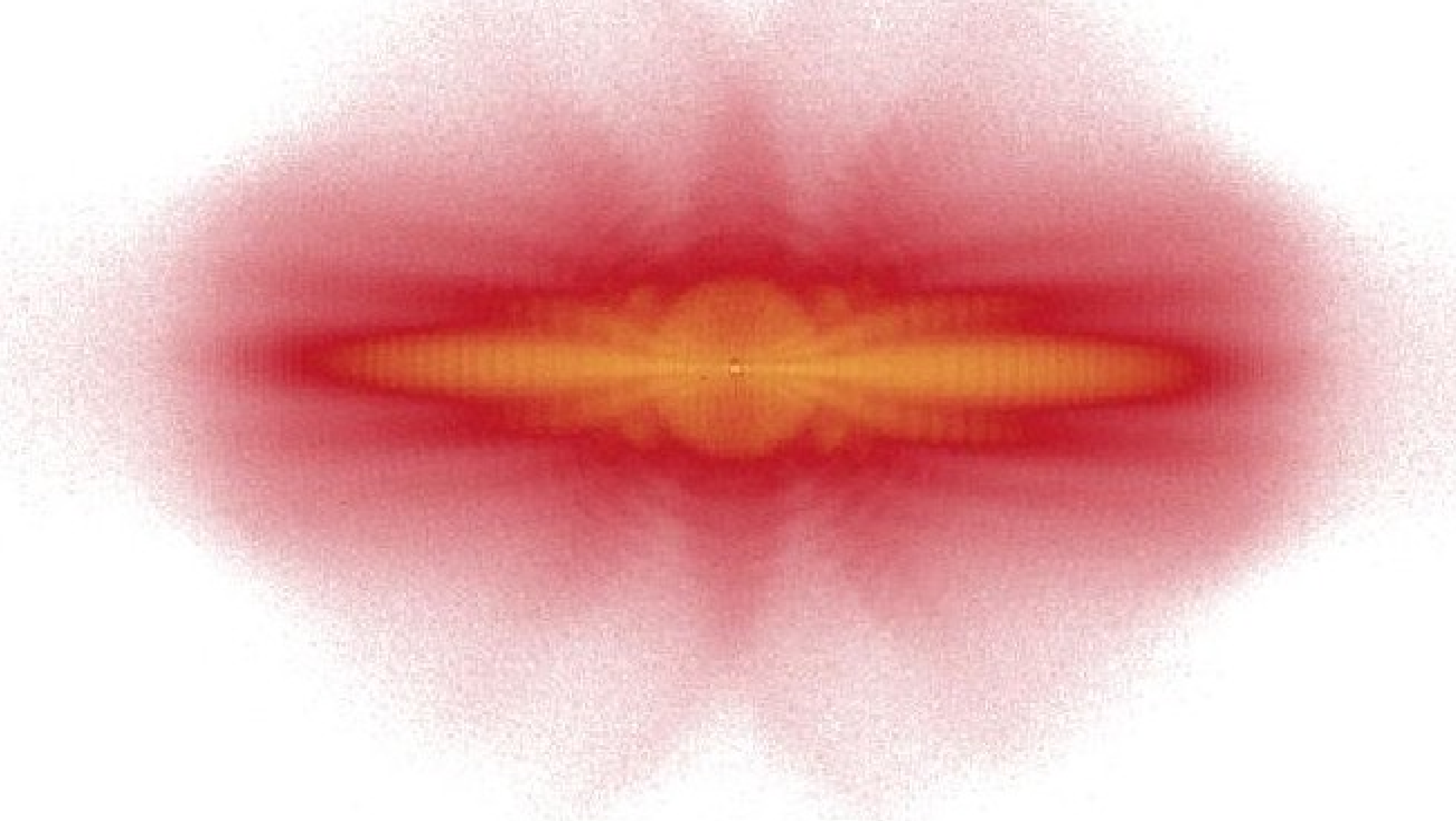
4-40 μm at a micropulse repetition rate of 25 MHz or 1 GHz (5 μs long)

macropulse repetition rate of 5 or 10 Hz

micropulse energies ~ 1 mJ @ 0.4% rms BW

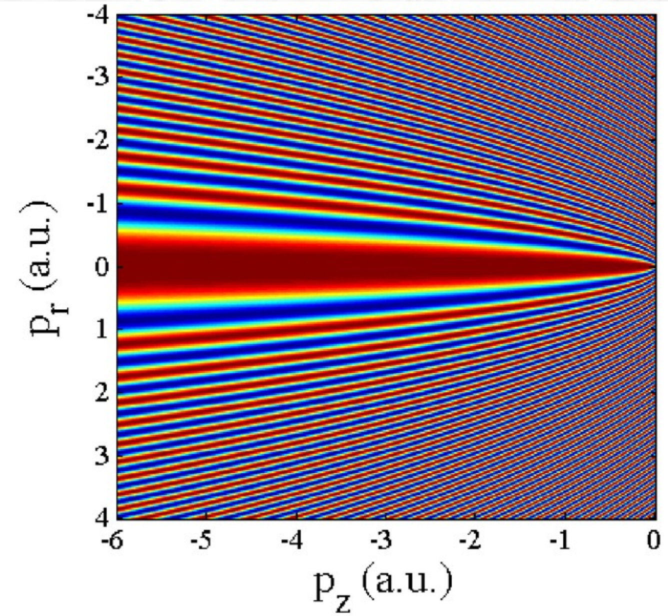
secondary focus at user experiments

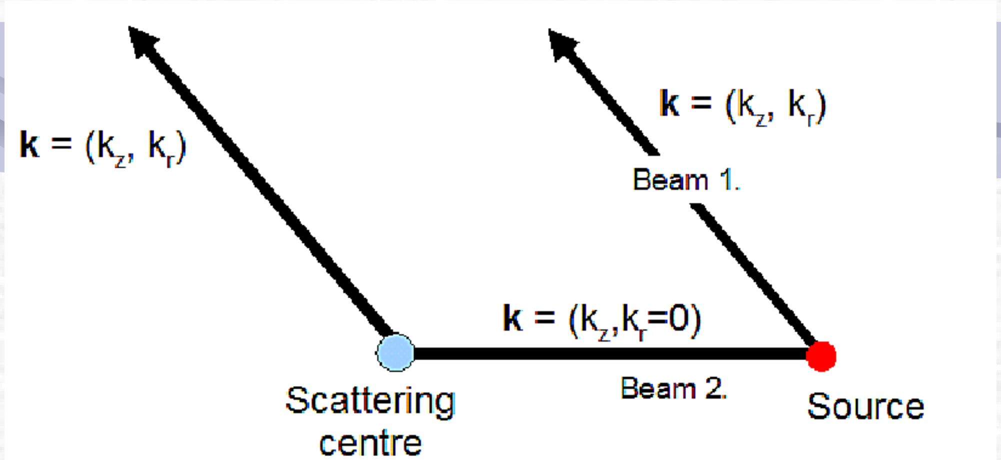
Proof of concept:
Shoot electron at its own atom



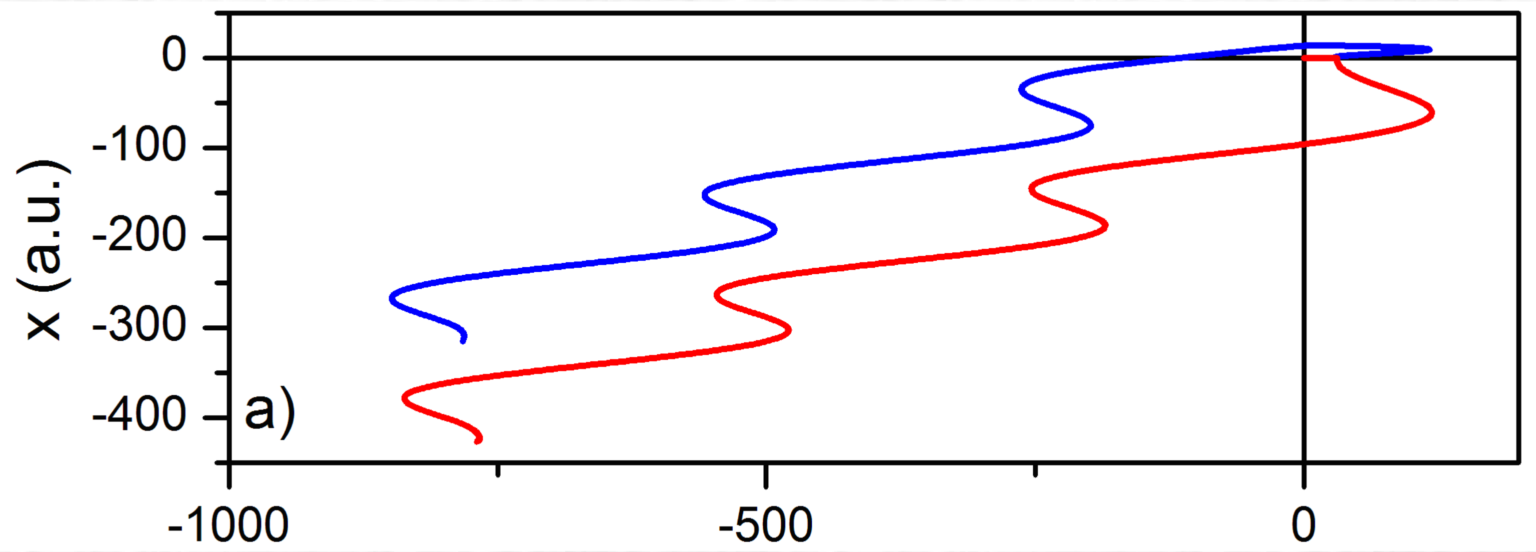
Huismans et al., Science 331, 61 (2011)

Strong-field ionization of Xe(6s) using 7 μm radiation

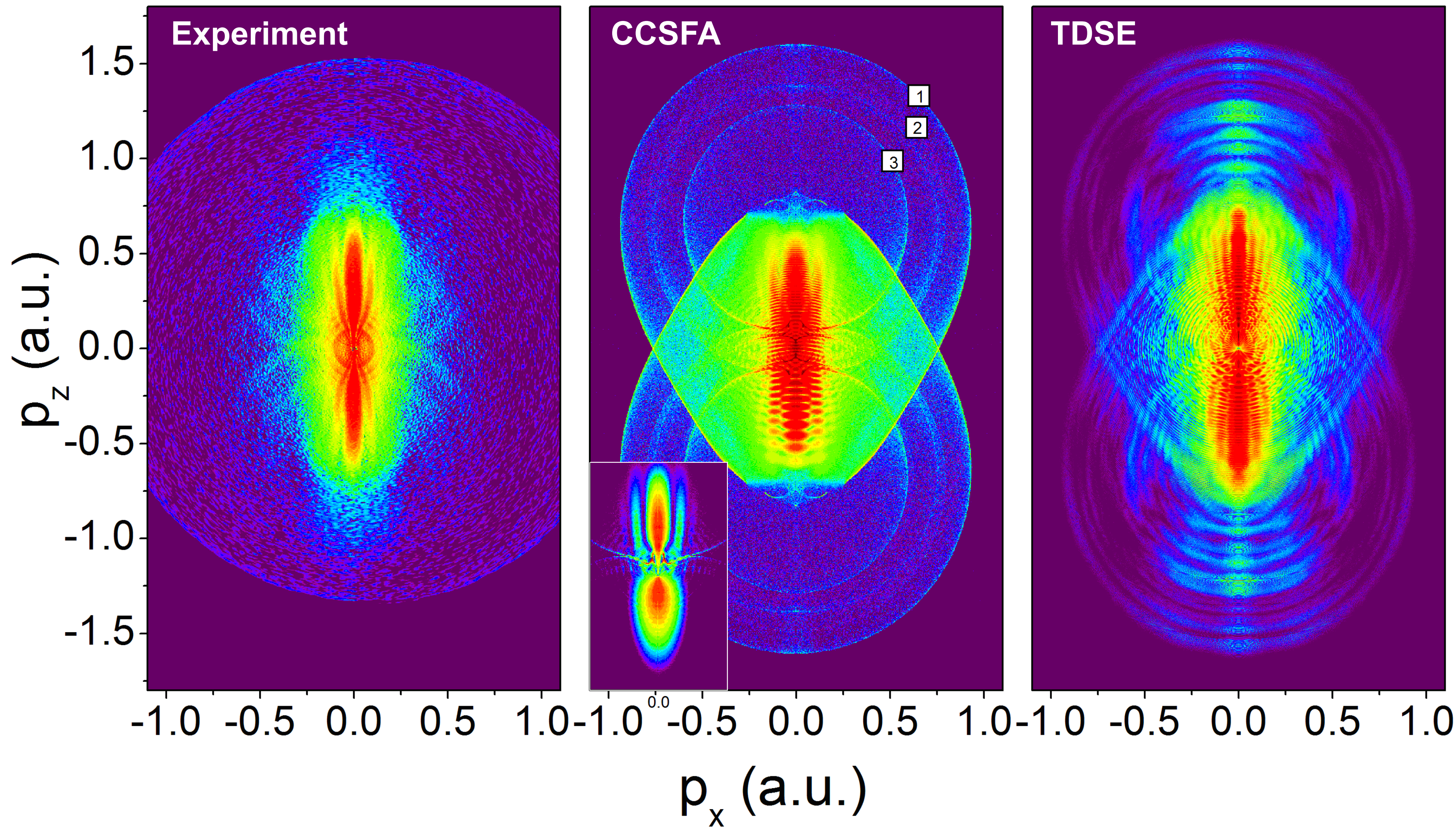




Tunnel ionization produces a source that emits a reference and signal wave



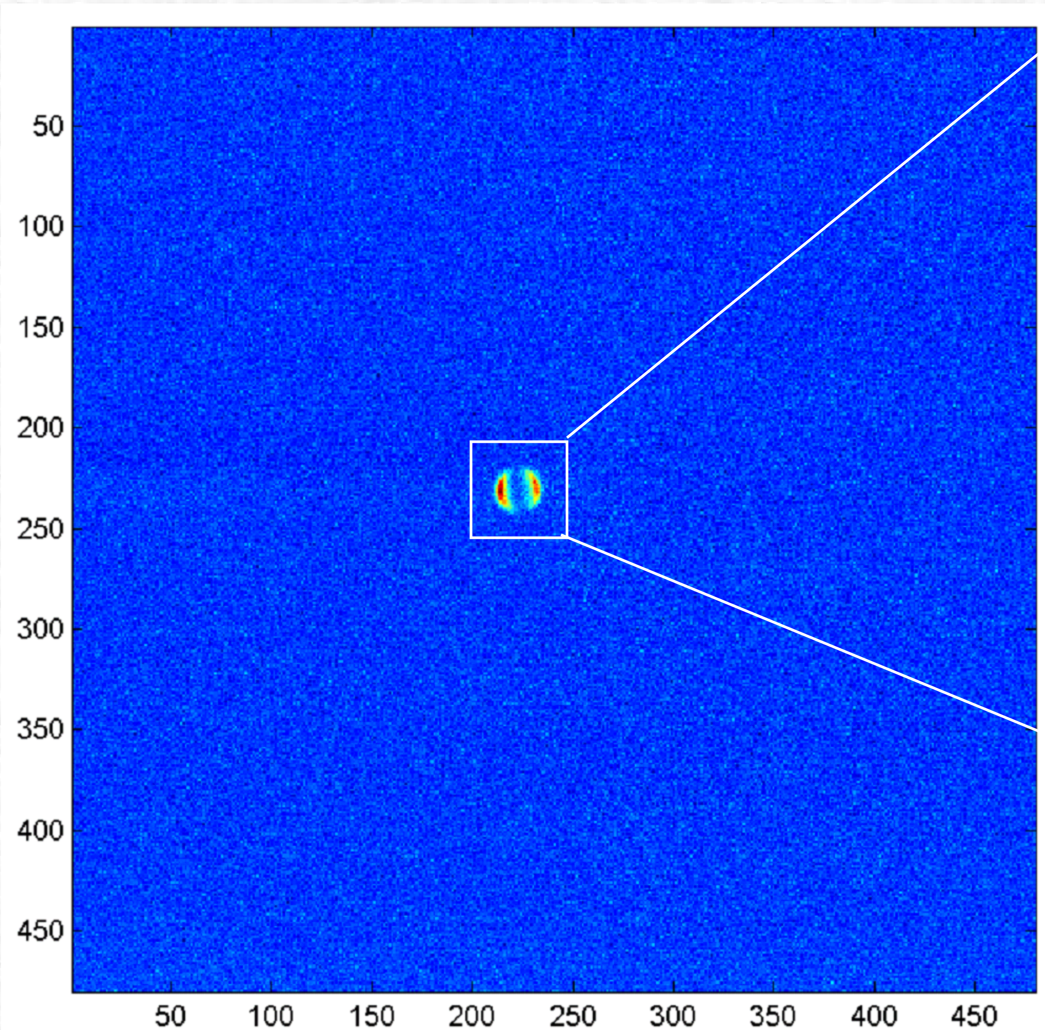
Strong-field ionization of Xe(6s) using 7 μm radiation

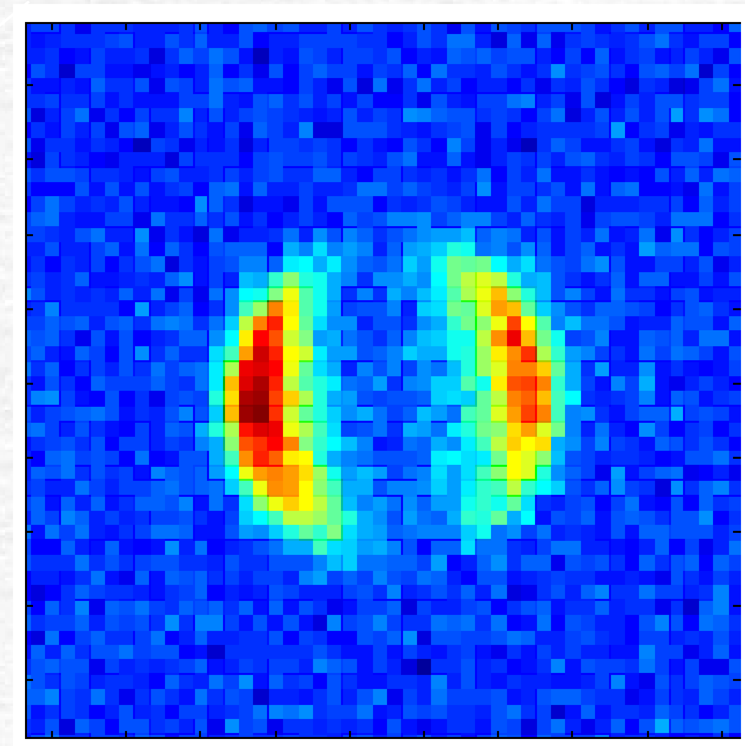


Huismans et al., Science 331, 61 (2011)


FLASH in Hamburg

Explore the utility of velocity map imaging at the FEL (FLASH Campaign 2007)

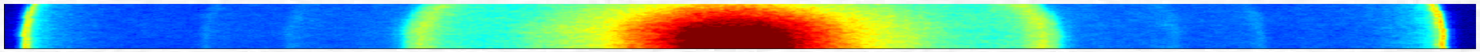


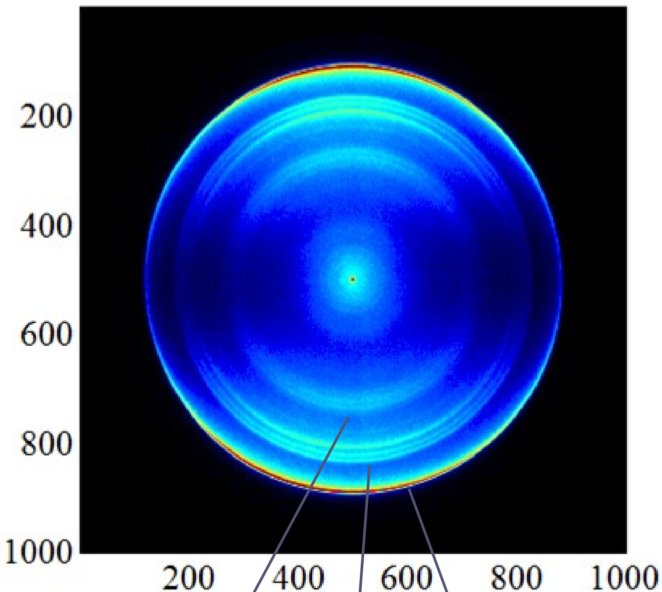


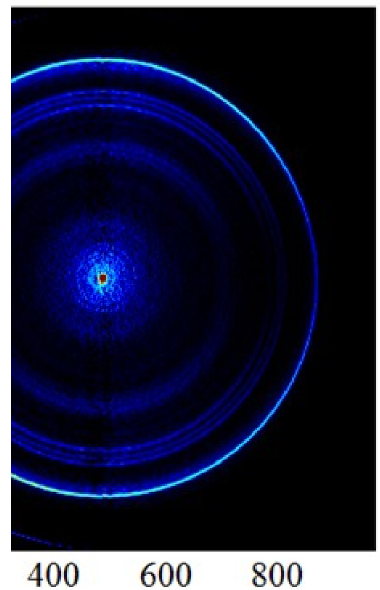
Recoil of D_2^+ in 46 eV photo-ionization of D_2 molecules

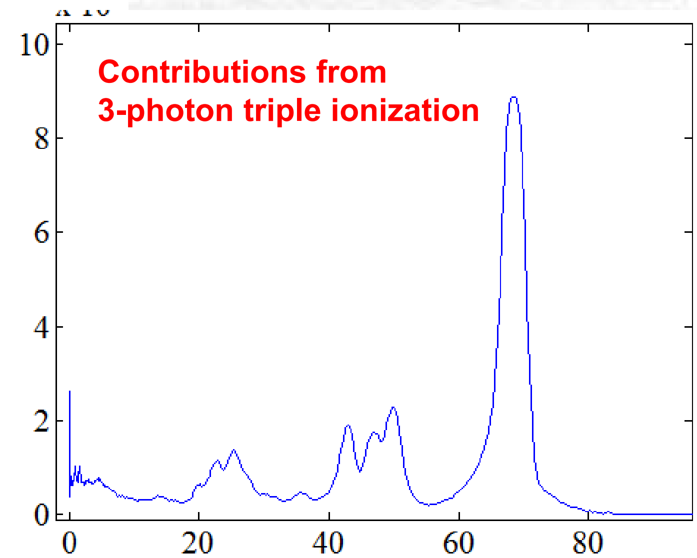
P. Johnsson et al., J. Mod. Opt. 55, 2693 (2008)

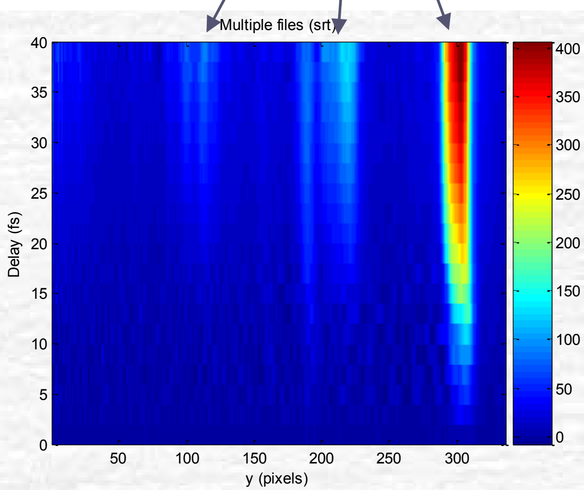
Neon PAD spectra (June 2009)









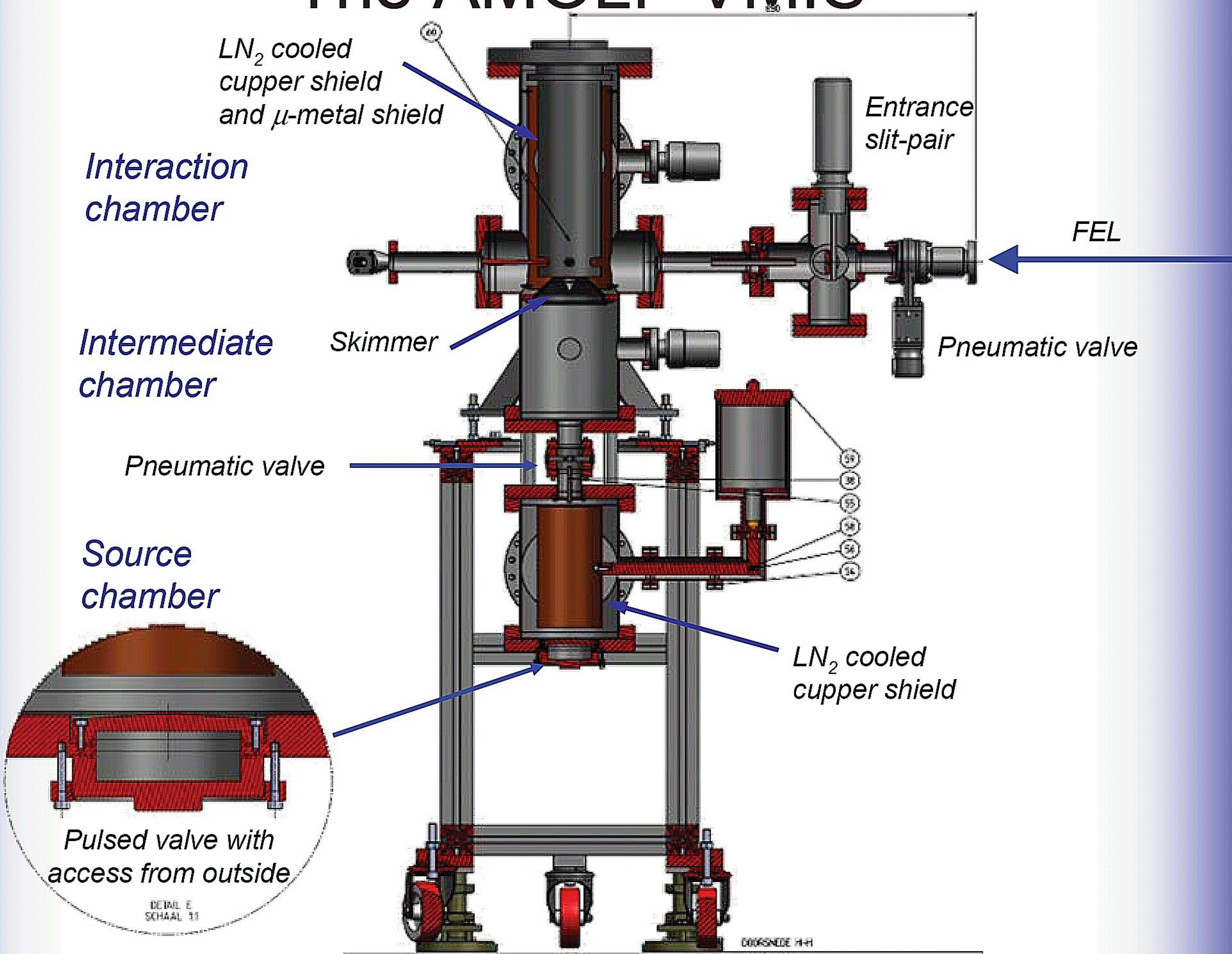


Sorted data using GMD: Allow to determine which channel is linear or non-linear with FEL energy

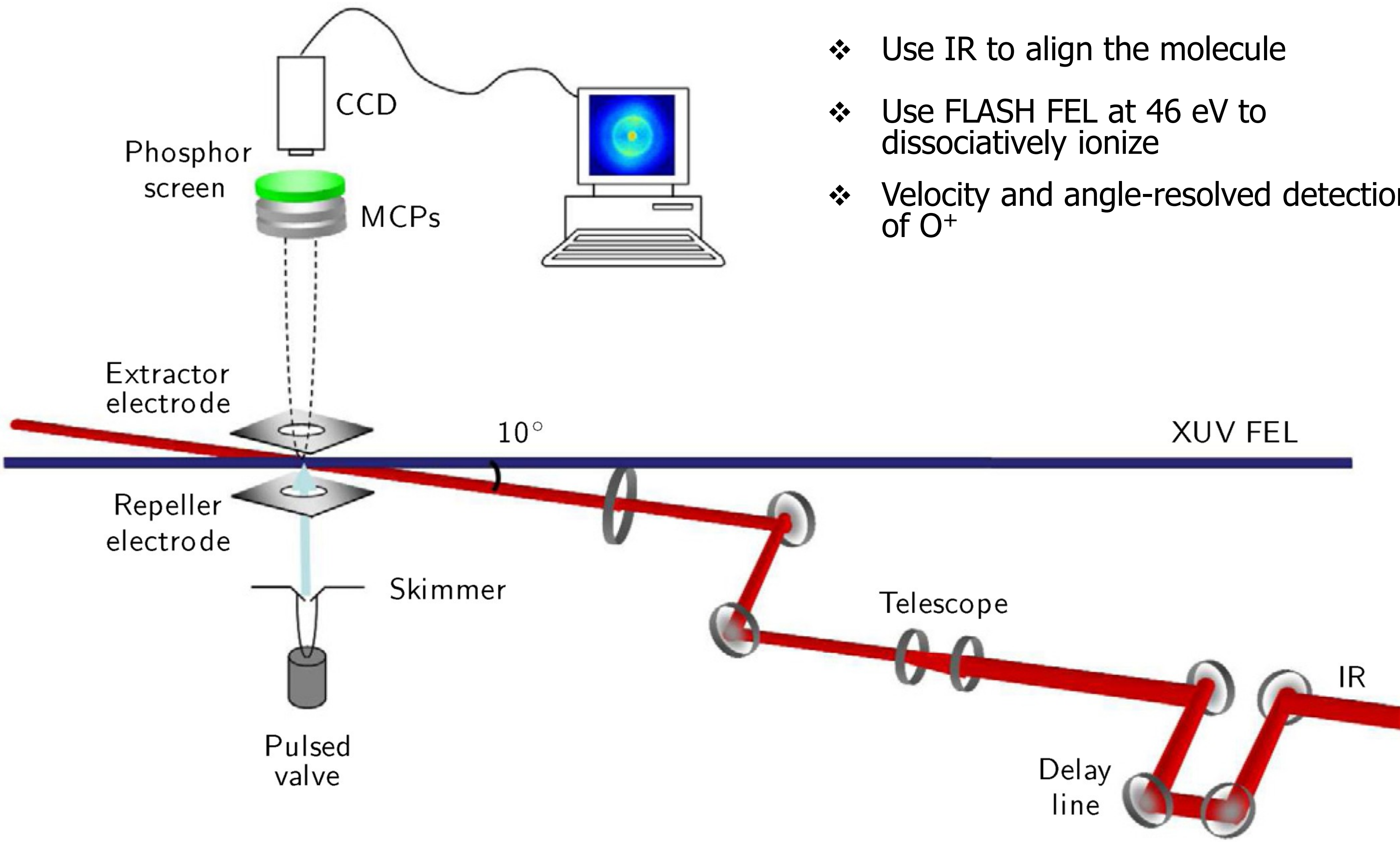
A. Rouzee et al, (submitted)

1	69,3 eV
2	49,9
3	46.9
4	42.8
5	35.6
6	25.4
7	22.7
8	20.0
9	1.6
10	0.9

The AMOLF VMIS



Pump-probe experiment on CO_2 alignment (FLASH Campaign 2008, BL2)

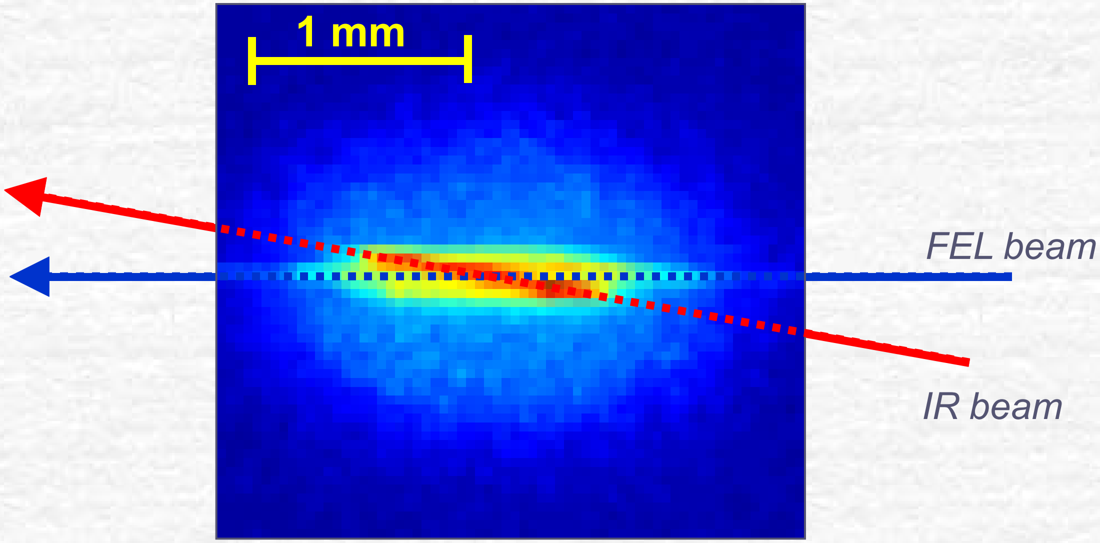


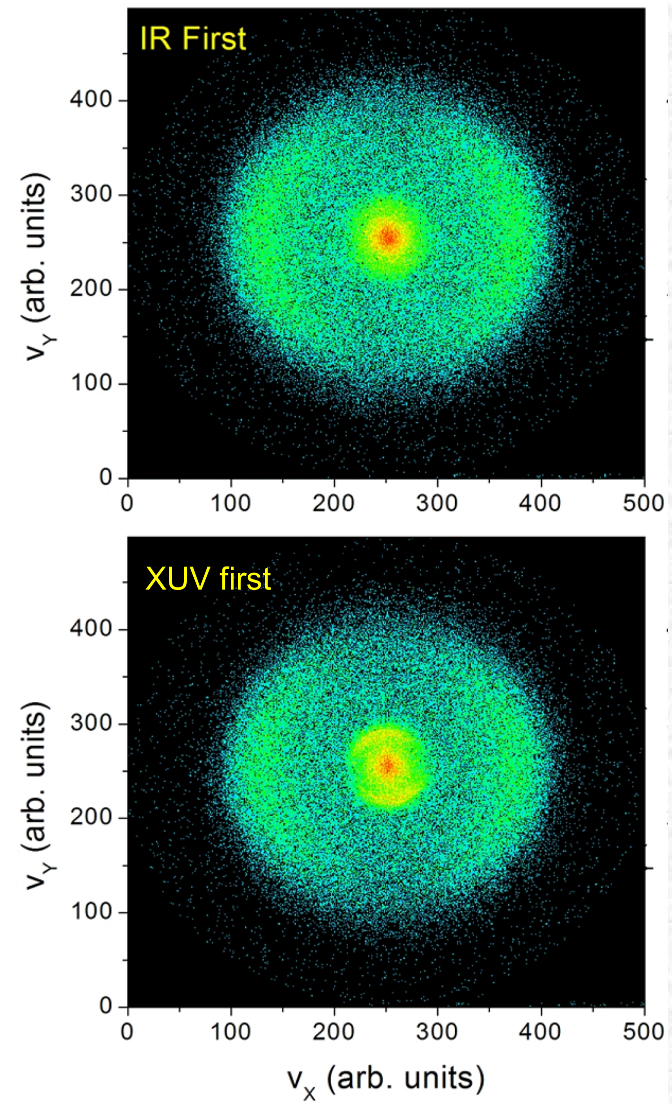
- Use IR to align the molecule
- Use FLASH FEL at 46 eV to dissociatively ionize
- Velocity and angle-resolved detection of O^+

Pump-probe experiment on CO_2 alignment (FLASH Campaign 2008, BL2)

Finding the two-color overlap

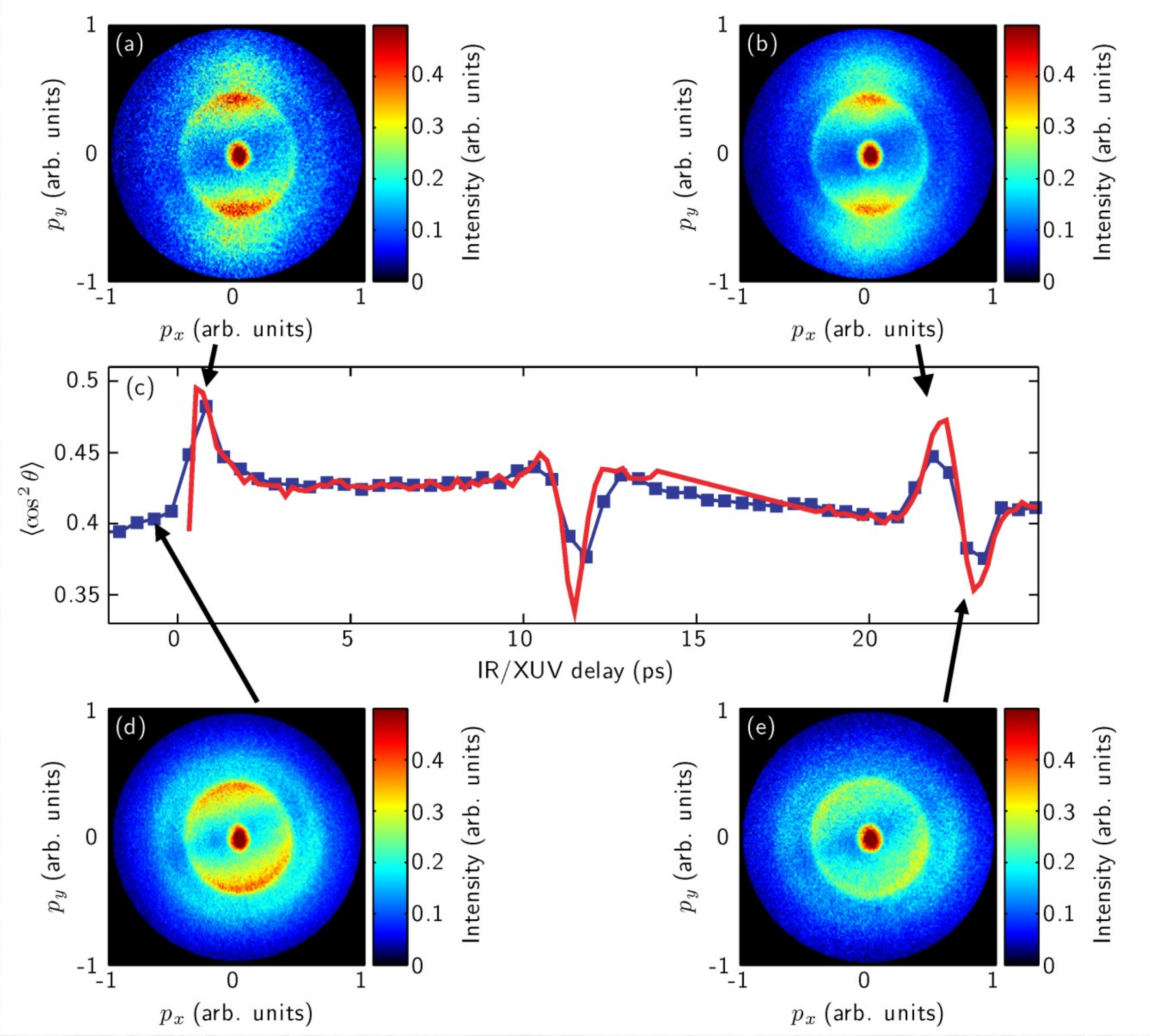
- Use bond-softening in H_2
- XUV-production of H_2^+
- IR-dissociation into $H^+ + H$
- Velocity and angle-resolved detection of H^+





P. Johnsson et al., Opt. Lett. 35, 4163 (2010)

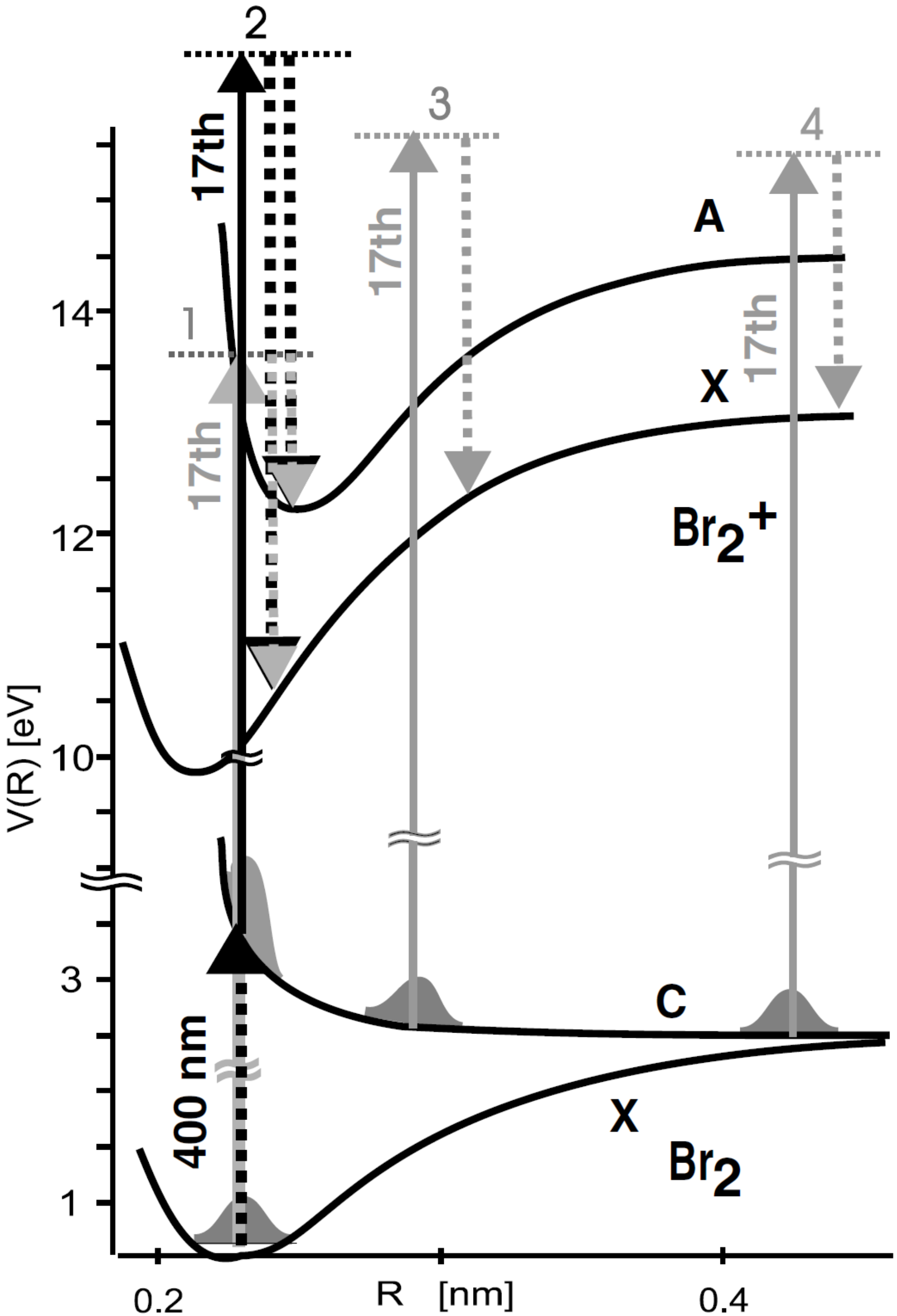
Pump-probe experiment on CO_2 alignment (FLASH Campaign 2008)



Raw data

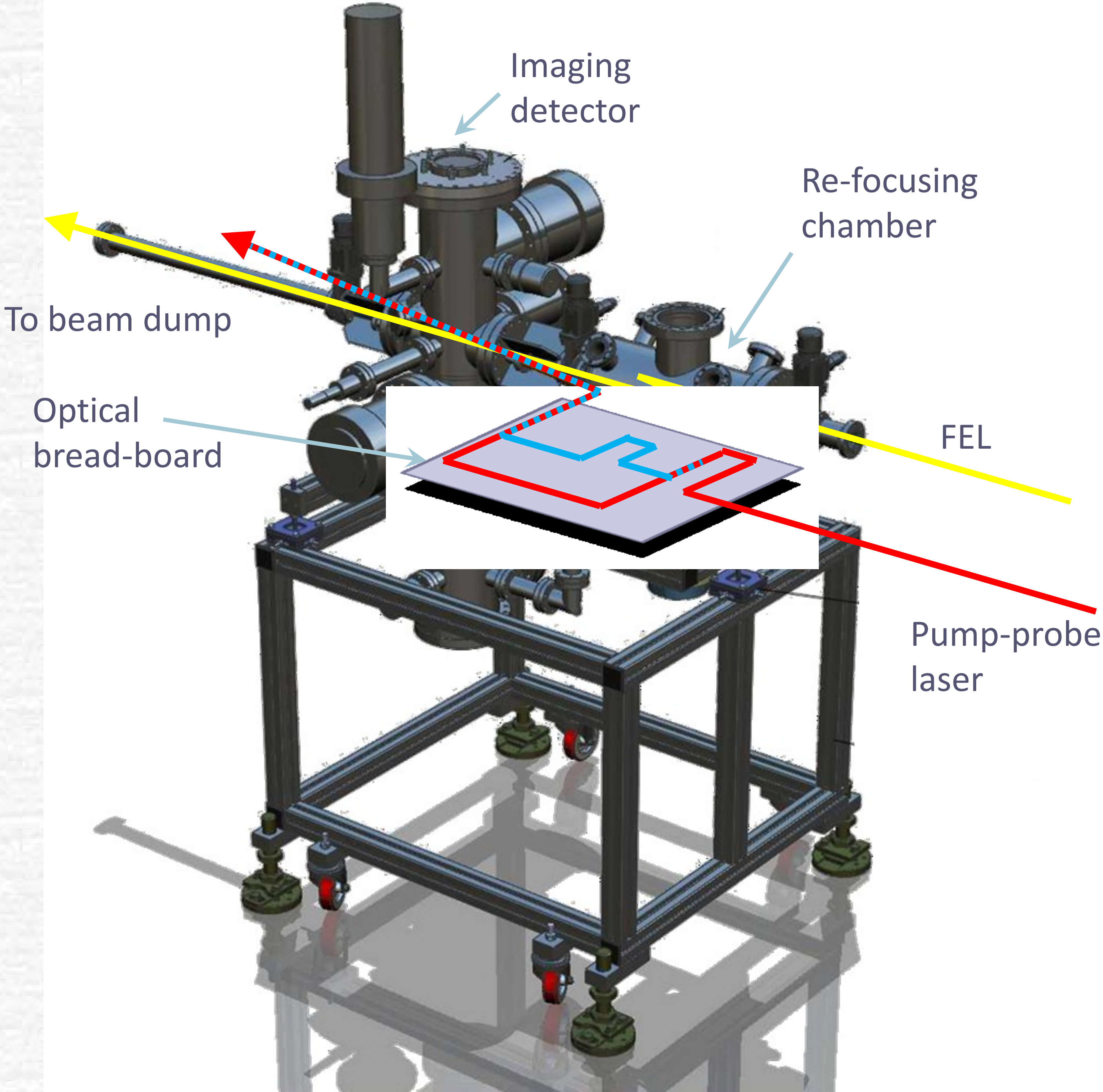
Sorted data using timing electro-optical (TEO) system (<100fs)

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

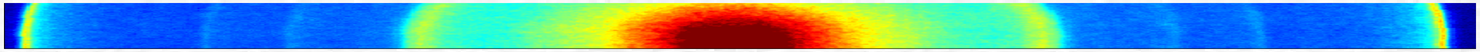


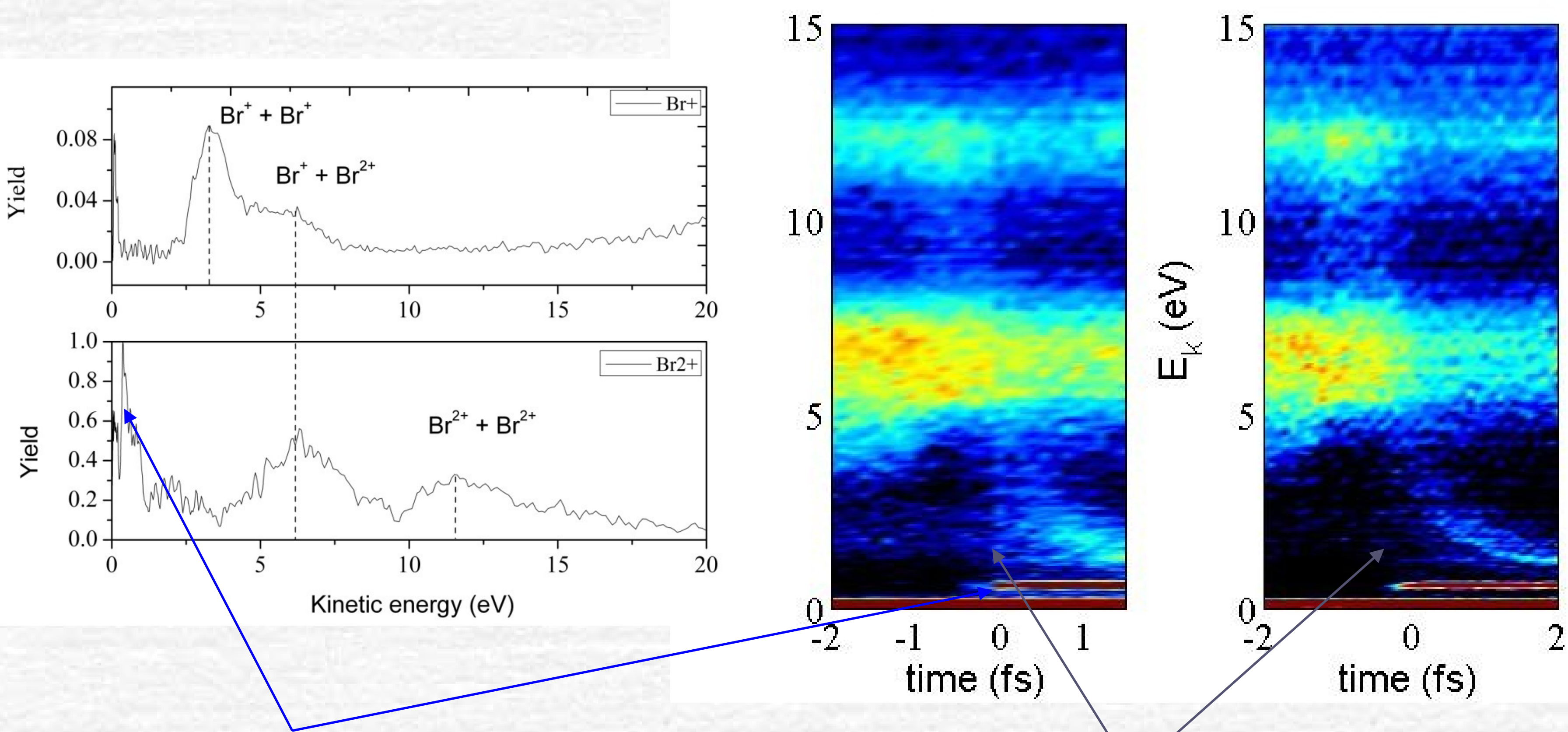
Br_2 photodissociation

- ❖ High vapor pressure
- ❖ Only moderately fast dynamics
- ❖ Easy to align
  (Rosca et al., J. Phys. B 34, 4919 (2001))
- ❖ Photodissociation at 400 nm
- ❖ Previous experiments with XUV probing
  (Leone et al., PRL 87, 193002 (2001), Wernet et al. PRL 103, 013001 (2009))


Imaging
detector
Re-focusing
chamber
To beam dump
Optical
bread-board
FEL
Pump-probe
laser

Evolution of kinetic energy spectra

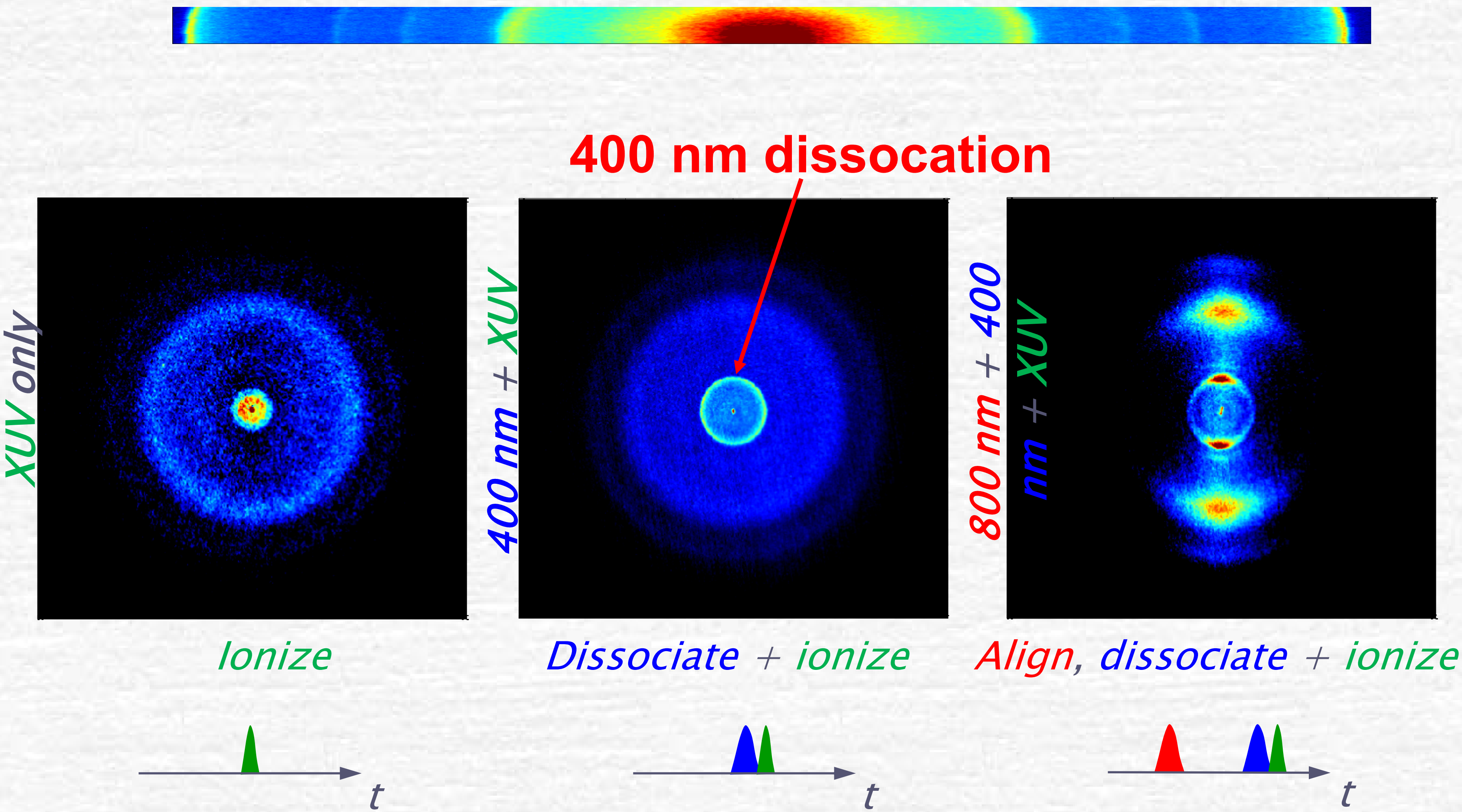




400 nm dissocation

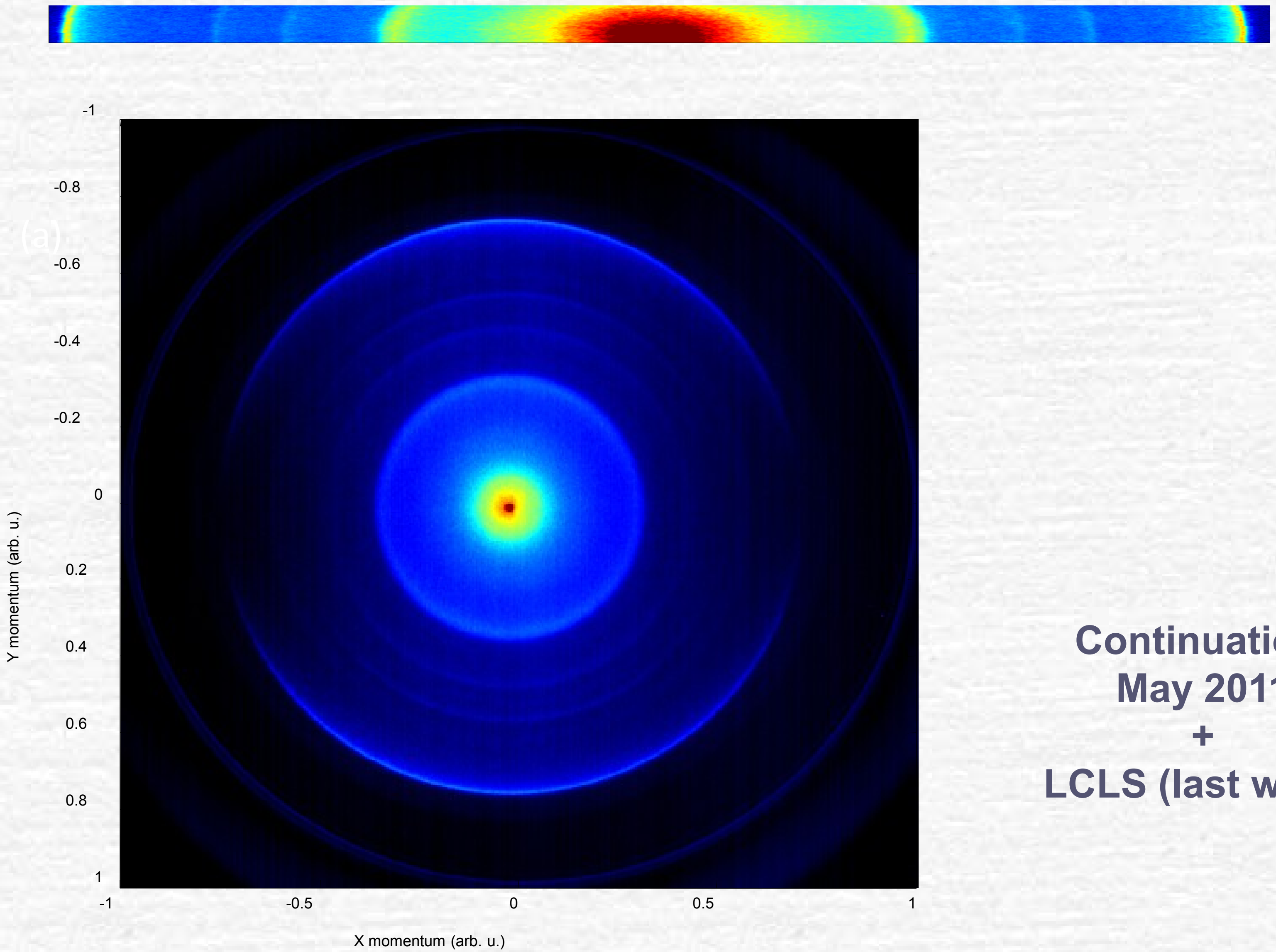
Importance of TEO sorting

Alignment and Planar Delocalization

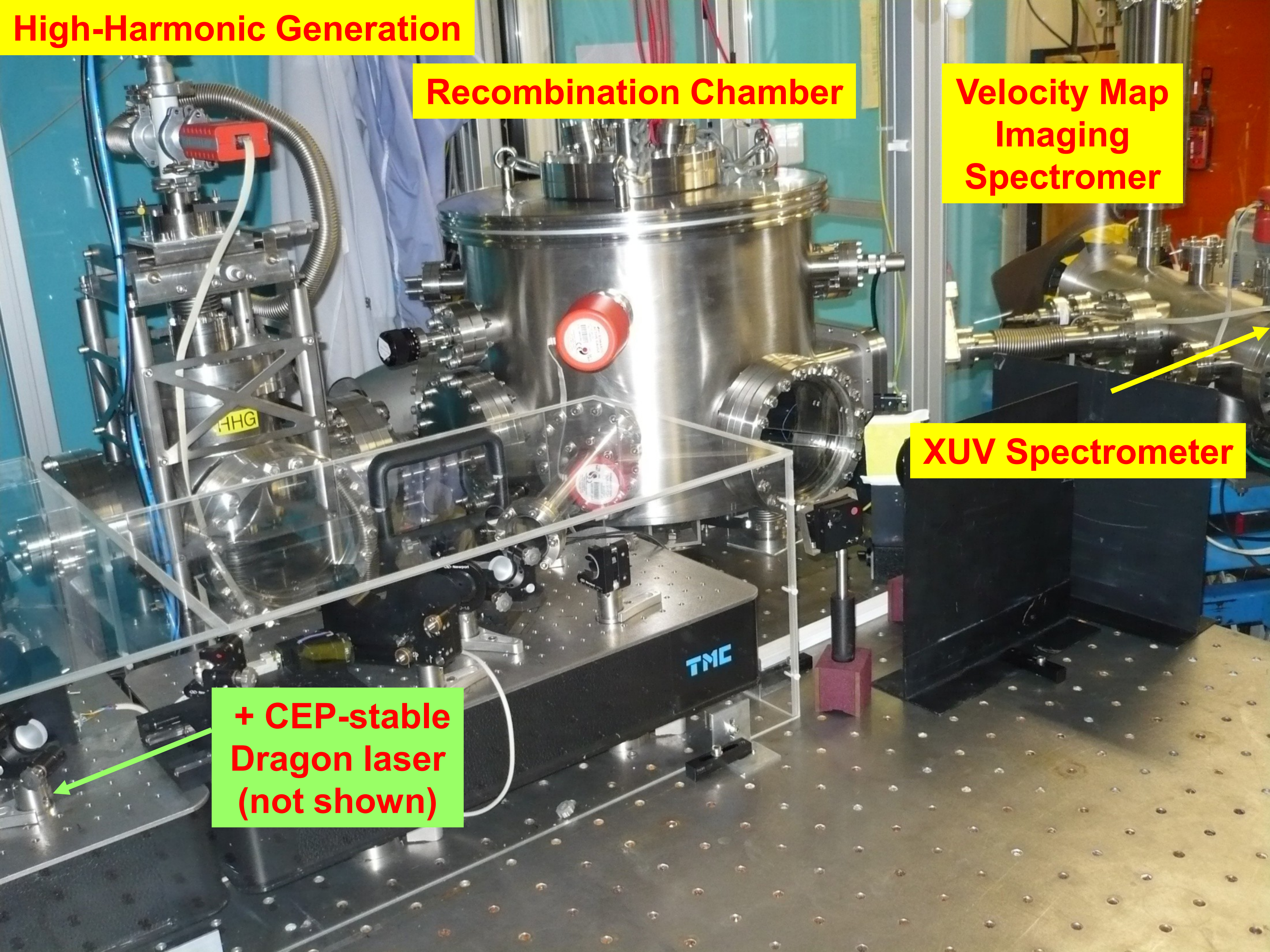


N. Berrah et al., J. Mod. Optics 57, 1015 (2010)

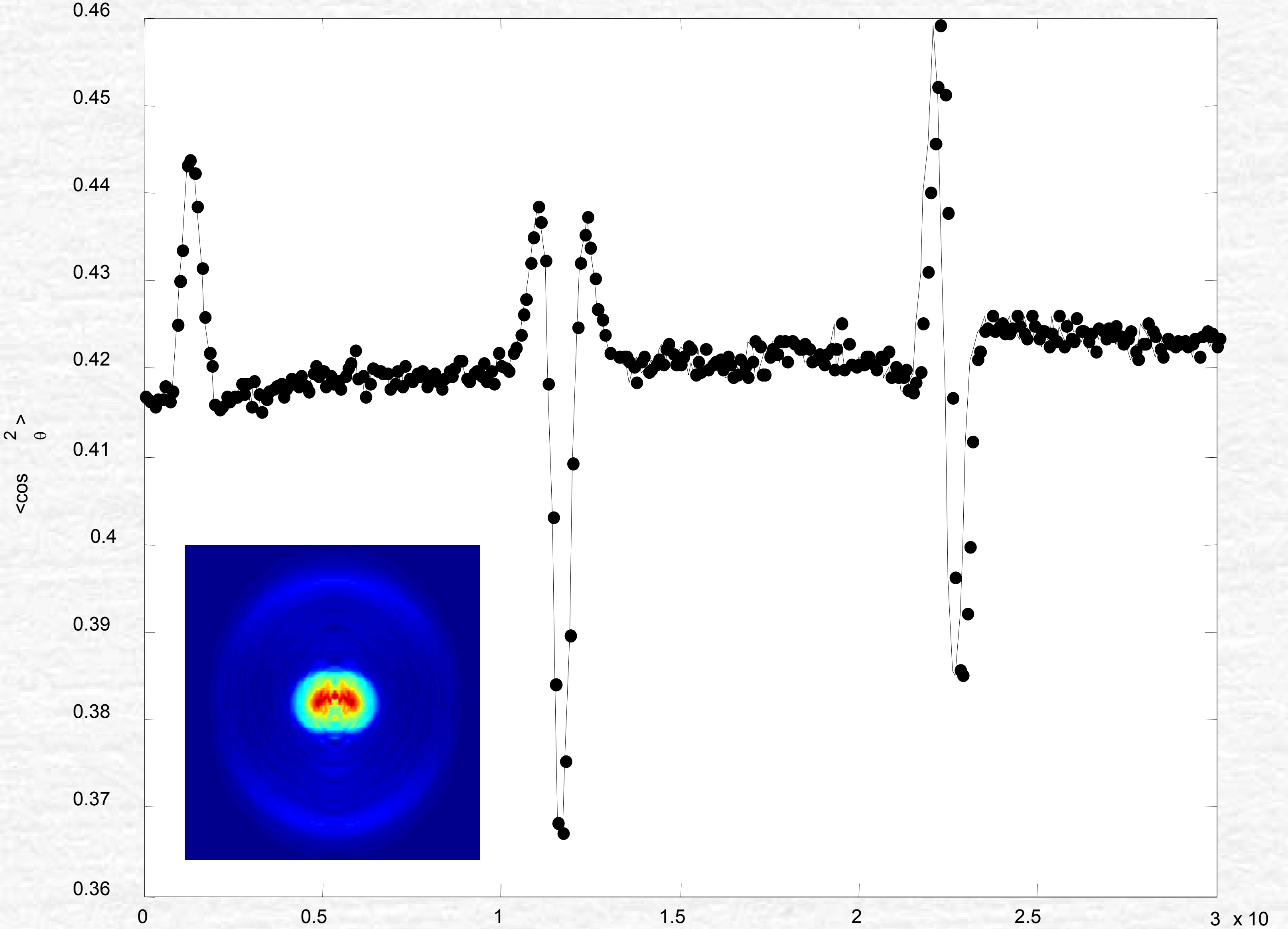
Electron spectra recorded in 800-400-XUV pump-pump-probe dissociative ionization of Br_2



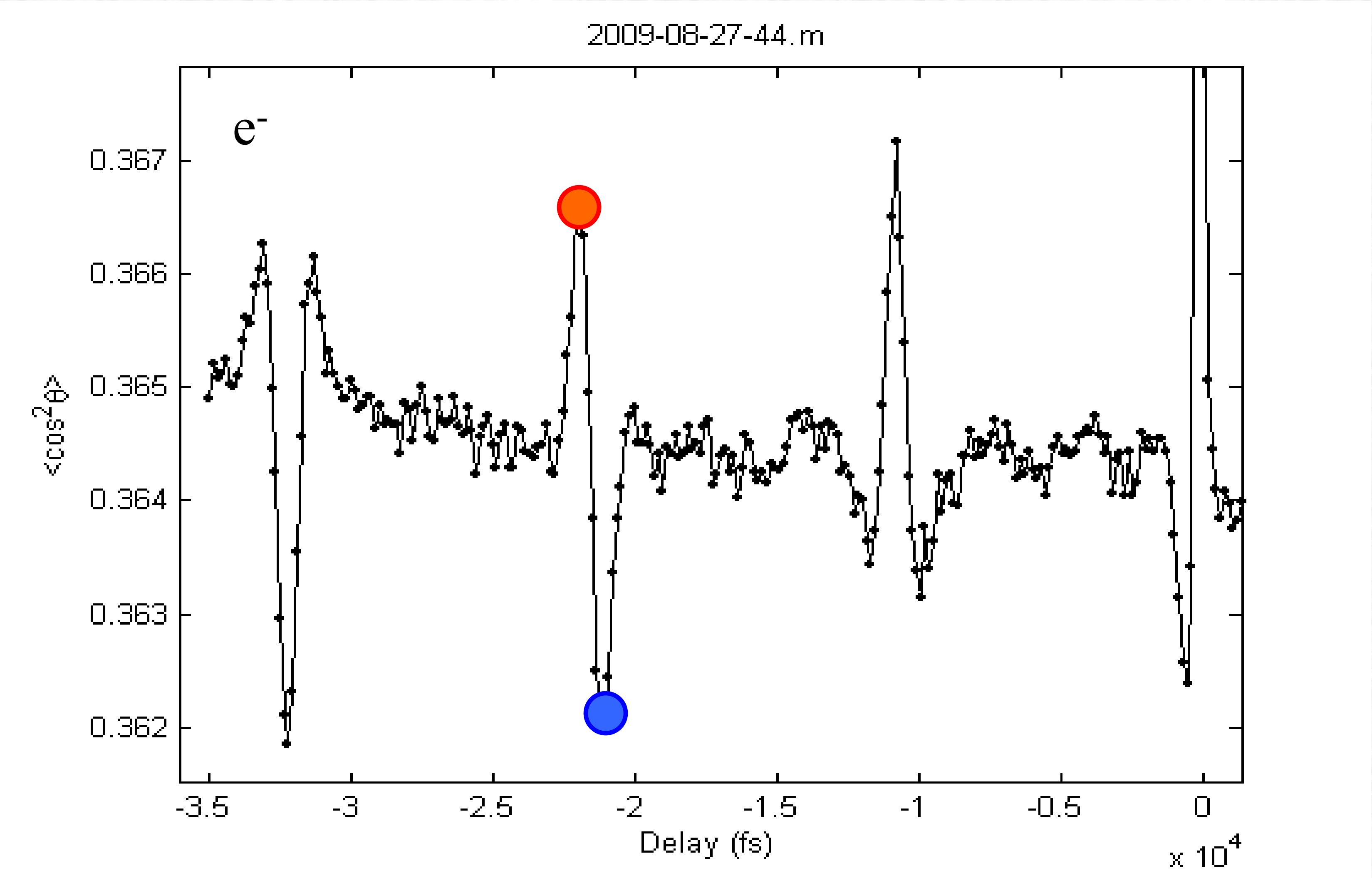
Continuation May 2011 + LCLS (last week)


High-Harmonic Generation
Recombination Chamber
Velocity Map Imaging Spectromer
XUV Spectrometer
+ CEP-stable Dragon laser (not shown)
HHG
TMC

Dynamic Molecular Alignment of CO_2

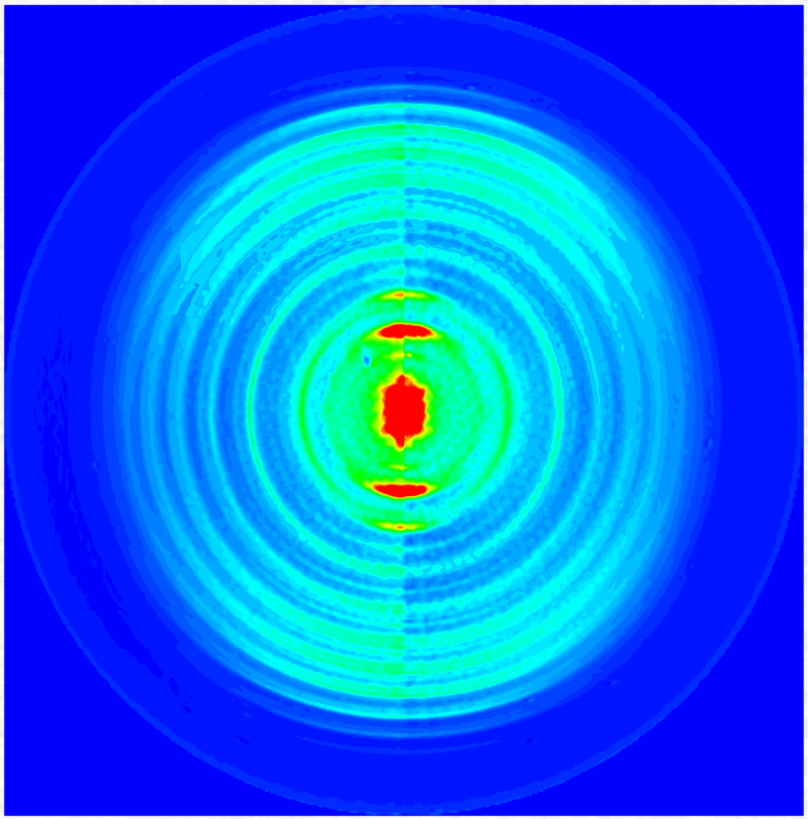


Photoelectron angular distributions from aligned CO_2 molecules

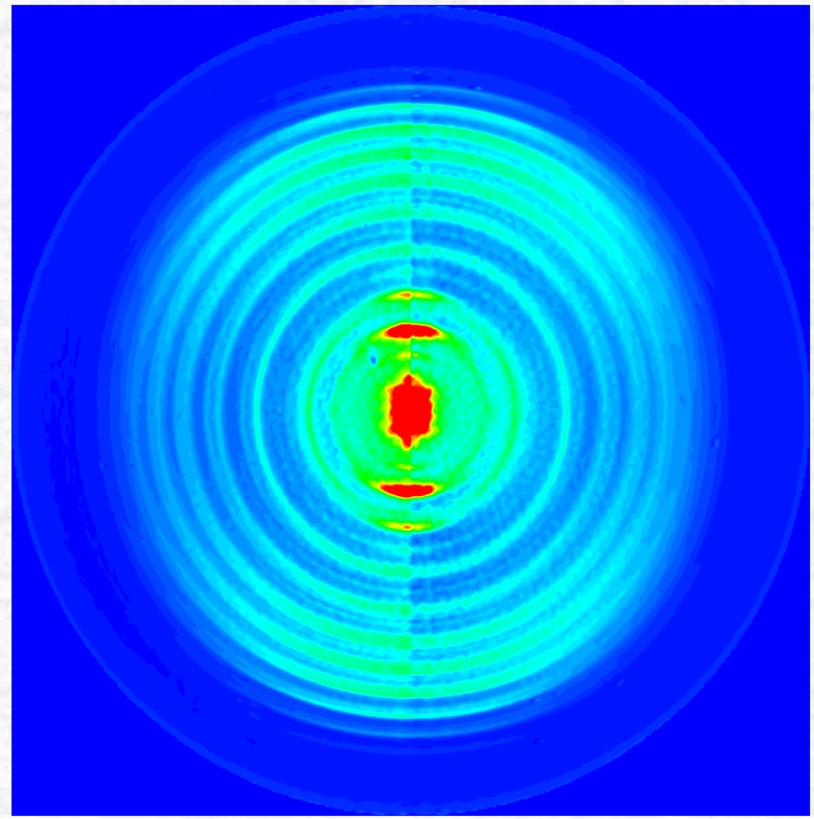



KRANT
KRANT

Photoelectron angular distributions from aligned CO_2 molecules

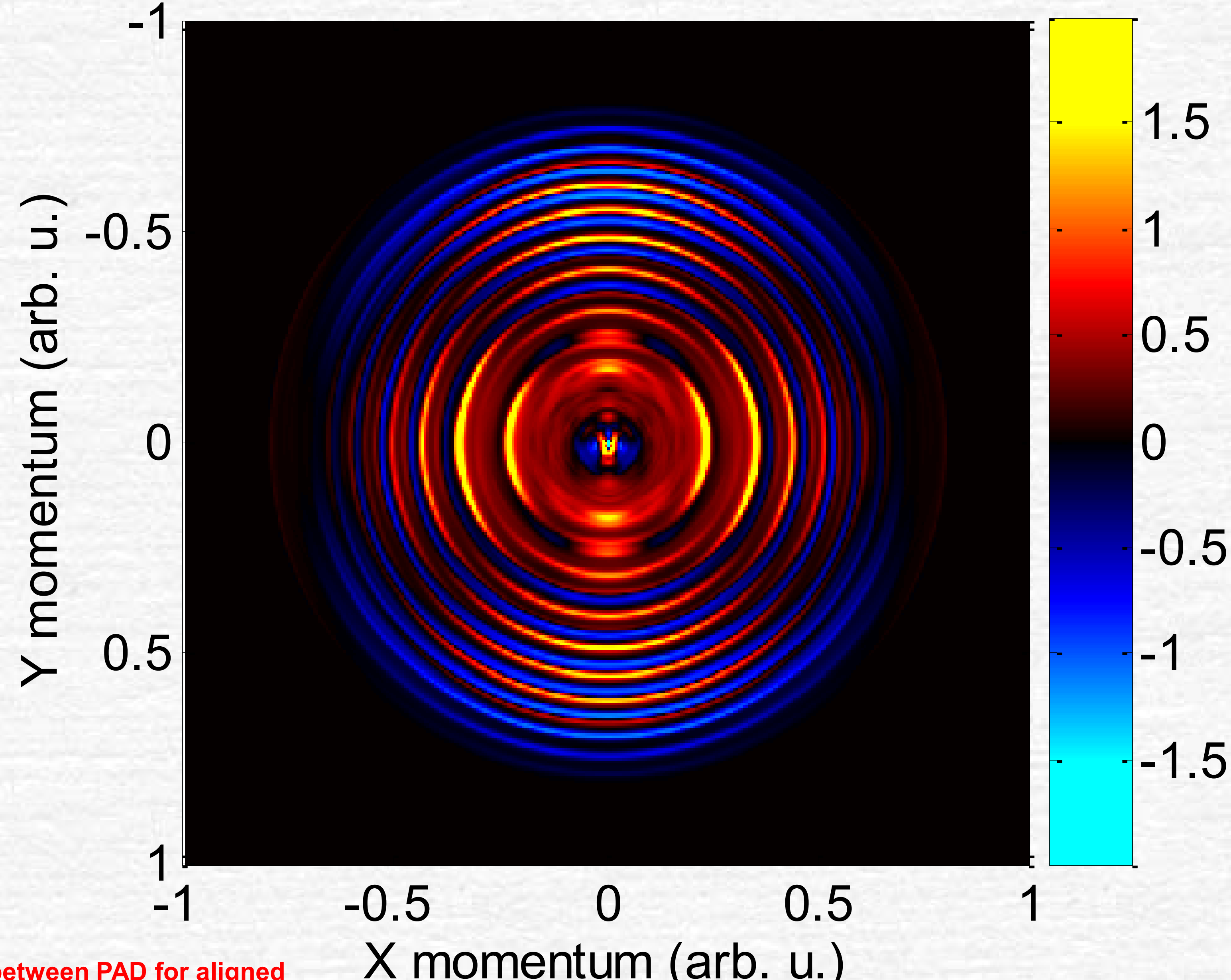


Alignment



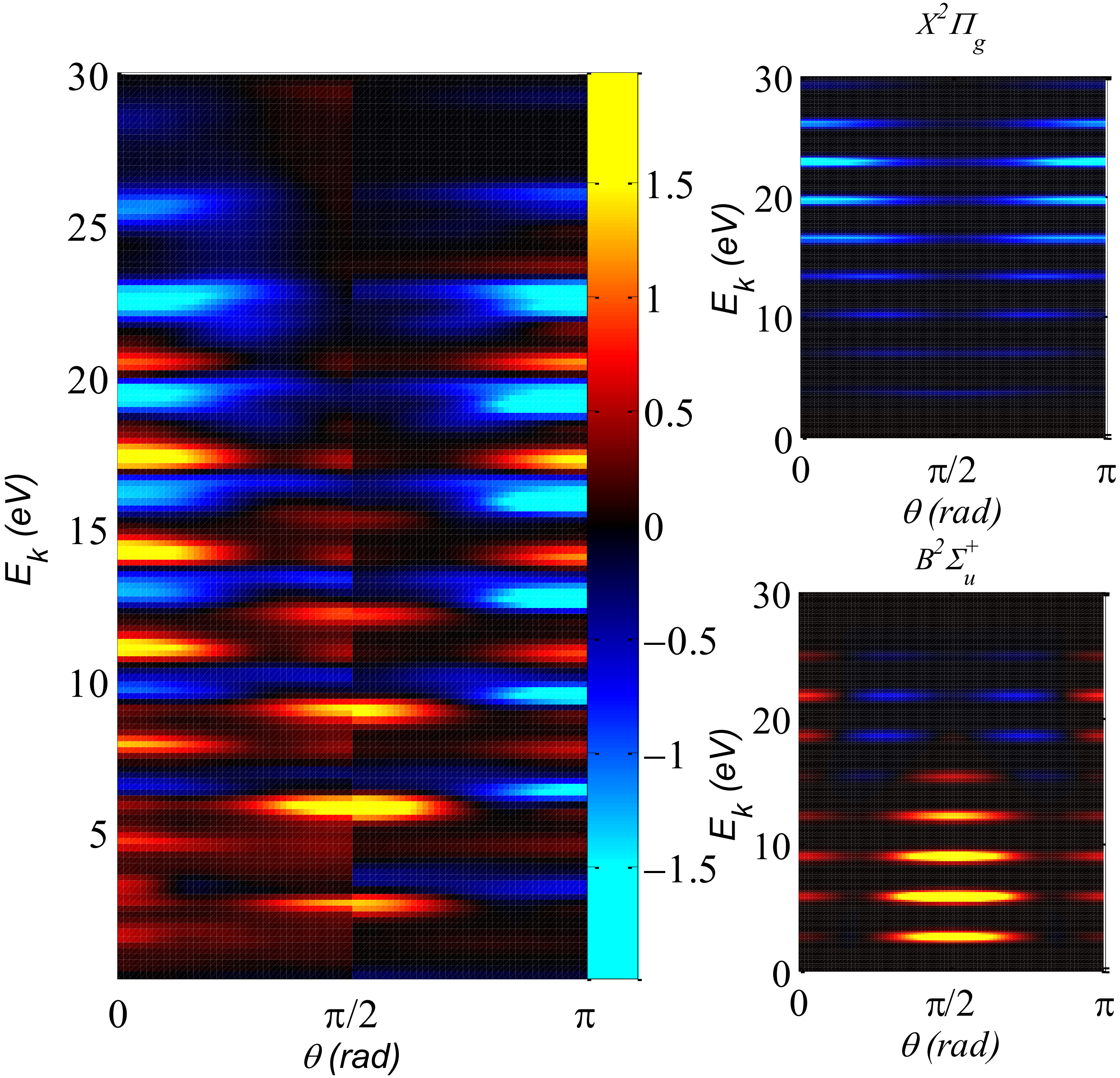
Planar Delocalization

Difference photoelectron image (inverted)



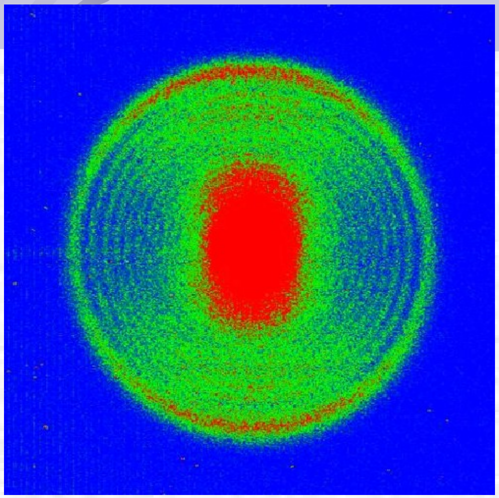
Difference between PAD for aligned and planar delocalized molecules

F. Kelkensberg et al. (in preparation)

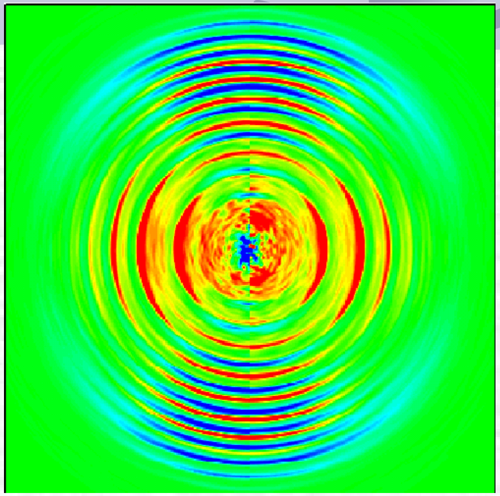


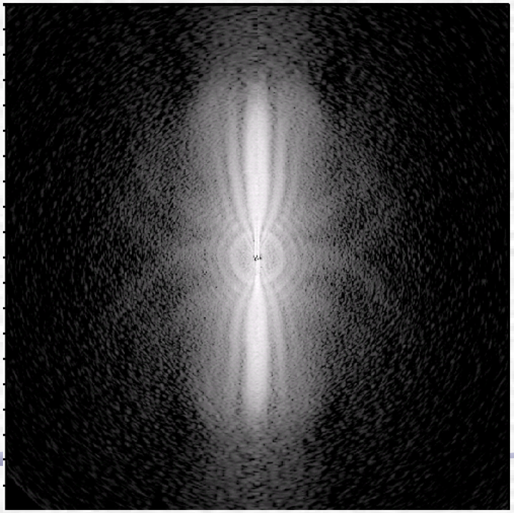
Theory by Robert Lucchese, Texas A&M

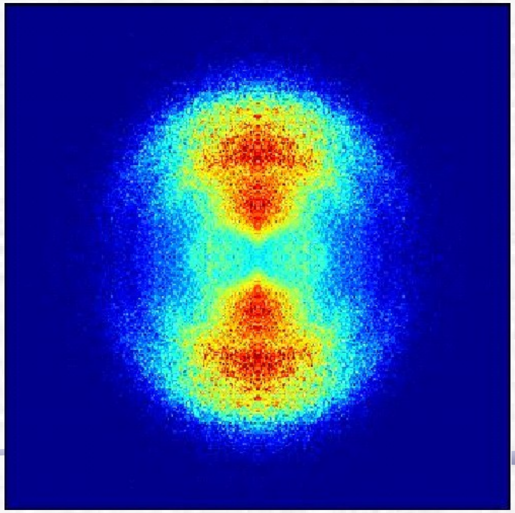
Take-home message

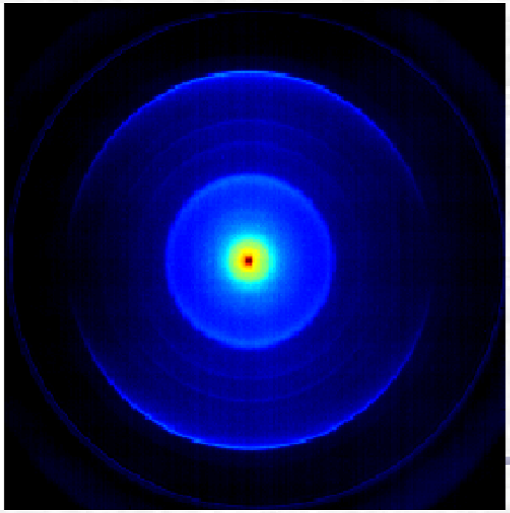


Use of the wave character of electrons allows novel experimental approaches that imprint structure and dynamics on measurable observables such as velocity map images









Novel XUV/x-ray sources (HHG and FELs) allow to push atomic & molecular science beyond the present state-of-the-art

❖Temporal properties → Electron dynamics

Example:

Attosecond time-resolved pump-probe spectroscopy in H_2

Wavepacket interferometry experiments give access to attosecond electron dynamics

❖Wavelength properties → Nuclear dynamics

Example:

Molecular frame photo-emission gives access to (time-dependent) orbital and structural information – pump-probe spectroscopy at FLASH and using an HHG source