# *Connecting Attosecond Science and XUV FEL Research*

Marc Vrakking

Attosecond Workshop
Imperial College, May 13th 2008

# Overview

- Present status of attosecond science

  *- recent example: electron localization on attosecond timescales in $H_2$ and $D_2$*
  *- assessment of strengths and weaknesses*

- Interplay between attosecond science and XUV FEL Research

- Results from recent campaign at FLASH FEL in Hamburg (April 2008)

# Acknowledgements

**FOM-AMOLF,NL**
**Per Johnsson** (formerly Lund)
**Freek Kelkensberg**
**Wing Kiu Siu**
**Ymkje Huismans**
**Arnaud Rouzee**
Omair Ghafur
**Tatiana Martchenko (now LOA)**
**Franck Lépine (now Lyon)**
Christian Siedschlag

**+technical support**
Rob Kemper
Hinco Schoenmaker
Ad de Snaijer

**Funding**:
NWO-VICI, NWO-ECHO, FOM-PR, EU

**Lund, SE**
Johan Mauritsson, Anne L'Huillier

**Louisiana State, USA**
Ken Schafer

**Garching, D**
**Matthias Kling**, Ferenc Krausz

**NRC Ottawa**
**Misha Ivanov**

**Milano, It**
**Giuseppe Sansone**, Mauro Nisoli

**FLASH, Hamburg**
**Stefan Duesterer**, Artem Azima, Franz Tavella, Nikola Stojanovic

# Sofar: Two ways that we can use attosecond pulses in experiments

1. XUV ionization followed by **acceleration** of the ionized electron in a strong IR field (**continuum**)

   Used in attosecond pulse characterization

   Used in attosecond interferometry experiments

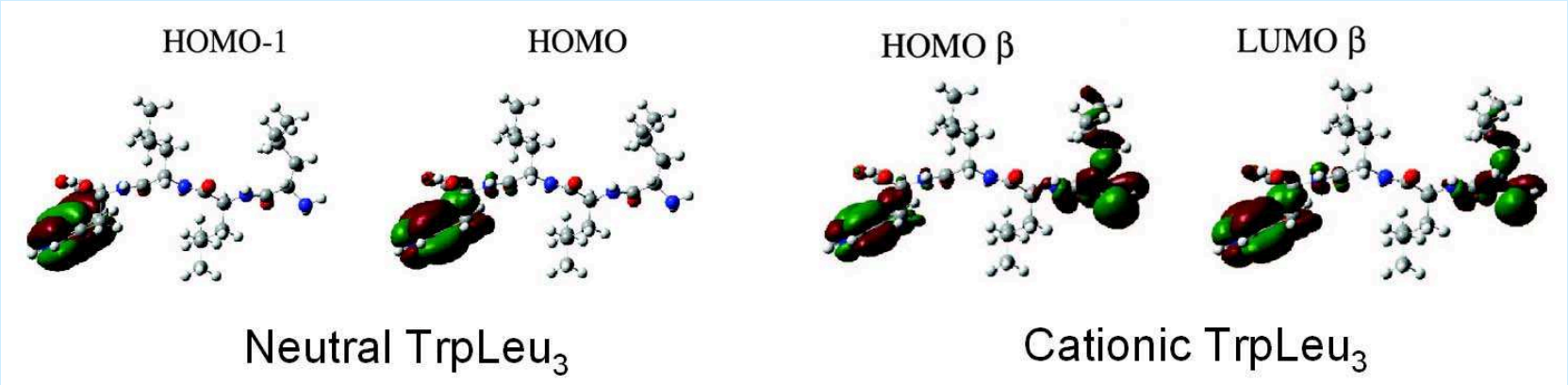
2. XUV excitation of **bound** states, followed by **ionization** in a strong IR field

   Used to study bound state dynamics and/or time-dependent ionization dynamics

# What do we want to do in attosecond science?

## Measure ultrafast electron dynamics and coupling of electronic and nuclear degrees of freedom

Prediction by F. Remacle and R. Levine (PNAS 103, 6793 (2005)): Ultrafast electron transfer is possible in large bio-molecules.




Also: electron dynamics in strong laser fields

- dynamic alignment
- Coulomb explosion
- control of electron localization

# Today: A third way that attosecond pulses can be exploited

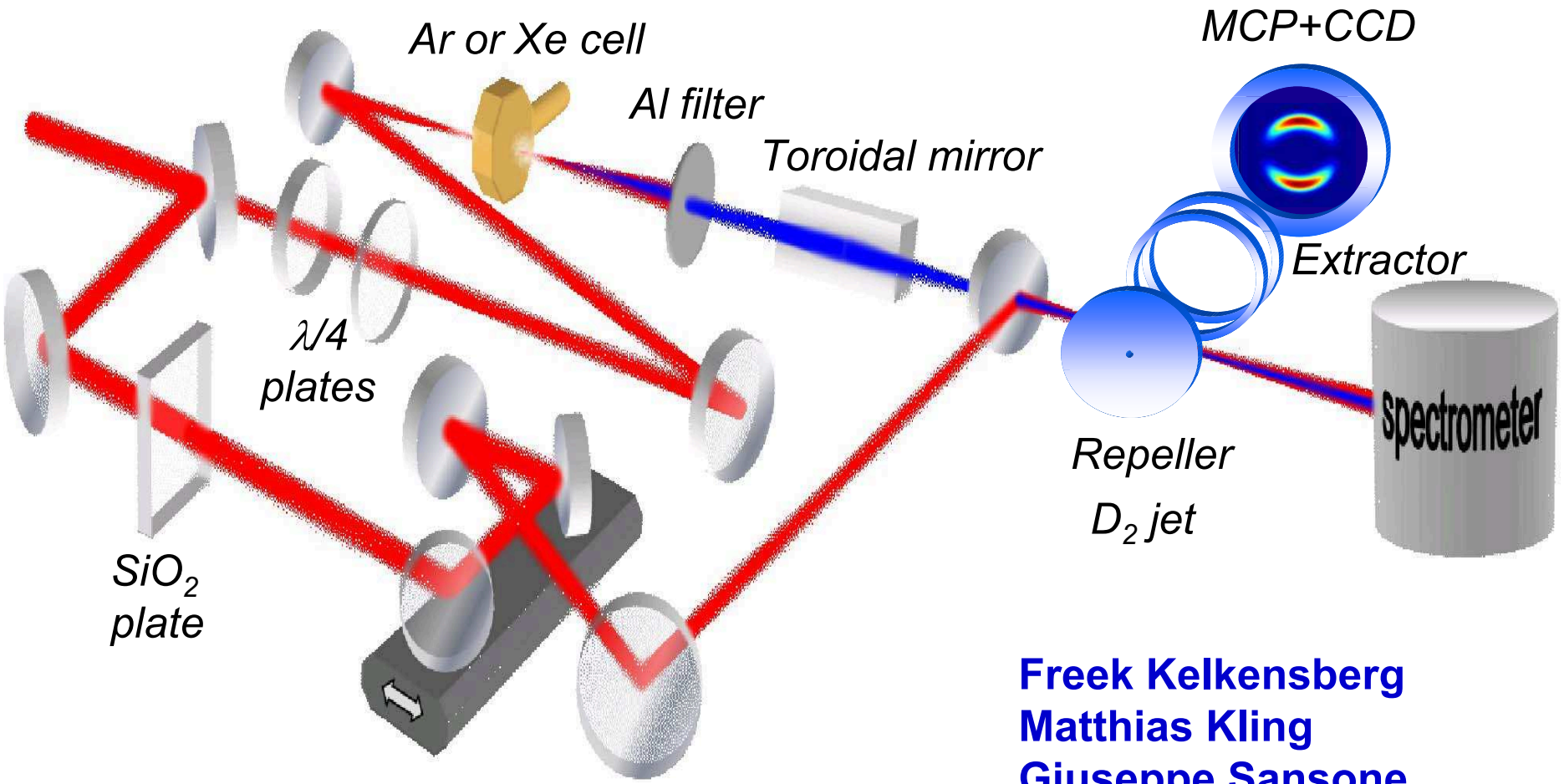
➔ **Rely on electron localization to probe electron dynamics**

Can be used in attosecond control of electron dynamics (Science 312, 246 (2006))

Can be used to observe electron dynamics on attosecond timescales

→ **Case study: XUV-IR pump-probe experiments on electron localization in $H_2$ and $D_2$**

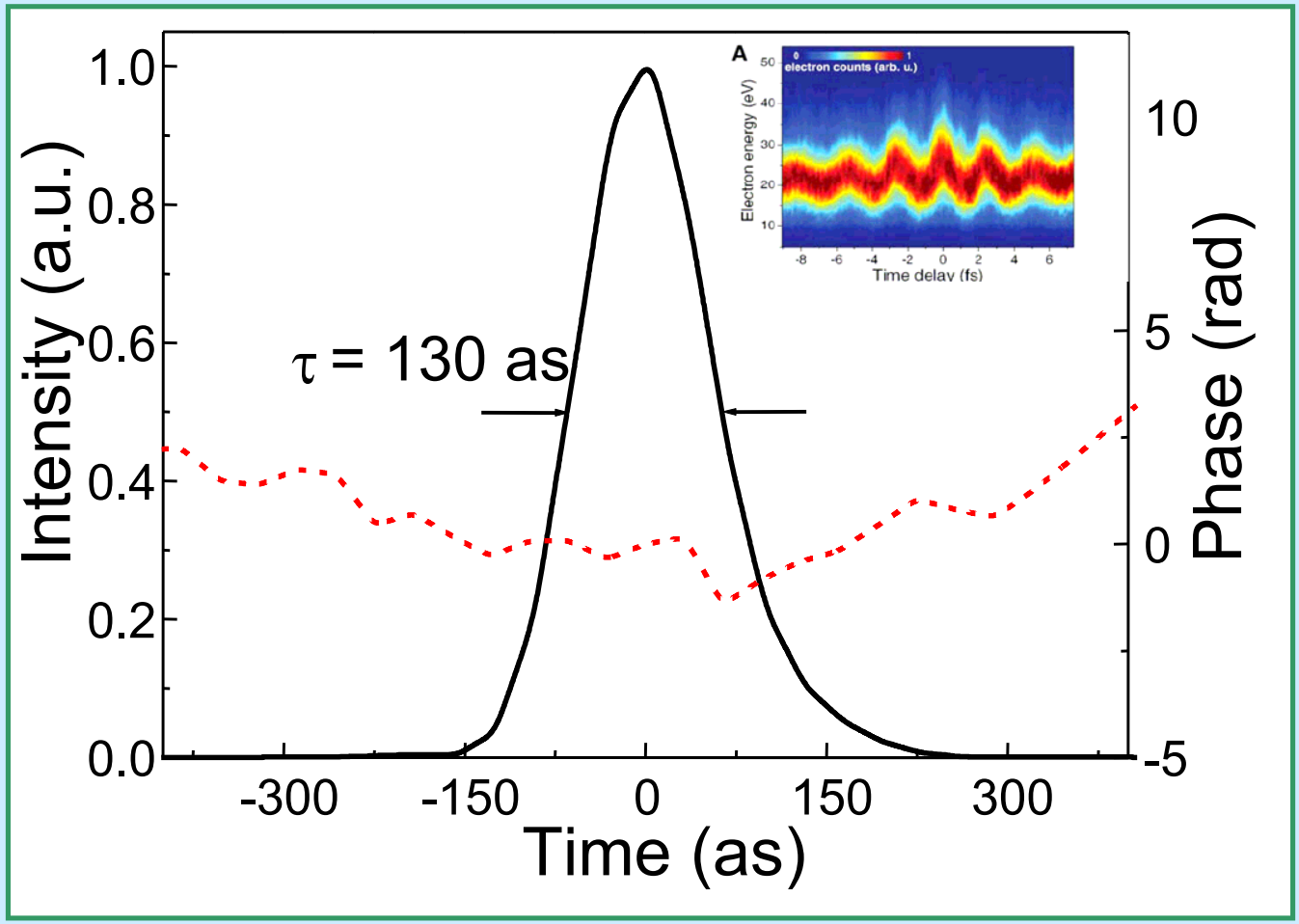
# Joint campaign
# AMOLF/Lund/Garching/Lyon/Milano




**Freek Kelkensberg**
**Matthias Kling**
**Giuseppe Sansone**
**Misha Ivanov**

# Enabling Technologies :

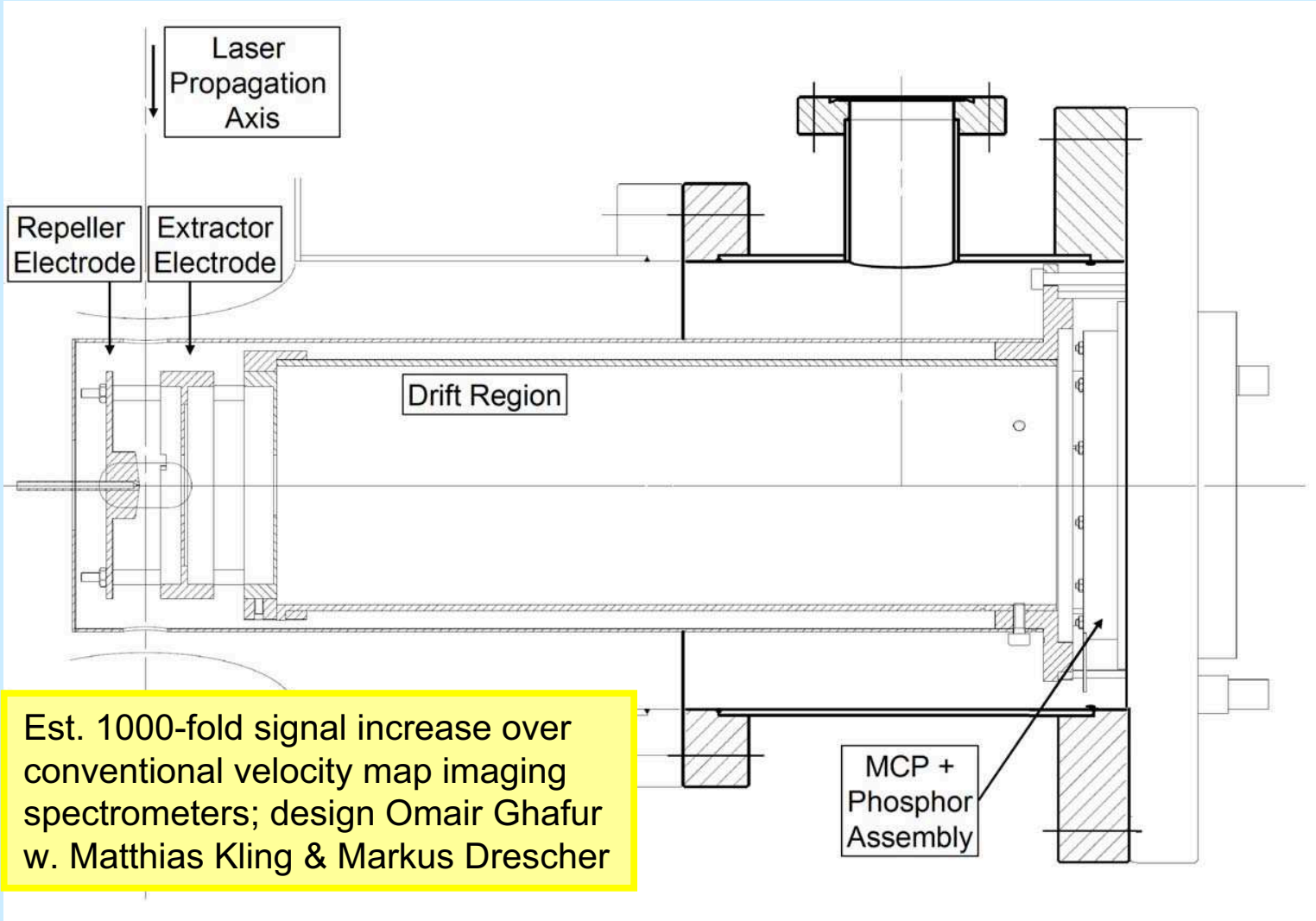
## A source of isolated attosecond laser pulses (Milano)




*G. Sansone et al., Science* ***314****, 443 (2006)*

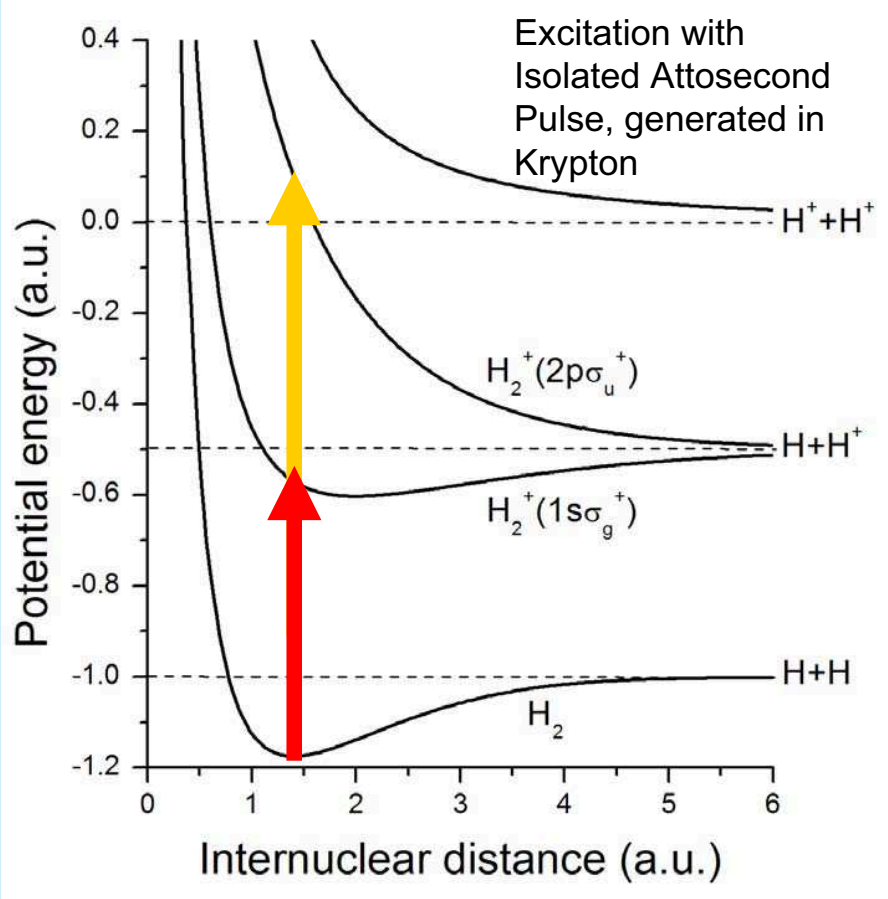
# Enabling Technologies :

## An imaging spectrometer with integrated gas injection (AMOLF)

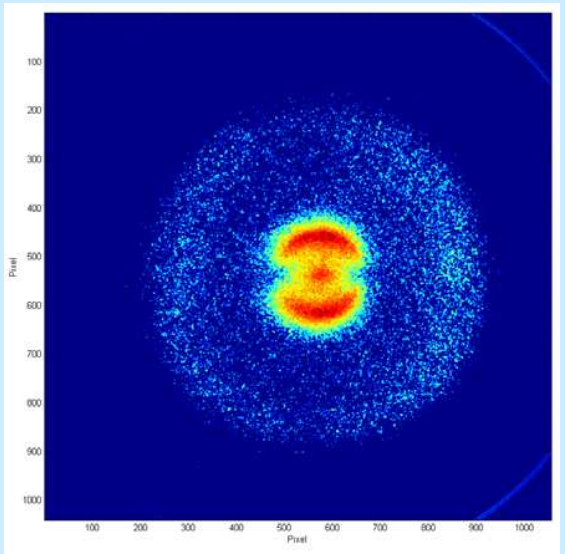



Est. 1000-fold signal increase over conventional velocity map imaging spectrometers; design Omair Ghafur w. Matthias Kling & Markus Drescher

# XUV-IR Pump-probe experiments on $H_2$ and $D_2$



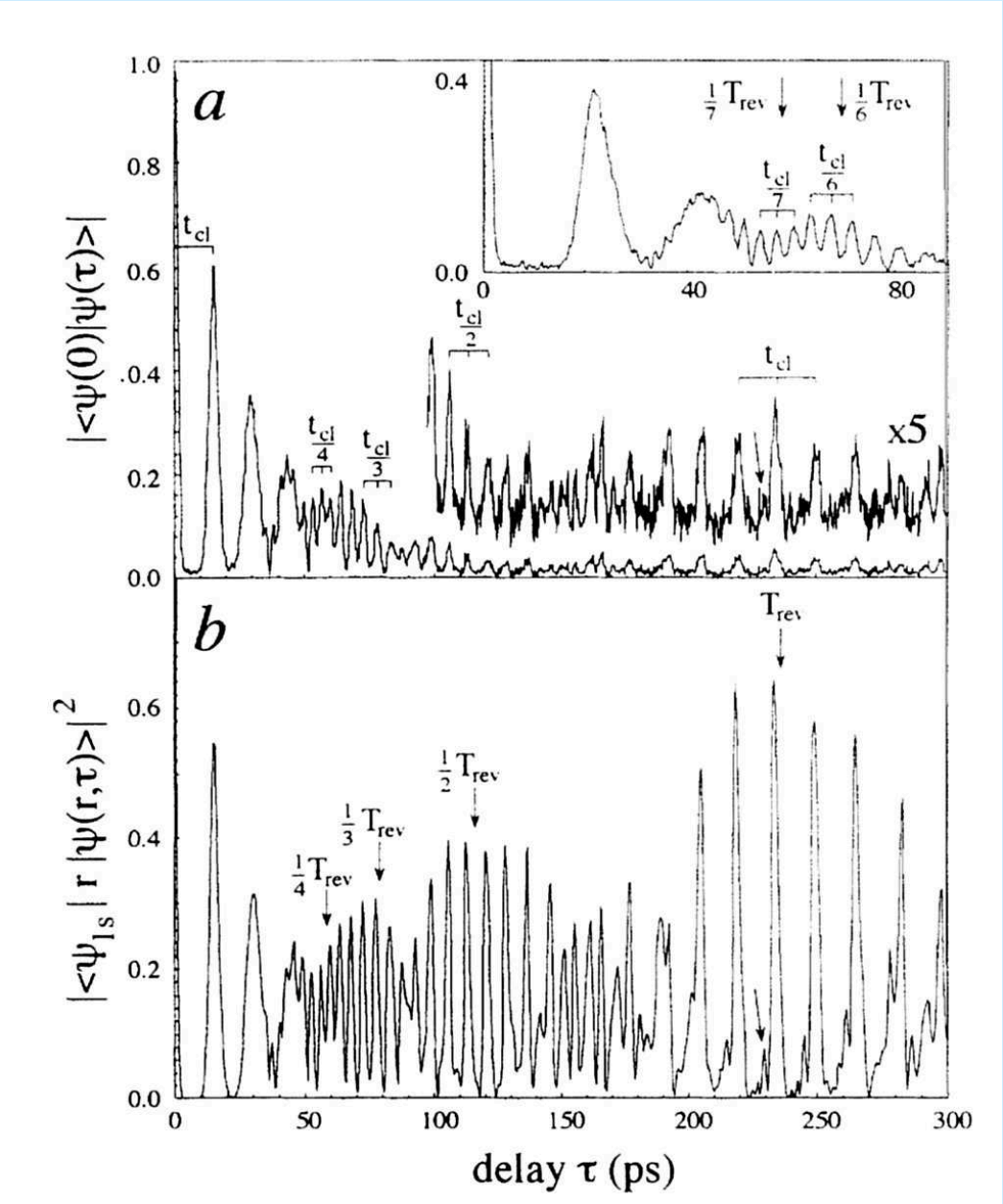

Use isolated attosecond pulse generated in Krypton to launch a wavepacket on the $2p\sigma_u^+$ state or the $1s\sigma_g^+$ state and investigate the subsequent IR interaction



# Status of Attosecond Science

- We're beginning to be able to do various types of pump-probe experiments that reveal electron dynamics and/or correlations between electronic and nuclear motion

- Where are the problems, what can't we do yet?




We can't do even the simplest attosecond XUV pump – attosecond XUV probe experiment!!

N.B. Studying electron dynamics with femtosecond lasers requires slowing down the dynamics by working with Rydberg atoms or molecules

Wals. et.al. Phys. Rev. Lett. 72, 3783 (1994).

# Two Families of Attosecond Laser Experiments

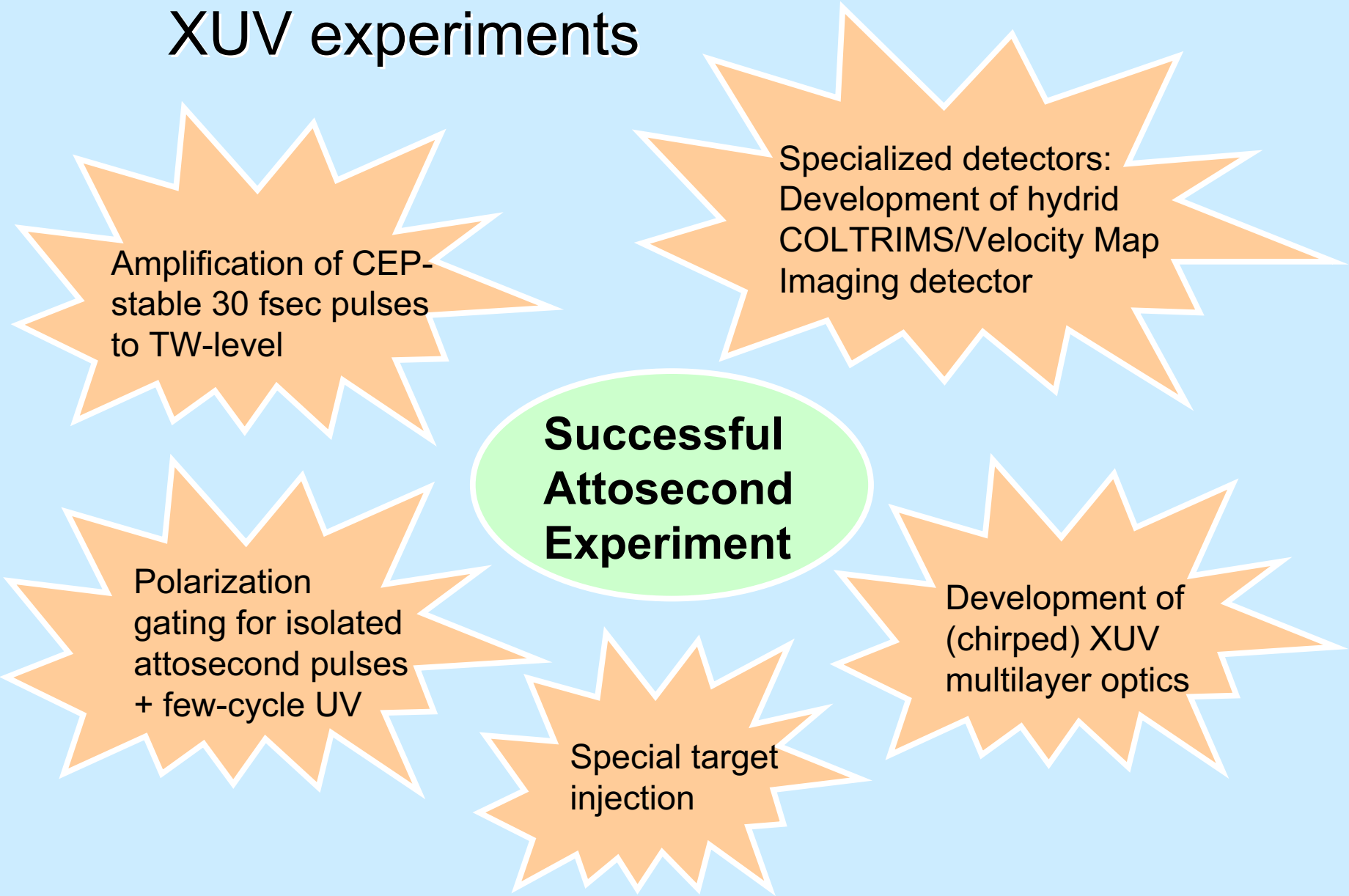
1. High harmonic generation using a many-cycle laser pulse → *train of attosecond laser pulses*

   State of the art is ~ 10 μJoule/harmonic ($3x10^{12}$ photons/harmonic at 30 eV) → non-linear ionization "heroic"

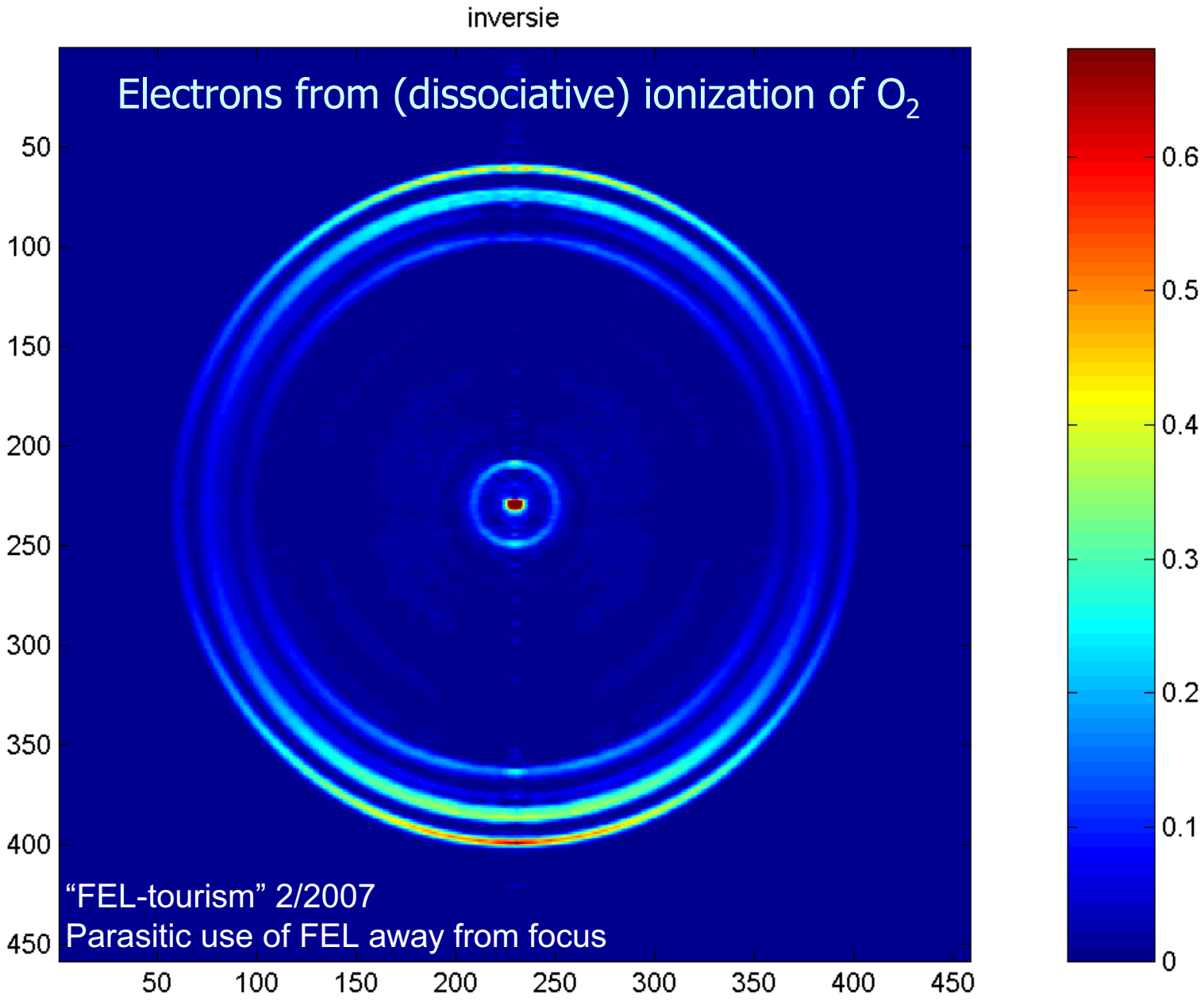
2. High harmonic generation using a few-cycle (CEP-stabilized) laser pulse or using a pulse with a time-varying polarization → *isolated attosecond pulses*

   Typically ~ $10^6$ photons/pulse or less

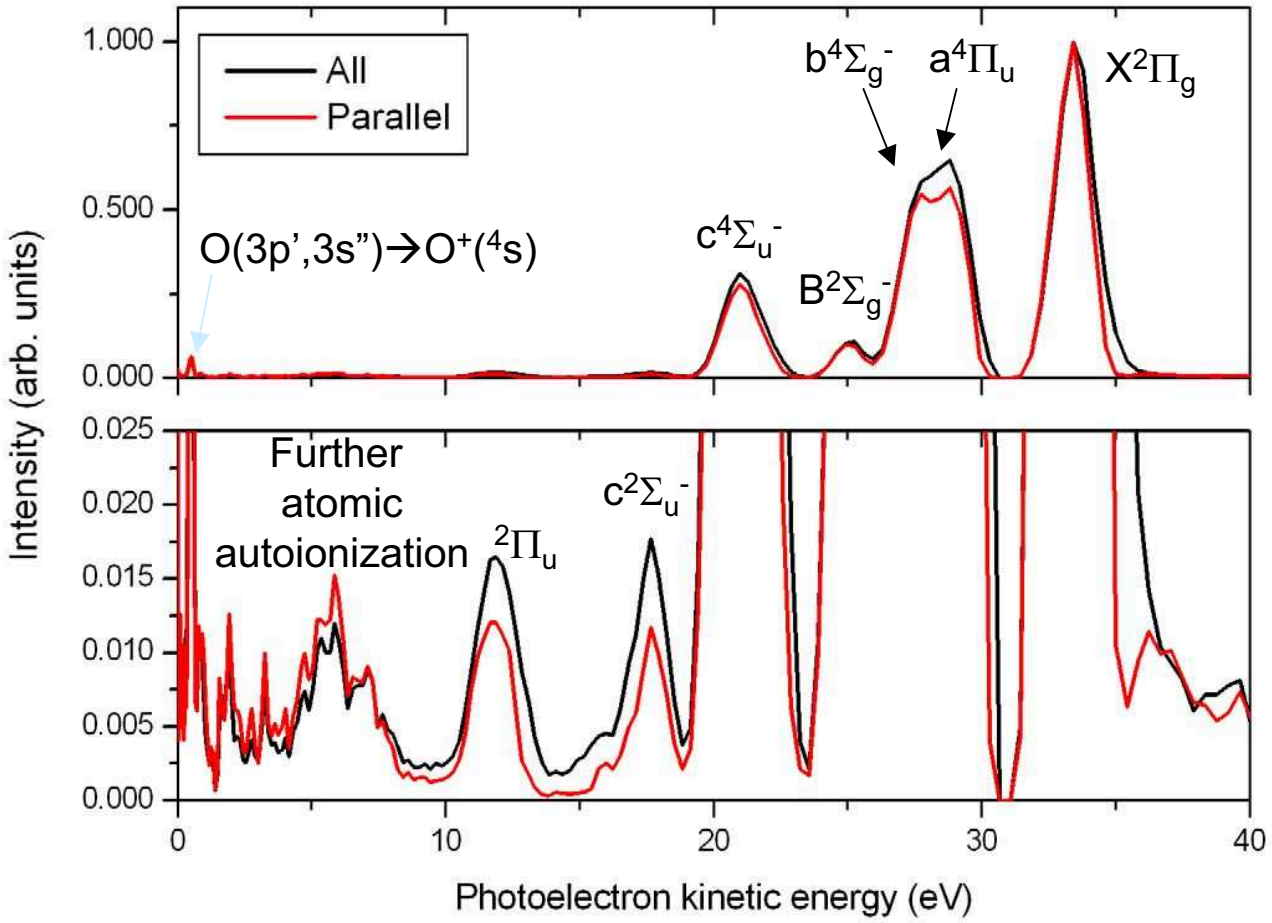
# 1) Our choices for going towards XUV-XUV experiments



# 2) Exploiting the complementarity between harmonics-based and FEL-based experiments


inversie
Electrons from (dissociative) ionization of $O_2$
50
100
150
200
250
300
350
400
450
50
100
150
200
250
300
350
400
450
0.6
0.5
0.4
0.3
0.2
0.1
0
"FEL-tourism" 2/2007
Parasitic use of FEL away from focus

# Exploring the utility of Velocity Map Imaging at the FEL : $O_2$

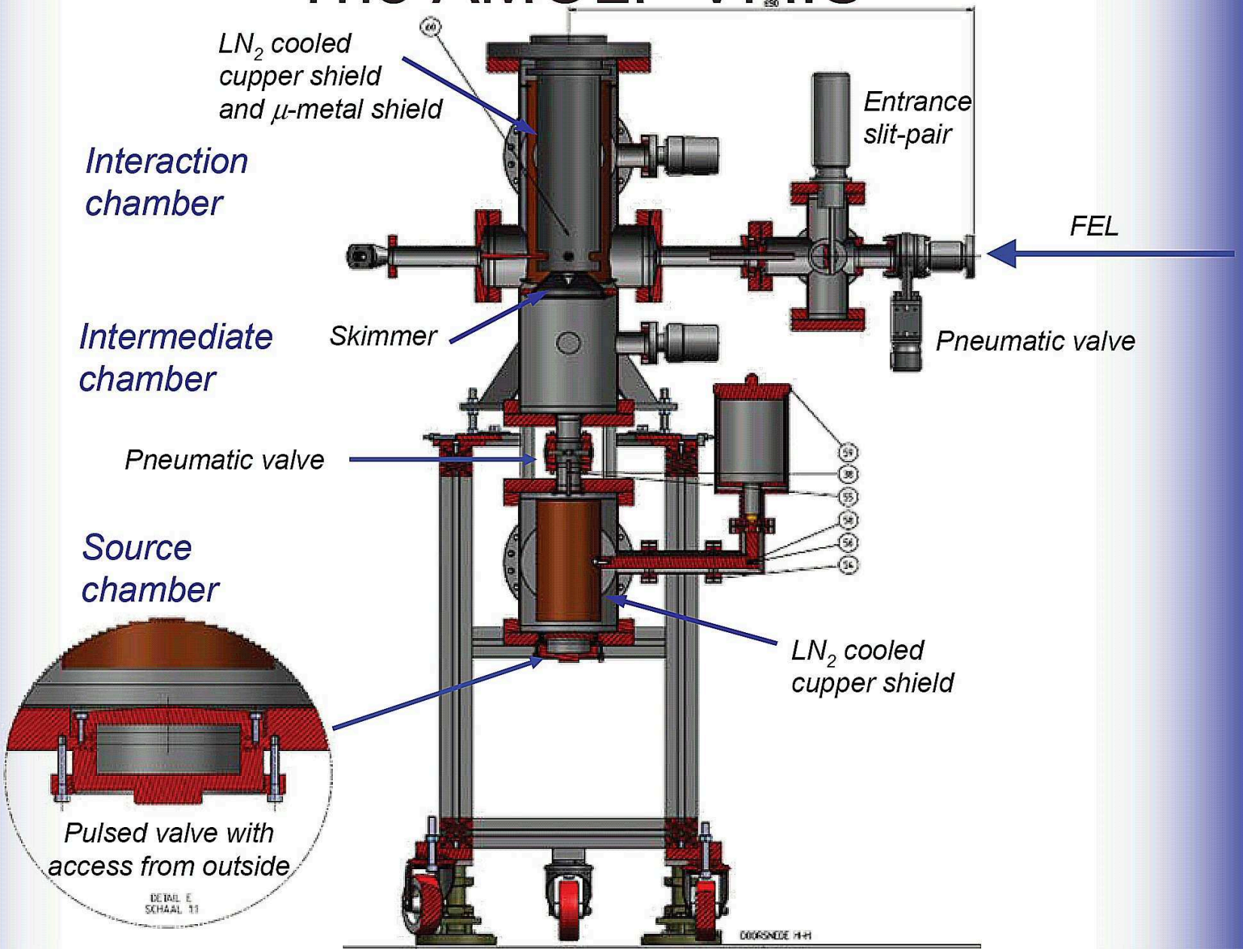


# Recent campaign: 4/2008

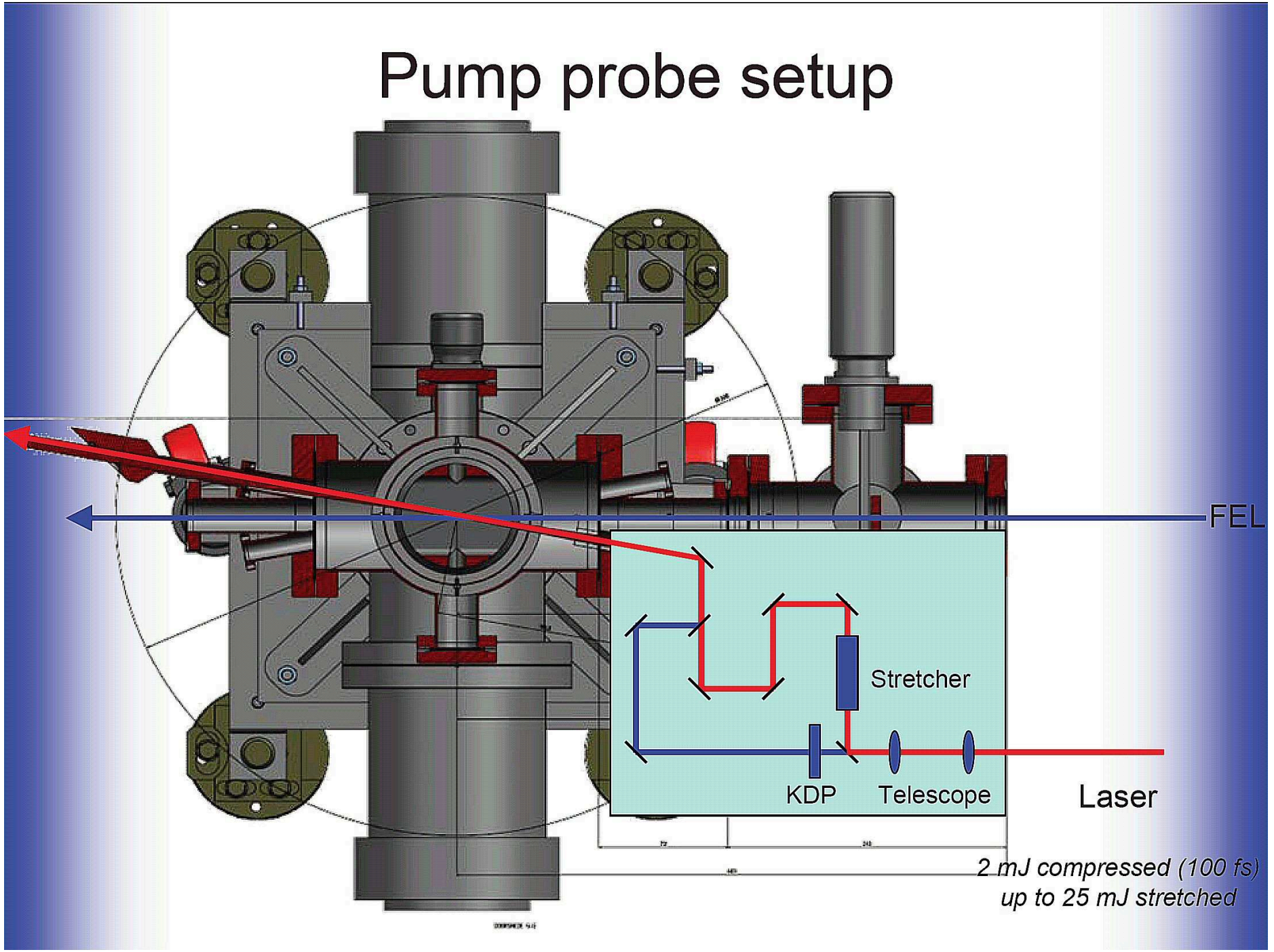
- Attempt to perform molecular IR-XUV pump-probe spectroscopy at FLASH

**Per Johnsson**
**Arnaud Rouzee**
**Wing Kiu Siu**
**Ymkje Huismans**
**Franck Lepine**
**Tatiana Martchenko**
**Stefan Duesterer c.s.**

# The AMOLF VMIS

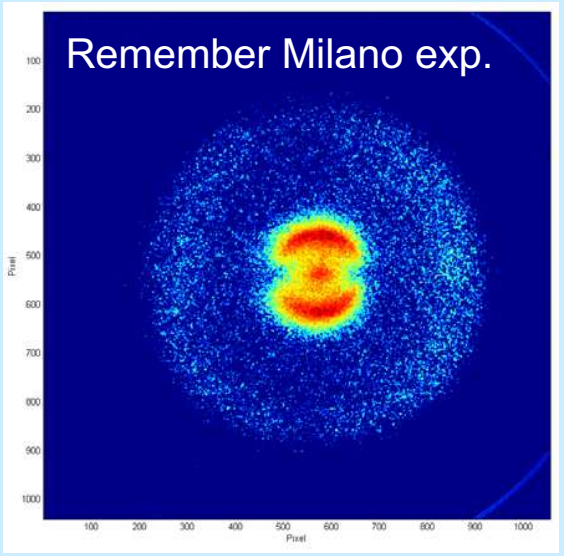


# Pump probe setup

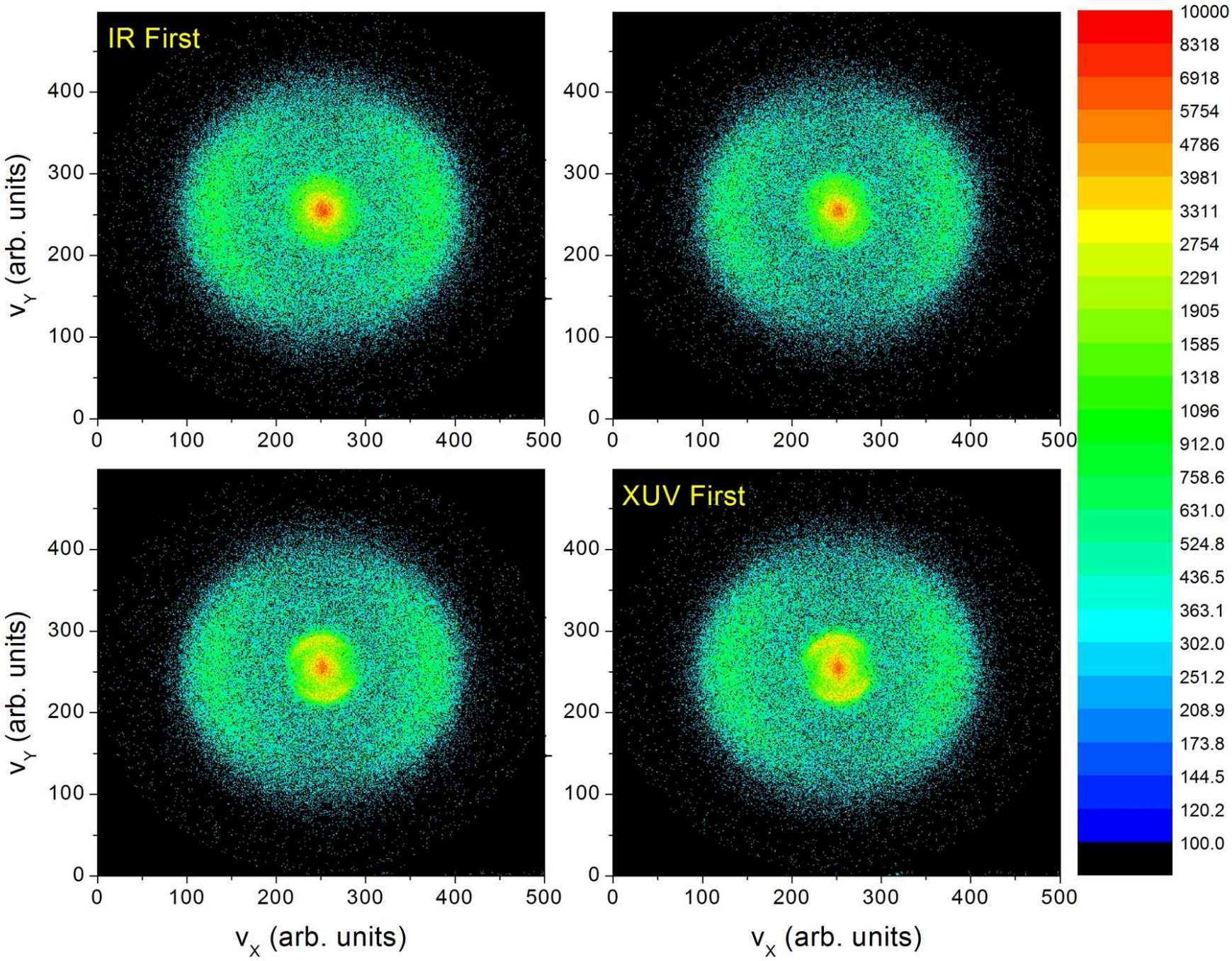


# Finding the two-color overlap

- Use bond-softening in $H_2$
- XUV-production of $H_2^+$
- IR-dissociation into $H^+ + H$




- Velocity and angle-resolved detection of $H^+$

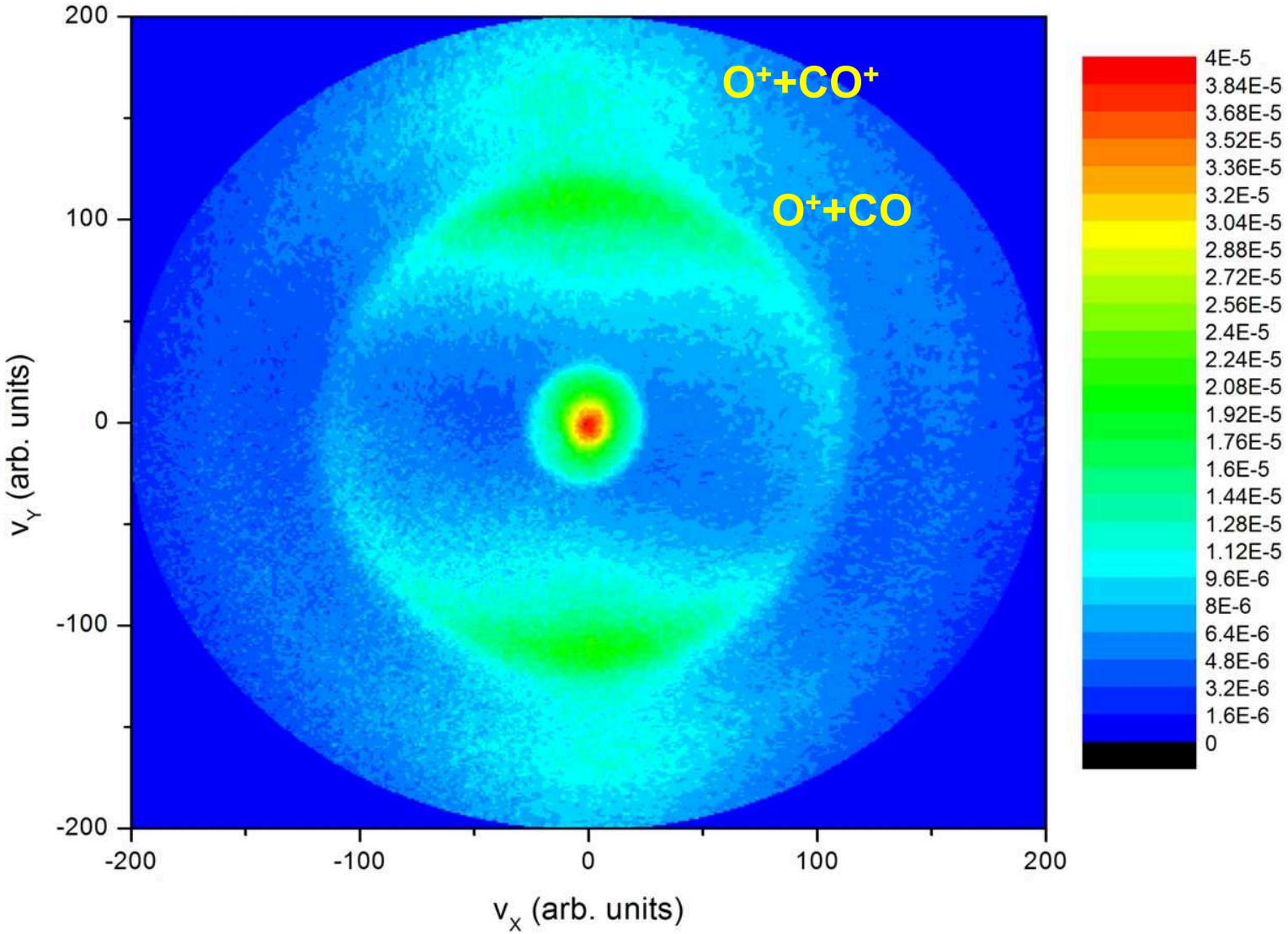

IR First
XUV First
$v_Y$ (arb. units)
$v_X$ (arb. units)
0
100
200
300
400
500
10000
8318
6918
5754
4786
3981
3311
2754
2291
1905
1585
1318
1096
912.0
758.6
631.0
524.8
436.5
363.1
302.0
251.2
208.9
173.8
144.5
120.2
100.0

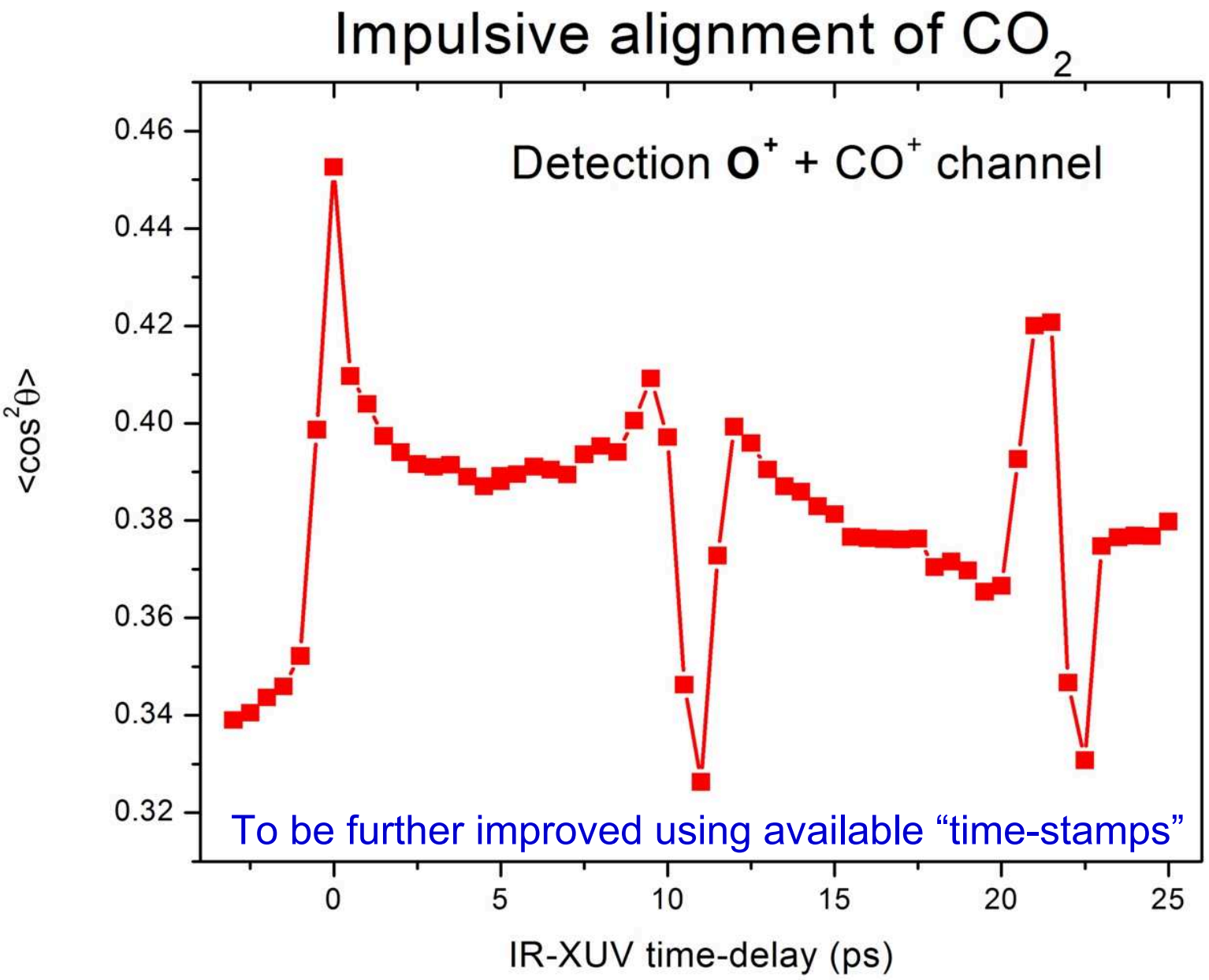
# Time-dependent alignment of $CO_2$

- Use IR to align the molecule
- Use FLASH FEL to dissociatively ionize
- Velocity and angle-resolved detection of $O^+$
- Step towards molecular frame dynamics (fragmentionation, imaging)

# $CO_2$

## IR alignment followed by XUV dissociative ionization

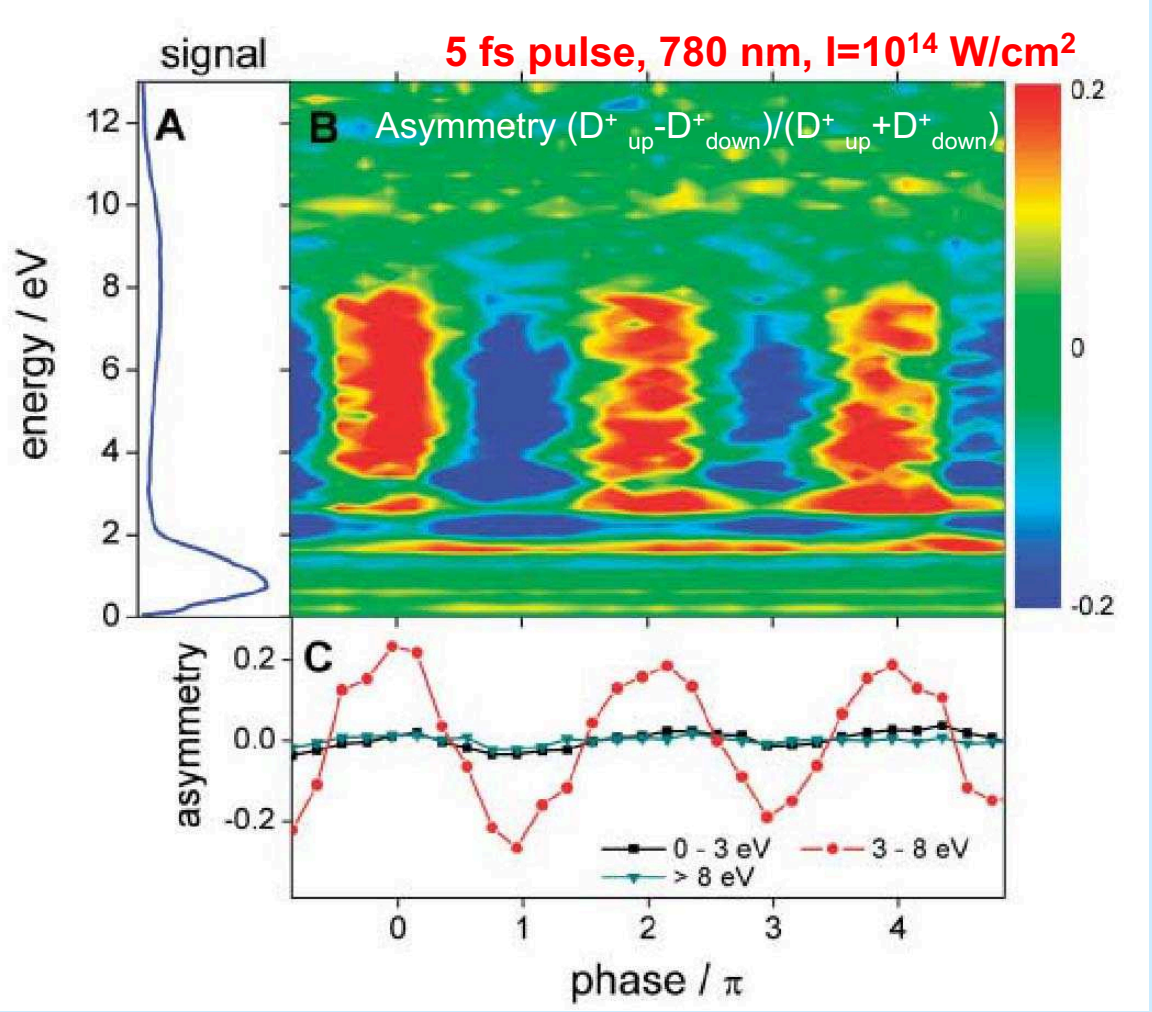



Impulsive alignment of $CO_2$
Detection $O^+$ + $CO^+$ channel
To be further improved using available “time-stamps”
<cos²θ>
0.46
0.44
0.42
0.40
0.38
0.36
0.34
0.32
0
5
10
15
20
25
IR-XUV time-delay (ps)

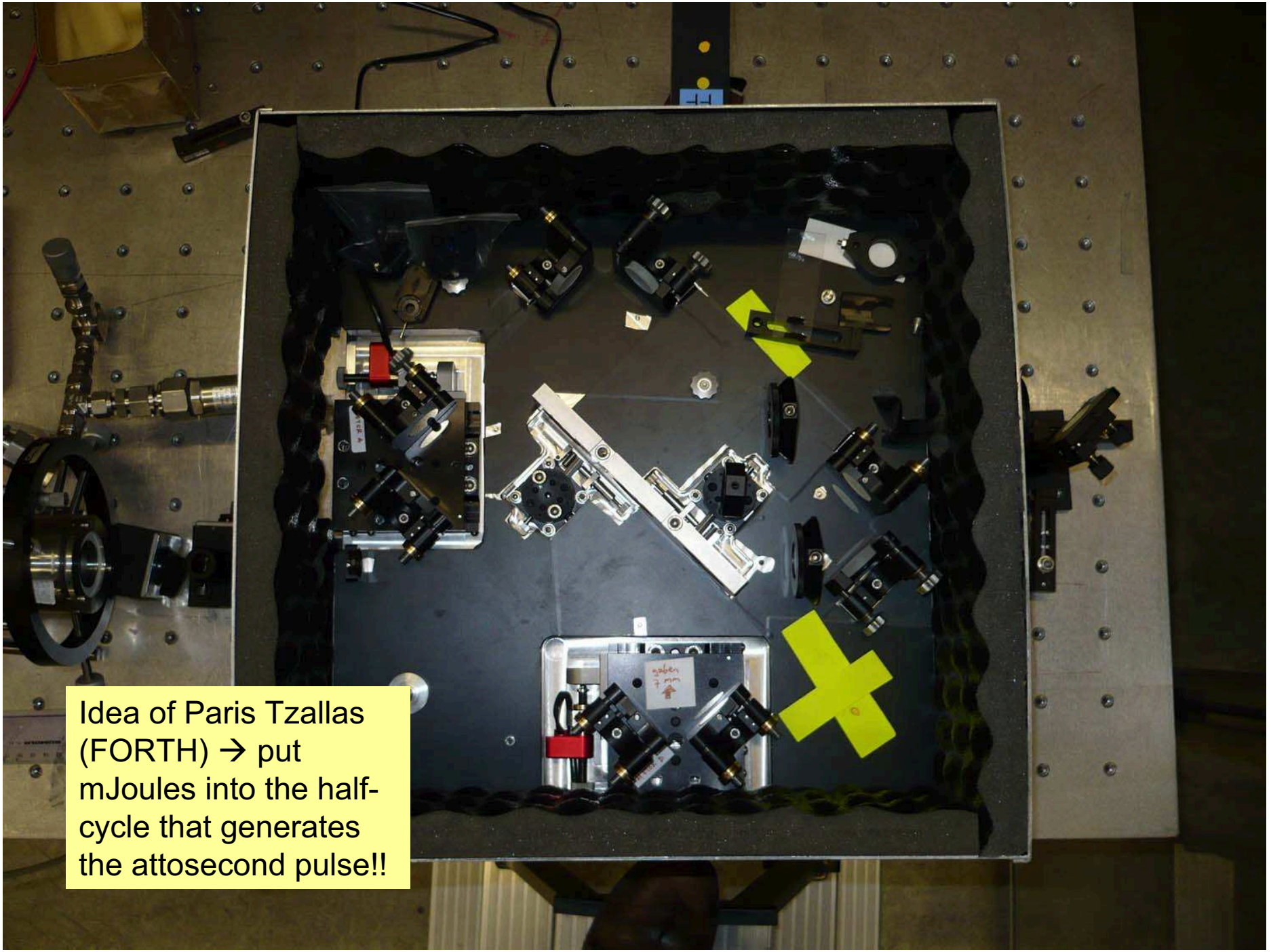
# Conclusions

- Attosecond science now allows to perform IR-XUV pump-probe experiments on a variety of systems
- Experimentation in small-scale attosecond laboratories and at large-scale FEL facilities is complementary, and can lead to a very useful cross-fertilization

# Inspiration: Electron localization in dissociative ionization of $D_2$




*M. Kling et al., Science 312, 246 (2006)*


Idea of Paris Tzallas (FORTH) → put mJoules into the half-cycle that generates the attosecond pulse!!

# $C_{60}$

## Dissociative and multiple ionization