Connecting Attosecond Science and XUV FEL Research

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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)

\[ \tau = 130 \text{ as} \]

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 reveals 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.

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 \( \sim 10 \, \mu\text{Joule/harmonic} \) (\( 3 \times 10^{12} \) 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 \( \sim 10^6 \) photons/pulse or less
1) Our choices for going towards XUV-XUV experiments

- Amplification of CEP-stable 30 fsec pulses to TW-level
- Specialized detectors: Development of hybrid COLTRIMS/Velocity Map Imaging detector
- Polarization gating for isolated attosecond pulses + few-cycle UV
- Development of (chirped) XUV multilayer optics
- Special target injection
- Successful Attosecond Experiment
2) Exploiting the complementarity between harmonics-based and FEL-based experiments
Electrons from (dissociative) ionization of $O_2$

"FEL-tourism" 2/2007
Parasitic use of FEL away from focus
Exploring the utility of Velocity Map Imaging at the FEL: \( \text{O}_2 \)

Image showing the photoelectron kinetic energy distribution with peaks labeled for various states as follows:

- \( \text{O}(3p',3s'') \rightarrow \text{O}^+(4s) \)
- \( \text{b}^4\Sigma_g^- \)
- \( \text{a}^4\Pi u \)
- \( \text{X}^2\Pi_g \)
- \( \text{c}^4\Sigma_u^- \)
- \( \text{B}^2\Sigma_g^- \)
- \( \text{c}^2\Sigma_u^- \)

Additional text reads:

Further atomic autoionization


diagram with peaks at specific energy levels.
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.
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 $H^+$

Remember Milano exp.
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 (fragmentation, imaging)
CO$_2$

IR alignment followed by XUV dissociative ionization

![Graph showing the IR alignment followed by XUV dissociative ionization of CO$_2$. The graph indicates the distribution of products such as O$^+$+CO$^+$ and O$^+$+CO.](image-url)
Impulsive alignment of CO$_2$

Detection O$^+$ + CO$^+$ channel

To be further improved using available “time-stamps”
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$_2$

5 fs pulse, 780 nm, $I=10^{14}$ W/cm$^2$

Asymmetry $(D^+_{\text{up}} - D^+_{\text{down}})/(D^+_{\text{up}} + D^+_{\text{down}})$

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