Participant presentation:

Aarhus University

Denmark

Lars Bojer Madsen

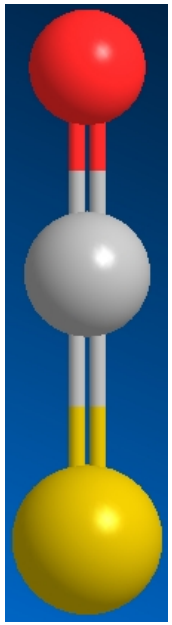
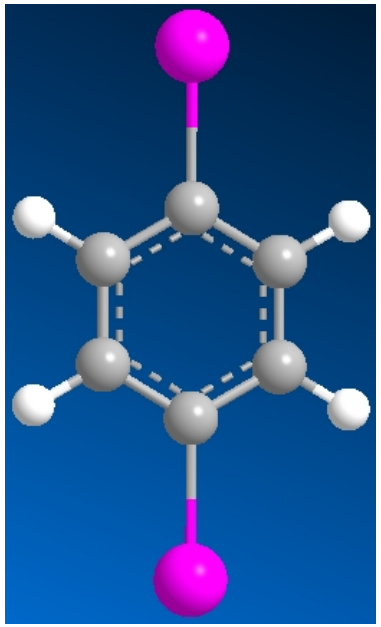
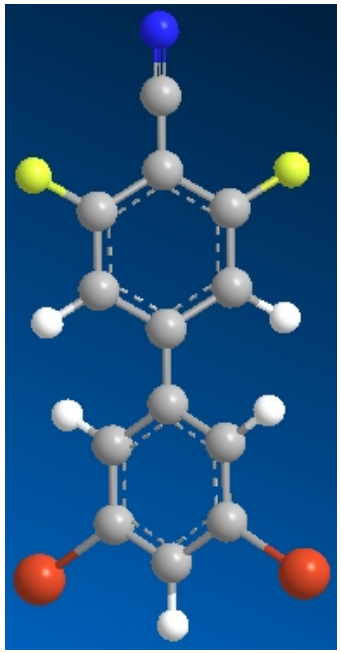
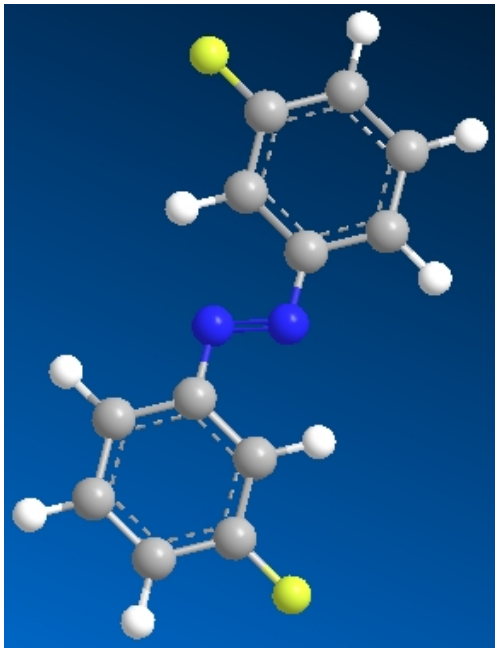
Department of Physics and Astronomy

Henrik Stapelfeldt

Department of Chemistry

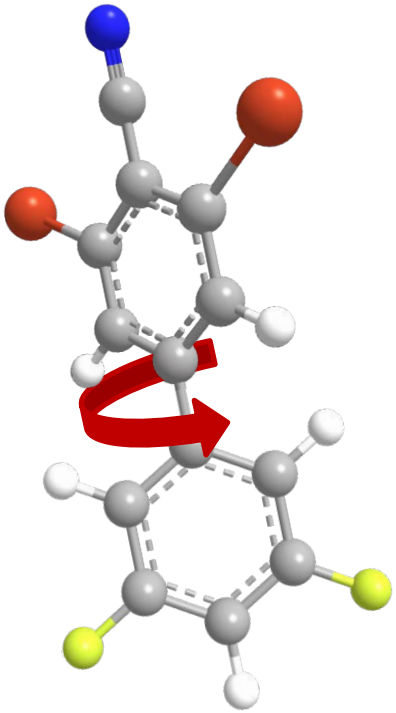
MEDEA Kick-off
MBI, Berlin
January 19-20, 2015

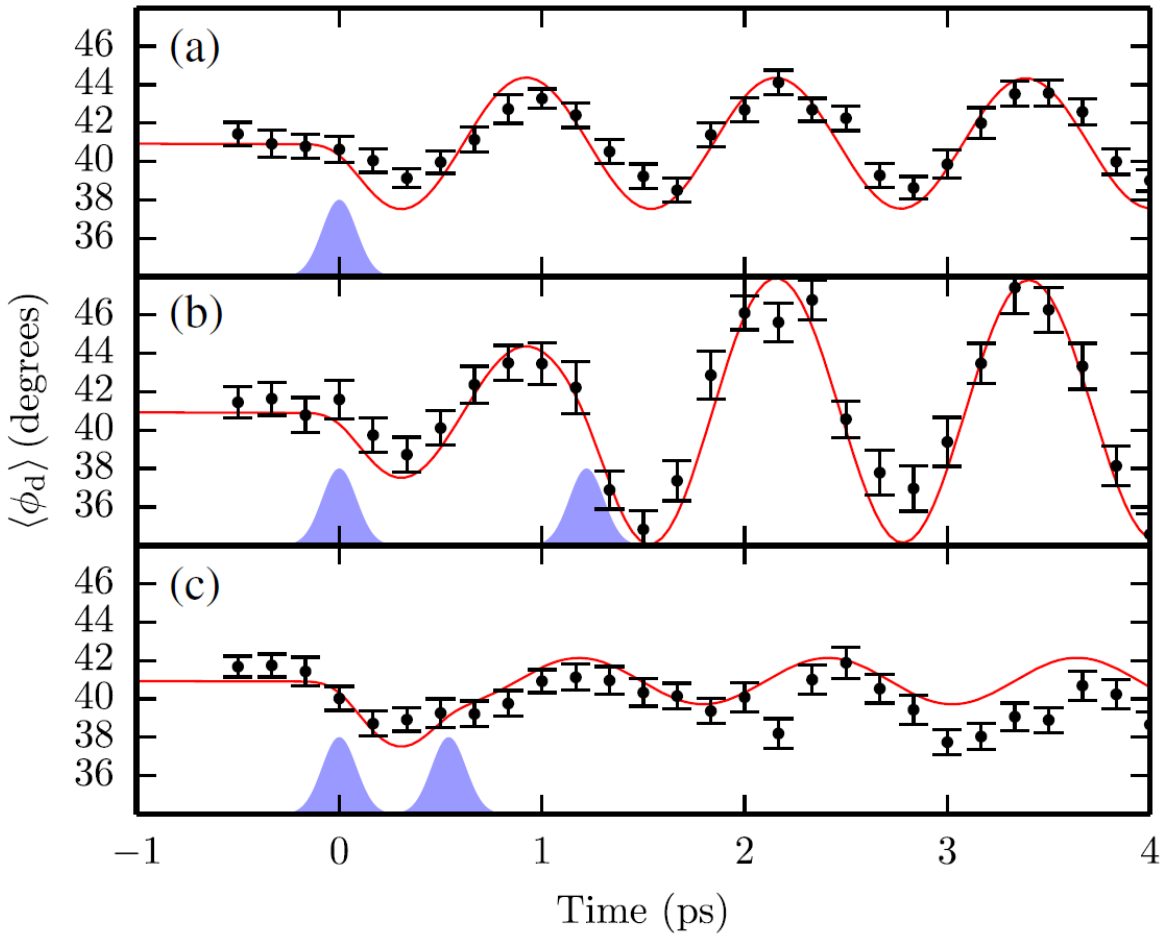
Alignment and Orientation of Gas-phase Molecules

Joint interests and projects with Jochen Küpper (DESY)
→ MEDEA

Use aligned / oriented molecules to image ultrafast structural dynamics

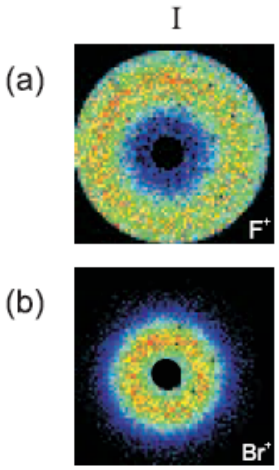




Christensen *et al.*
Phys. Rev. Lett. **113**, 073005 (2014)

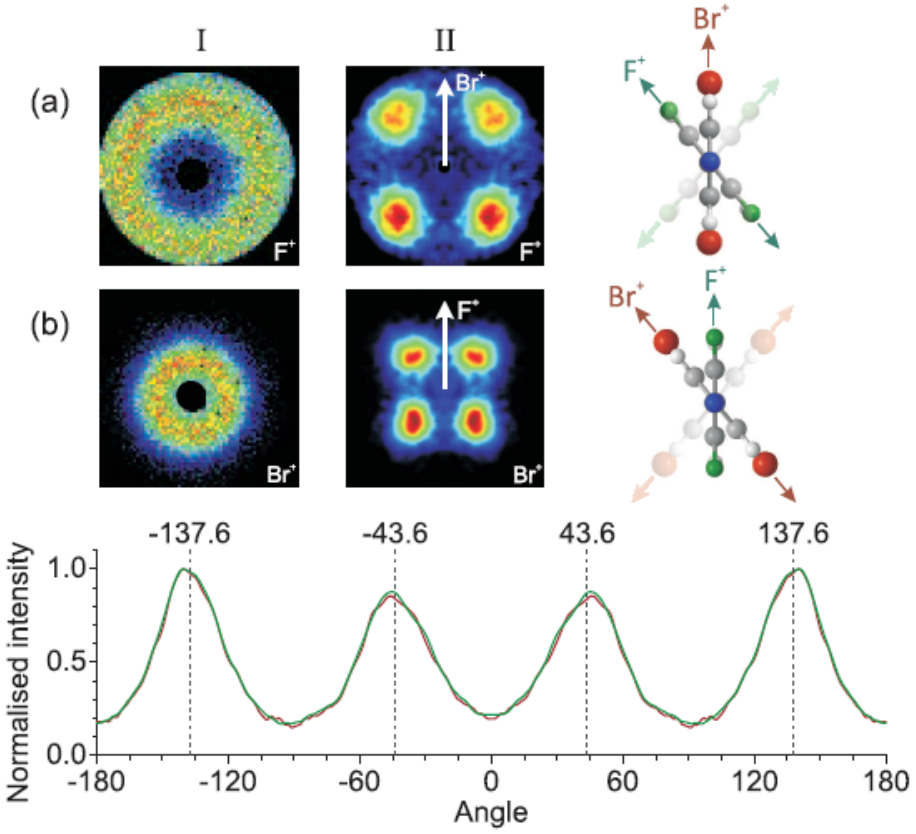
Correlations of experimental observables

Dihedral angle



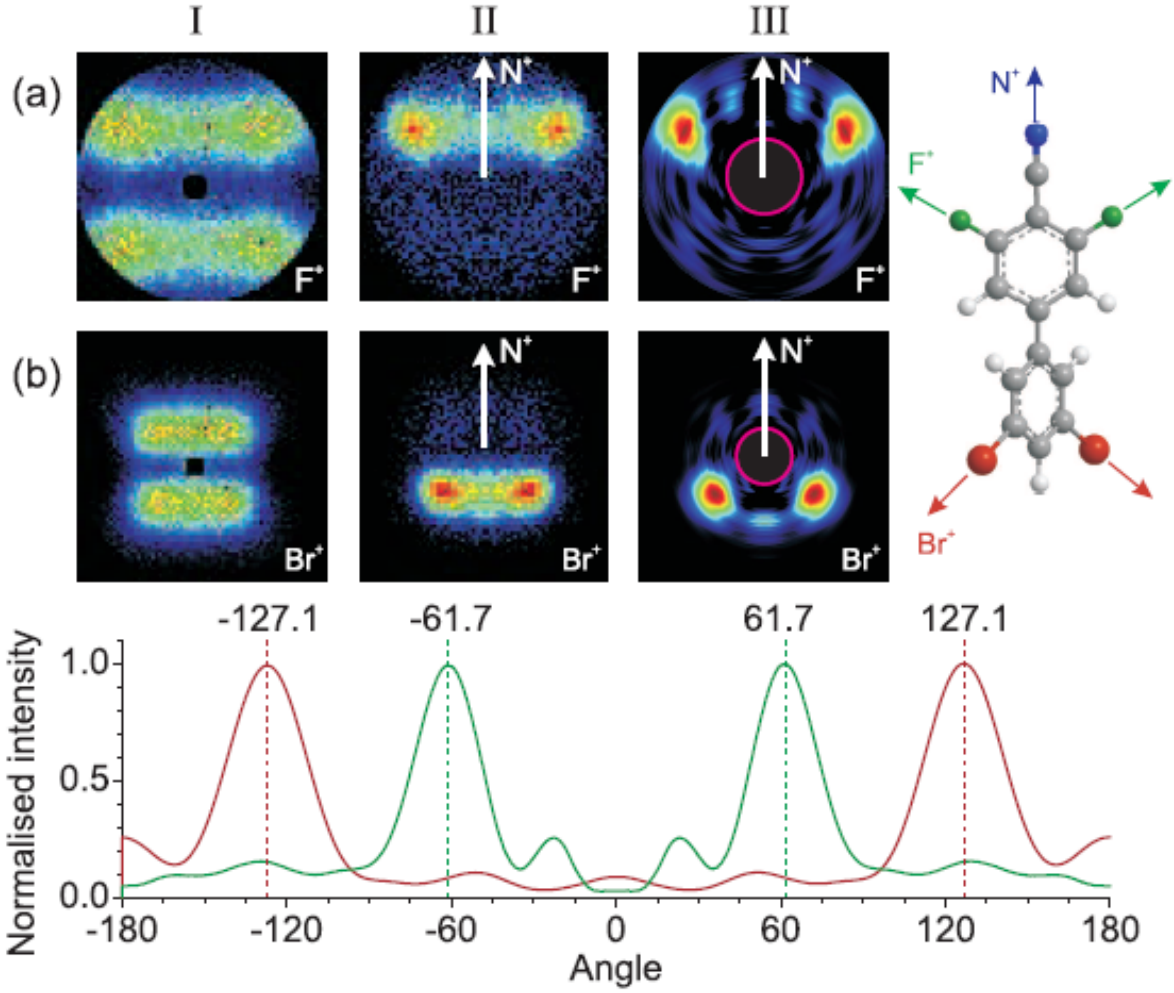
Correlation between ion recoil directions

Covariance detection of F^+ and Br^+ ions using PImMS
$\rightarrow$ Dihedral angle



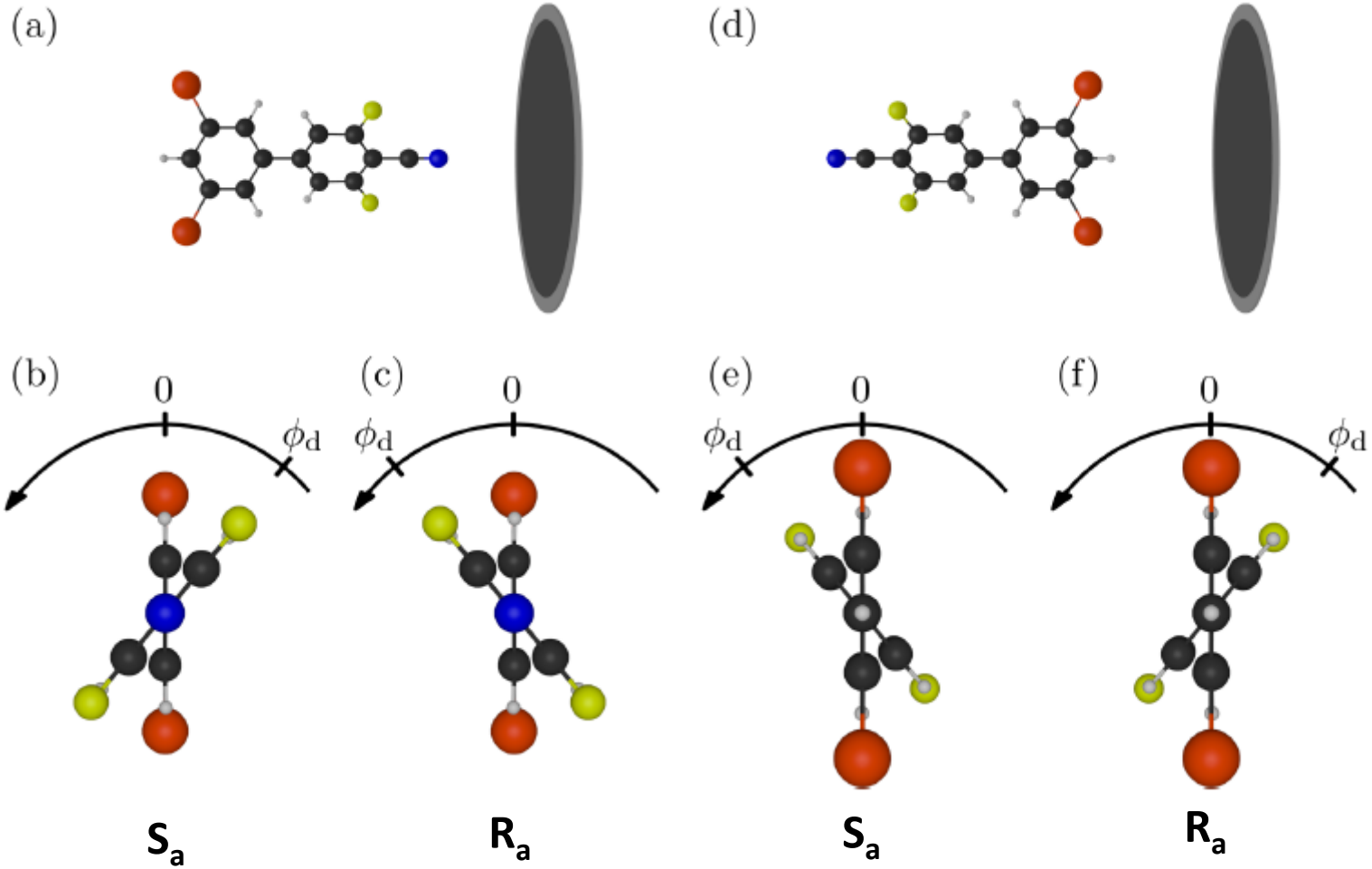
Slater *et al*. Phys. Rev. A. **89**, 011401(R) (2014)

More structural information



Correlations of experimental observables

Imaging specific enantiomers

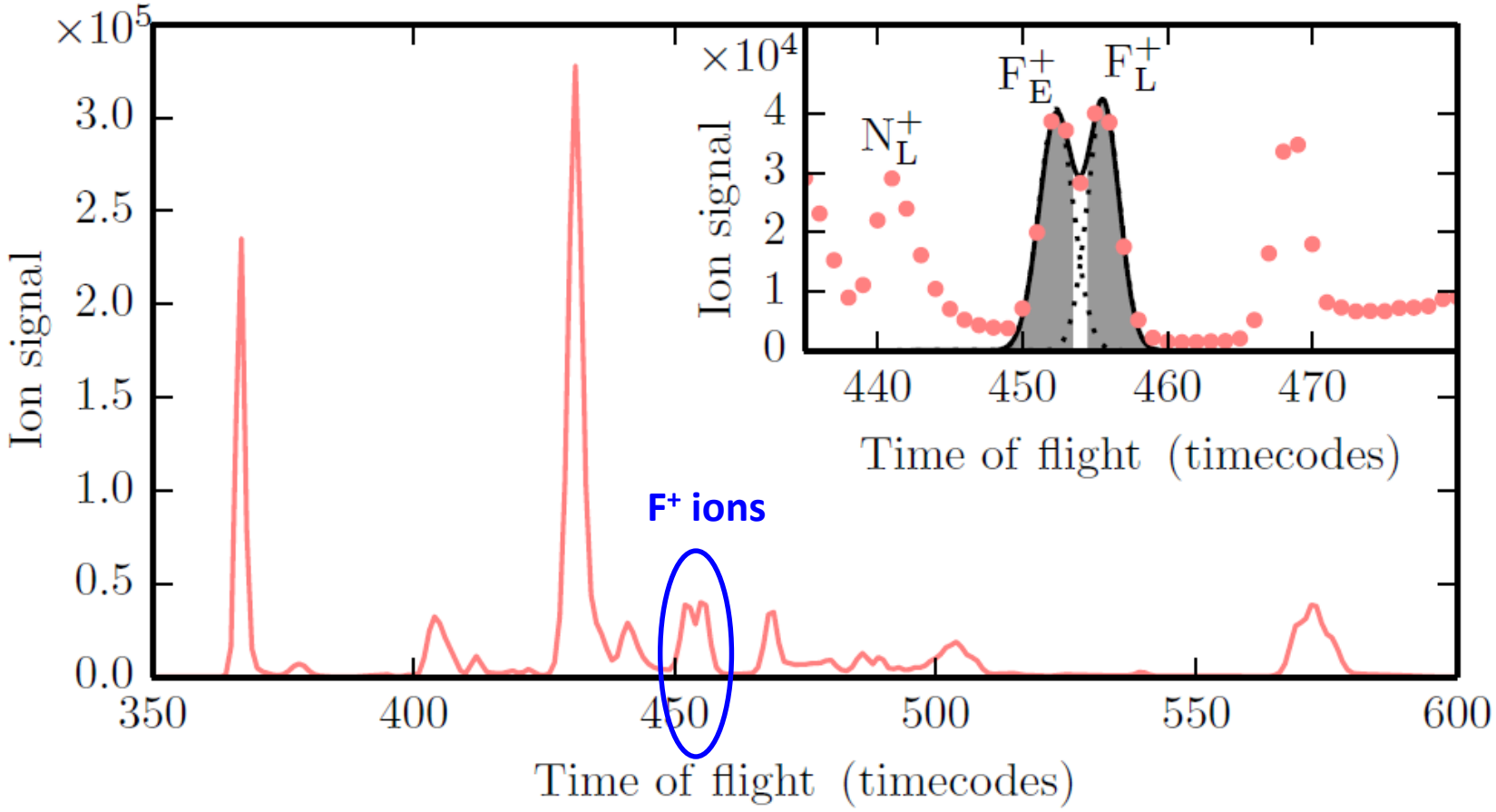


S_a **R_a** **S_a** **R_a**

ϕ_d : 140° / 320° 40° / 220° 40° / 220° 140° / 320°

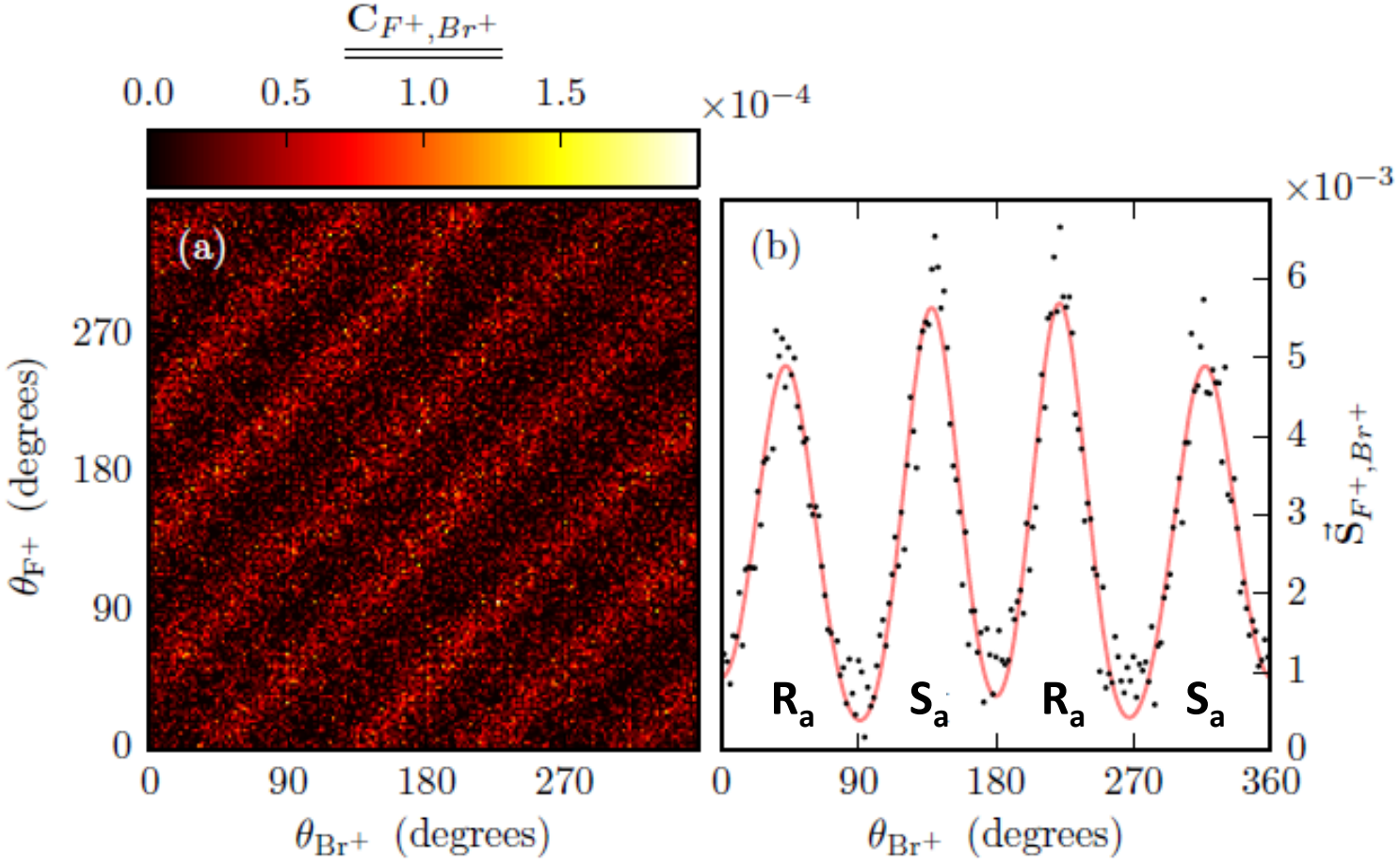
Correlations of experimental observables

Imaging specific enantiomers



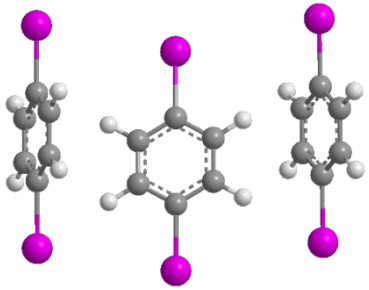
Correlations of experimental observables

Imaging specific enantiomers

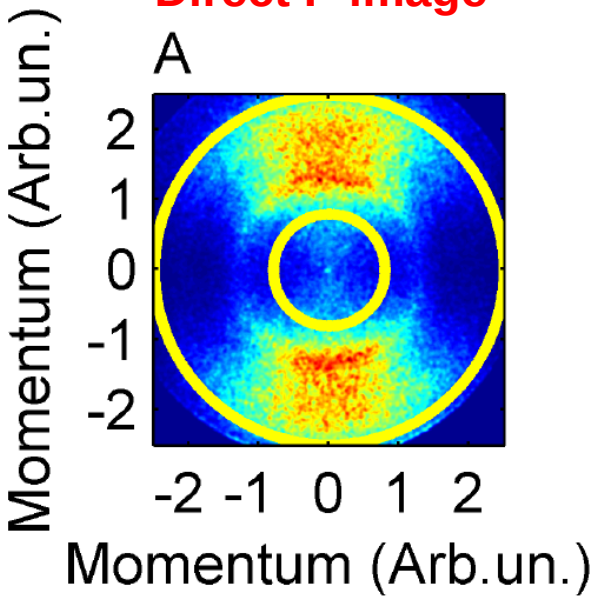


Correlations of experimental observables

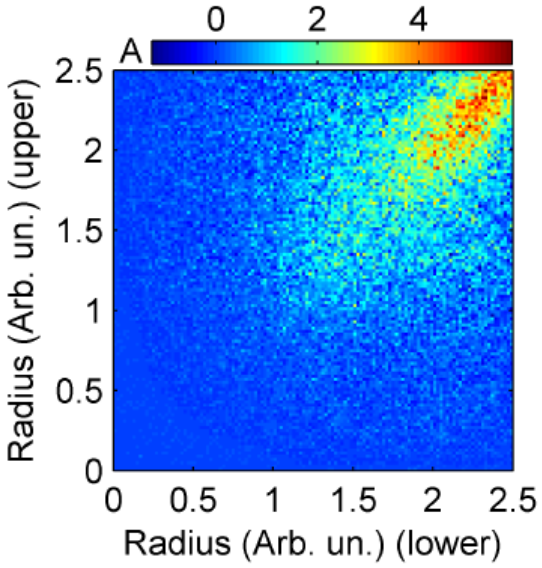
Coulomb explosion of di-iodobenzene



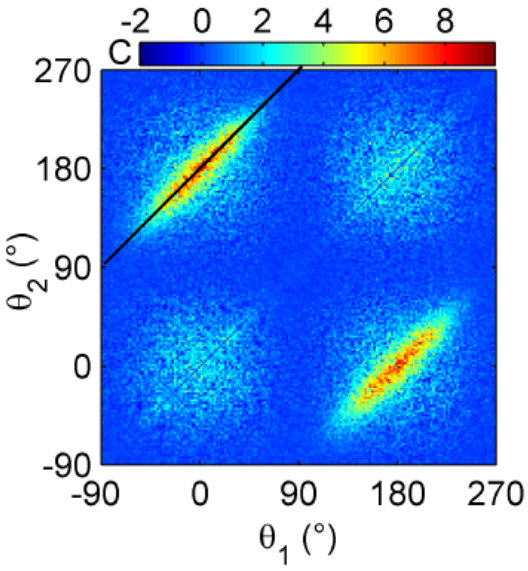
Direct I^+ image



Radial covariance



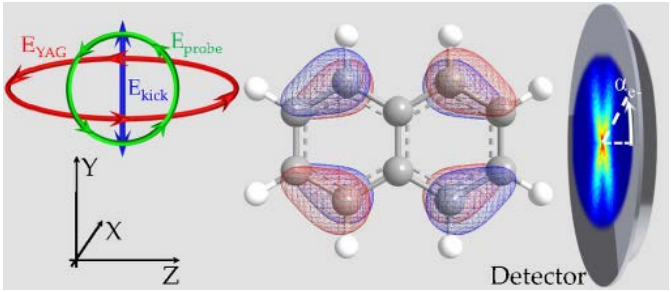
Angular covariance

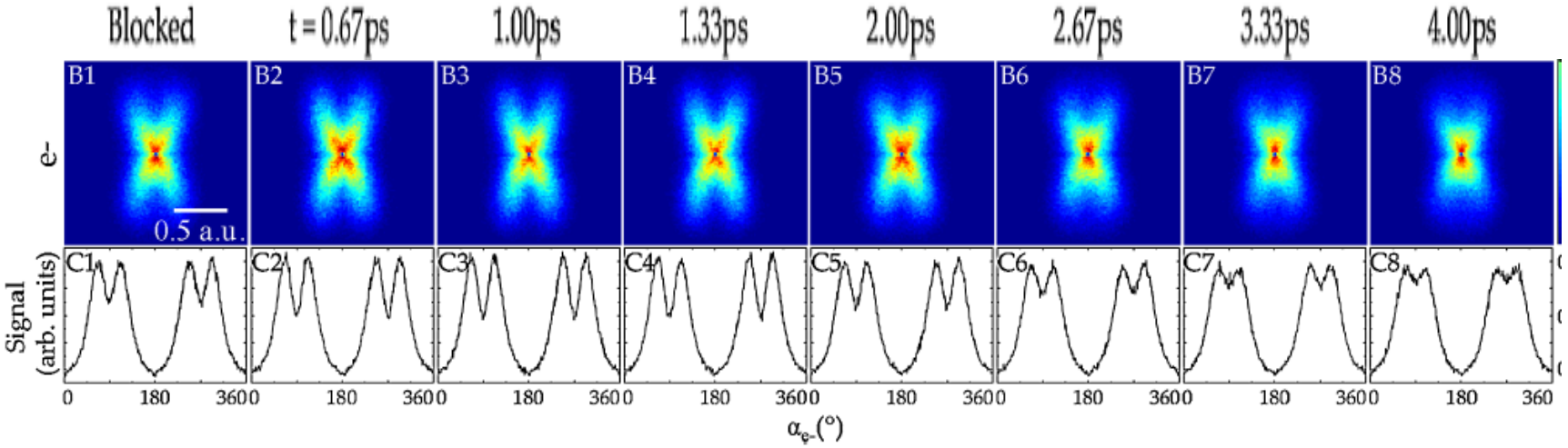


Ionization of aligned molecules with (ultrashort) laser pulses

Imaging electrons

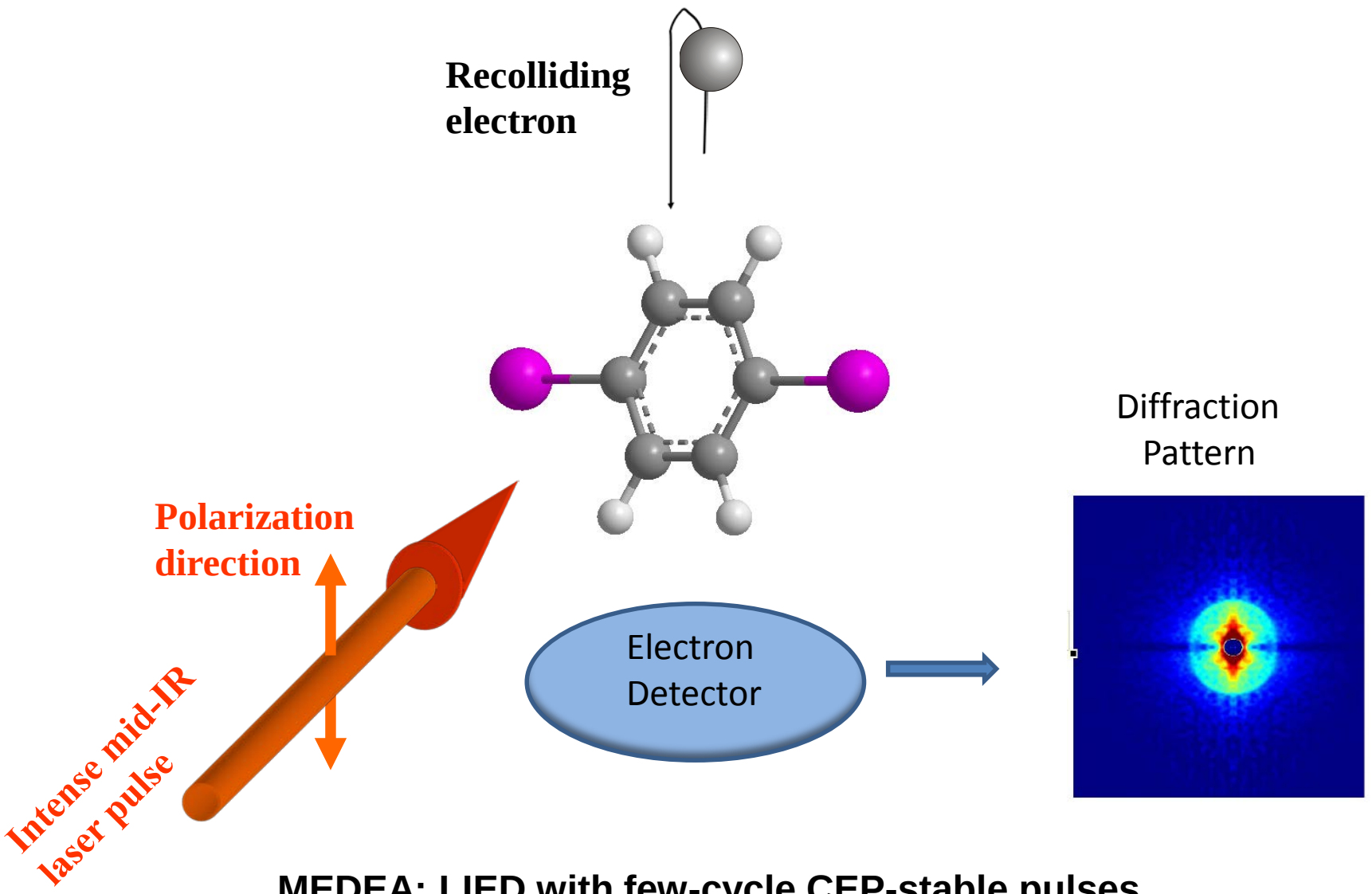
Single Ionization of naphthalene with circularly intense polarized pulse





MEDEA: Ionization with few-cycle pulses and VUV / XUV pulses
Saclay + Milan

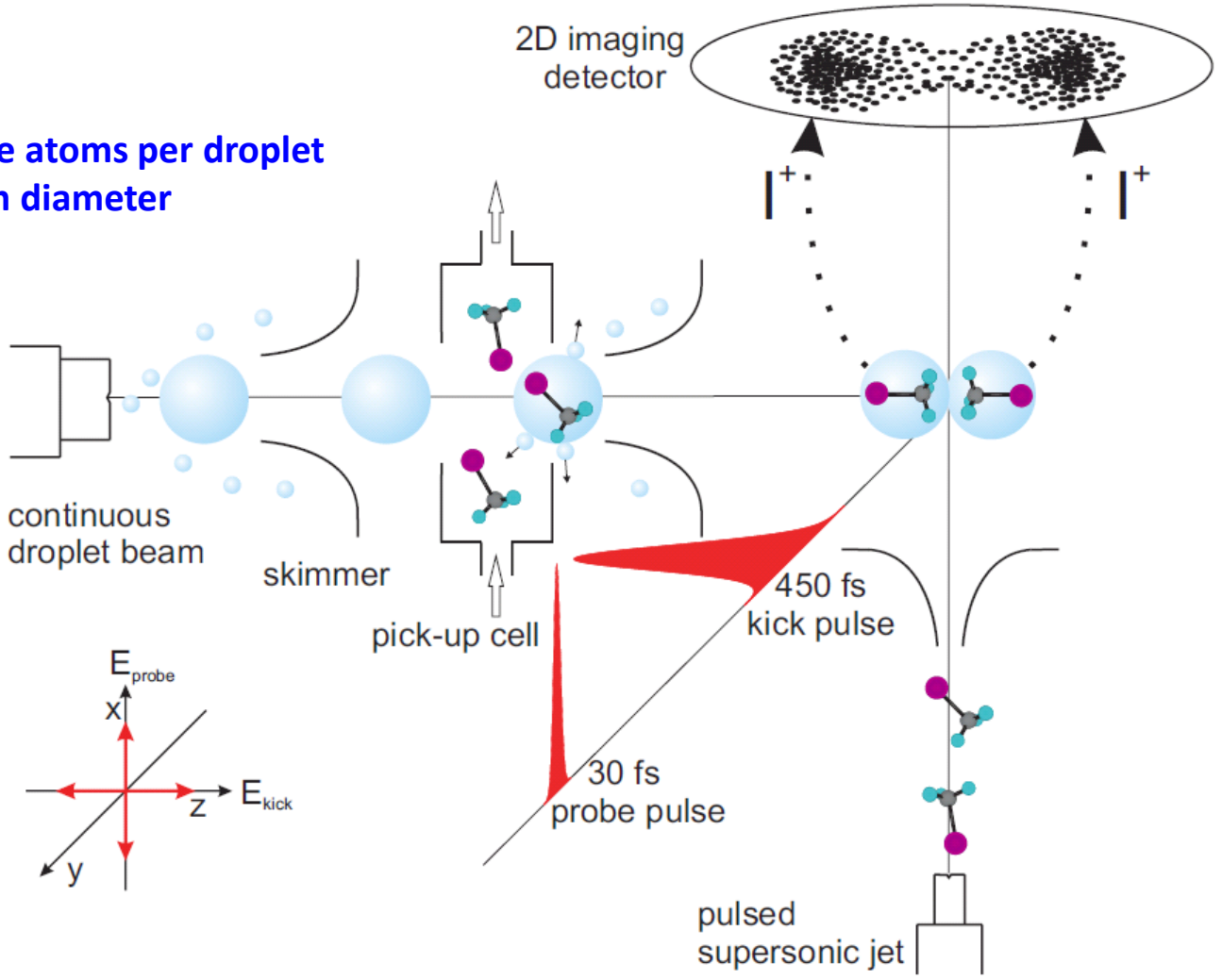
Laser-induced electron diffraction



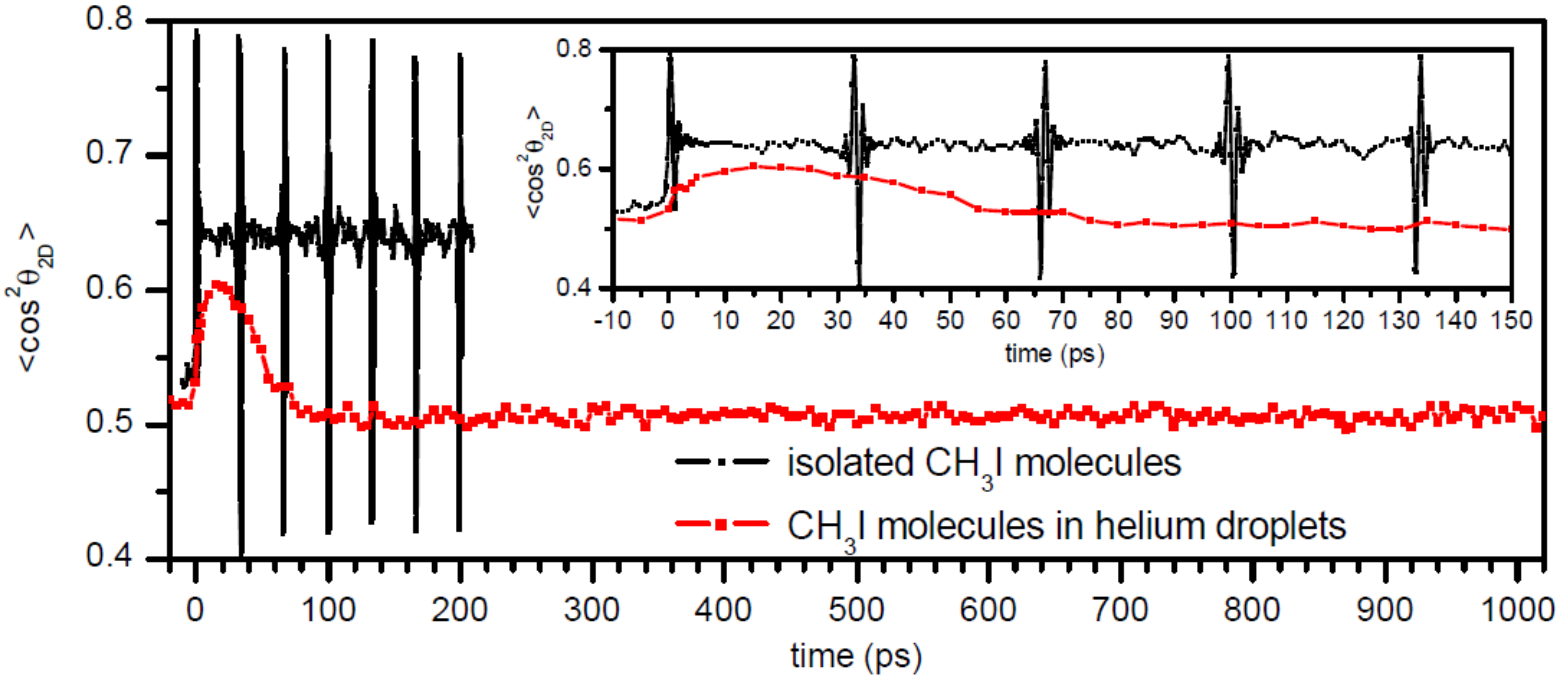
MEDEA: LIED with few-cycle CEP-stable pulses
Saclay + Milan + MBI

Alignment of Molecules in He droplets

10^4 He atoms per droplet
10 nm diameter



Alignment of Molecules in He droplets



Pentlehner et al. Phys. Rev. Lett. 110, 093002 (2013)

MEDEA: Freiburg

(Some of) The group

AU-theory (Lars Bojer Madsen)

Main expertise: Theoretical strong-field and attosecond physics

Since 2012 a focus area has been

– *How to deal with more than a single-active electron*?

AU-theory Research

- Processes that are studied
  - pump-probe, (multi)-photon ionization, light generation, transient absorption
- Theory that is being developed and used
  - TDSE, TD-RAS-SCF, TD-RAS-CI, TD-RAS-CC,
  - Tunneling, Strong-Field Approximation, Classical Trajectory Monte Carlo, Monte Carlo Wave Packet

Some examples from 2014

PHYSICAL REVIEW A **89**, 063416 (2014)

Time-dependent restricted-active-space self-consistent-field theory for laser-driven many-electron dynamics. II. Extended formulation and numerical analysis

Haruhide Miyagi and Lars Bojer Madsen

PHYSICAL REVIEW A **90**, 062508 (2014)

Time-dependent generalized-active-space configuration-interaction approach to photoionization dynamics of atoms and molecules

S. Bauch,[1,2] L. K. Sørensen,[1,2] and L. B. Madsen[1]

PHYSICAL REVIEW A **90**, 063408 (2014)

Dissociative ionization of H_2^+ using intense femtosecond XUV laser pulses

Lun Yue and Lars Bojer Madsen

PRL **113,** 103005 (2014) PHYSICAL REVIEW LETTERS week ending 5 SEPTEMBER 2014

Low-Energy Photoelectrons in Strong-Field Ionization by Laser Pulses with Large Ellipticity

D. Dimitrovski,[1,*] J. Maurer,[2] H. Stapelfeldt,[2] and L. B. Madsen[3]

PRL **113,** 223002 (2014) PHYSICAL REVIEW LETTERS week ending 28 NOVEMBER 2014

Nonlinear Dichroism in Back-to-Back Double Ionization of He by an Intense Elliptically Polarized Few-Cycle Extreme Ultraviolet Pulse

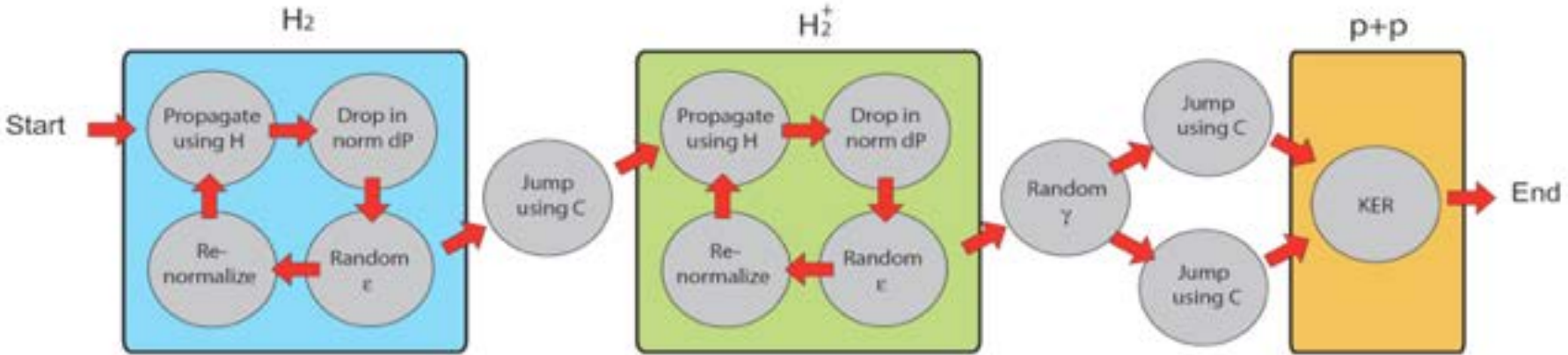
J. M. Ngoko Djiokap,[1] N. L. Manakov,[2] A. V. Meremianin,[2] S. X. Hu,[3] L. B. Madsen,[4] and Anthony F. Starace[1]

Work within MEDEA WP-1

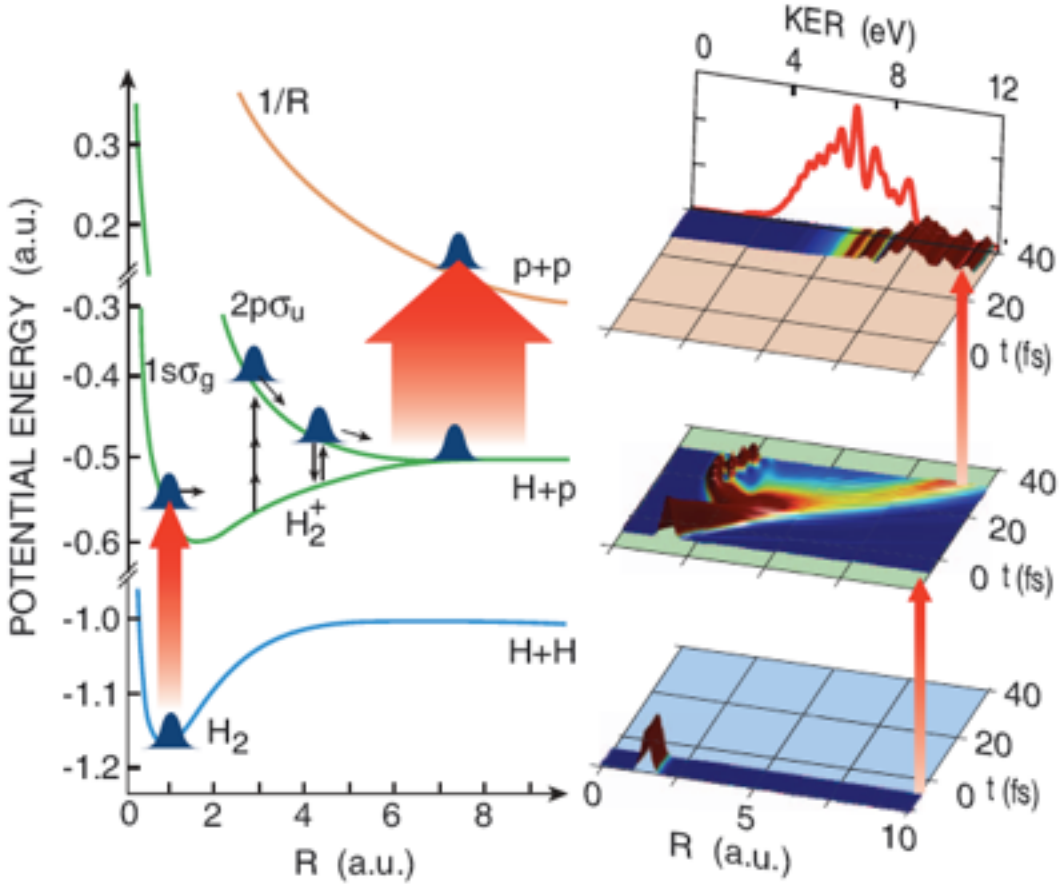
- "Interpretation and theoretical support will be provided by MAD using an ab initio quantum model to simulate the coupled electron-nuclear dynamics. However, this model cannot be applied to diatomic molecules with more than two electrons, because of the amount of computational power required in these cases exceeds that presently available. Therefore, the development of new theoretical models with predictive power for the analysis of this process is an absolute prerequisite for progress in the understanding of general diatomic molecules with attosecond XUV-IR pump-probe spectroscopy. To this end, *AU-theory (ESR AU-1) will consider the usefulness of Monte Carlo wavepacket techniques in describing the nuclear kinetic energy release spectra. Based on prior validation of the method by comparison with experimental results on H_2 and D_2, AU-theory will investigate the possibilities of extending it to attosecond XUV-IR pump-probe scenarios, including a model description of autoionization*. "

Monte Carlo wave packet (MCWP) method

- Describes open quantum systems (electron leaks out)
- Non-hermitian evolution of wave functions
- Equivalent to master equation
- Stochastic jumps between states (Hilbert spaces)



MCWP method to Dissociative double ionization



$$H = H_s - \frac{i}{2}\sum_m C_m^\dagger C_m$$

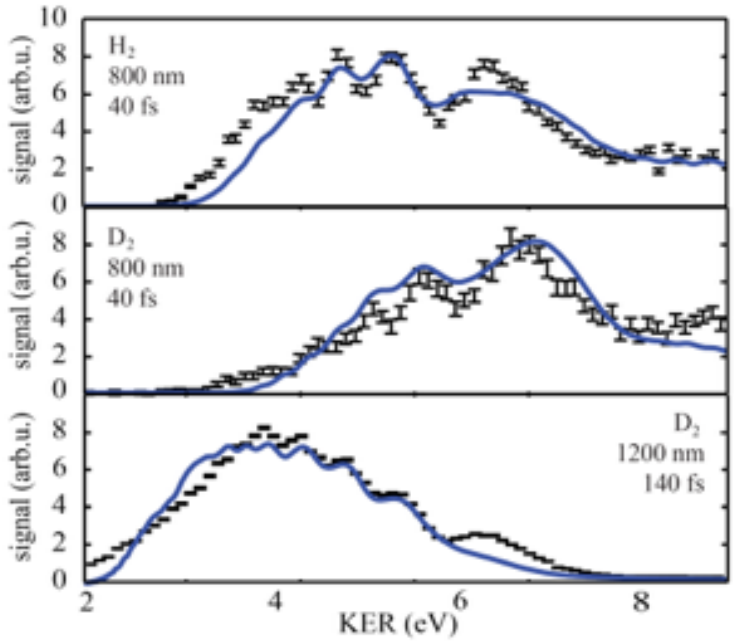
Example of results

Final result

- Average over many runs and focal-volume
- Include many different instants of ionization

Experiment

- Staudte et al., Phys. Rev. Lett **98**, 073003 (2007)
- 800 nm, 40 fs, 10^{14} W/cm^2



Leth, Madsen, Mølmer, PRL 103, (2009).

Summery

- AU-theory has experience with strong-field and attosecond physics and associated theoretical modeling. Currently efforts are made to elucidate the time-dependent many-electron problem with new methodology. In MEDEA focus will be on further development of the Monte Carlo Wave Packet Method for dissociative ionization in relation to experiments from MEDEA network groups.