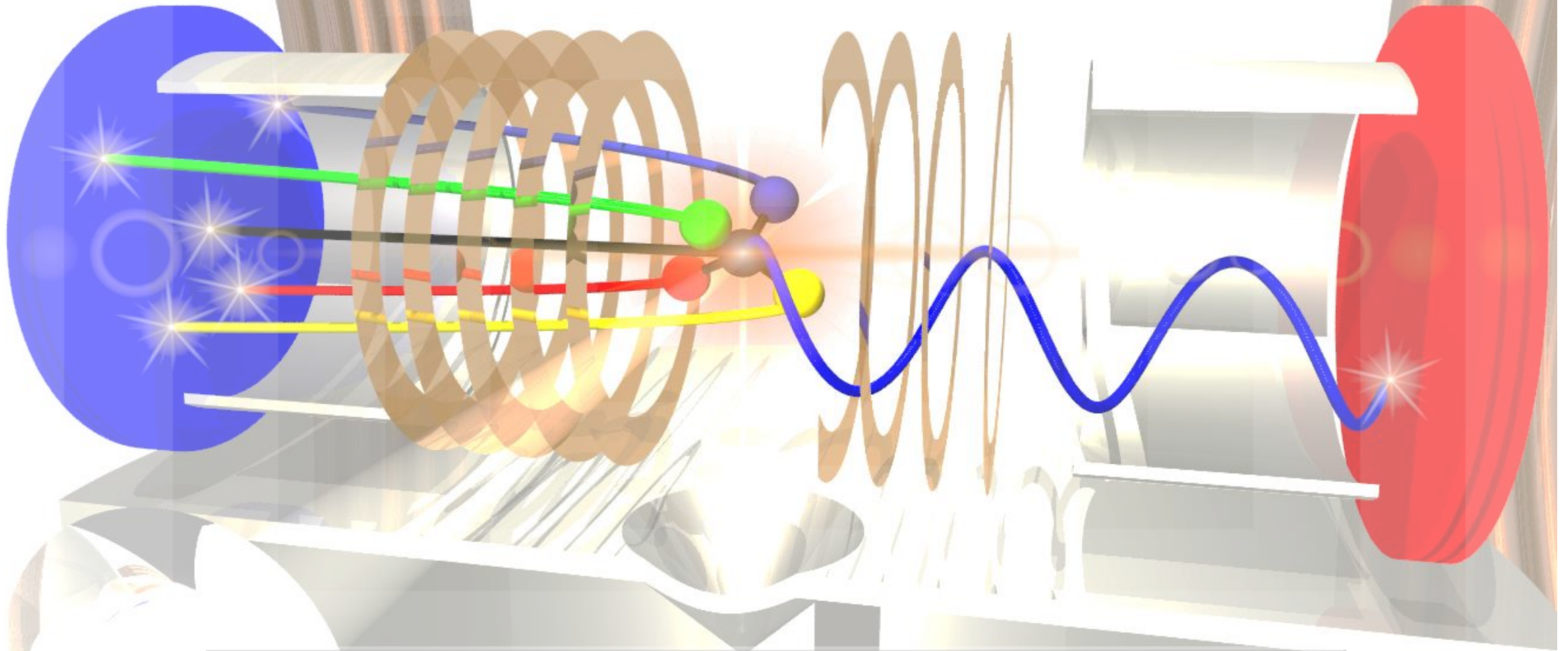


The Power of Coincidence

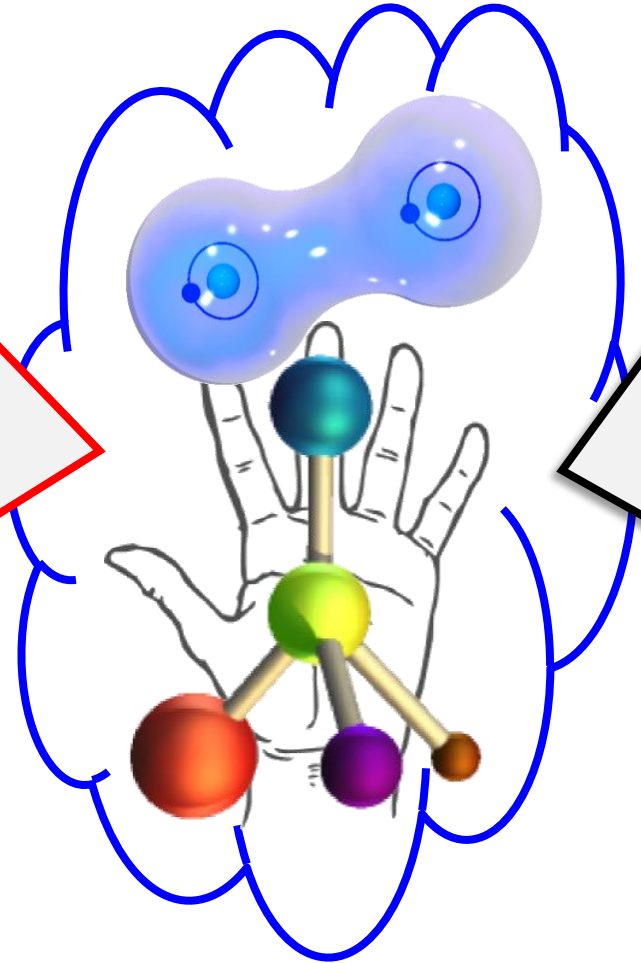


Reinhard Dörner, Goethe University Frankfurt

$$\Psi(\vec{R}_i, \vec{r}_j, t)$$

Imaging Molecules (nuclei) :

- Diffraction
- Coulomb Explosion Imaging



Imaging Molecules (electrons) :

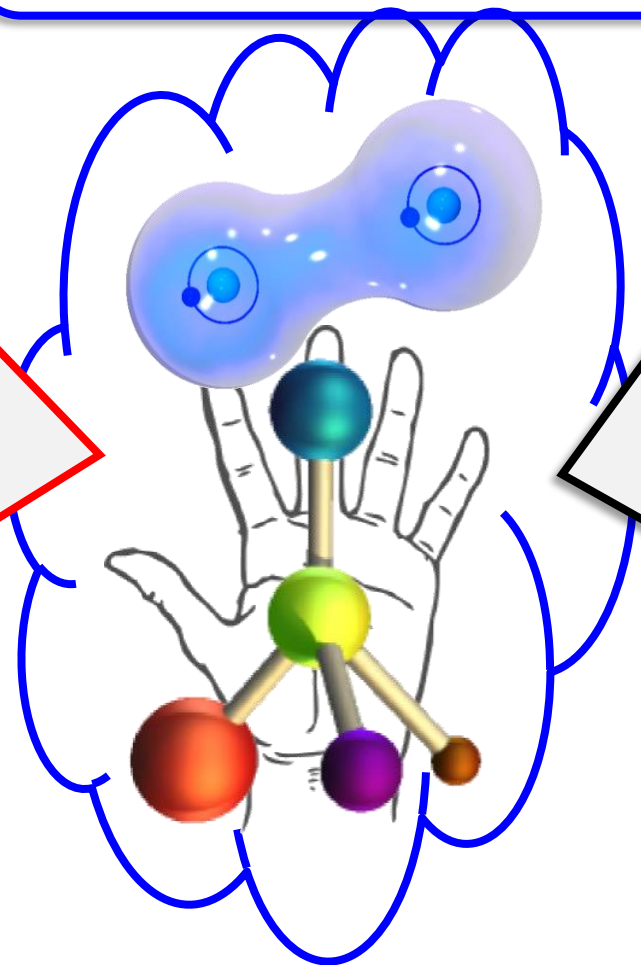
- Image Electrons
- Momentum Space
- Correlations

Coincidence Imaging

$$\Psi(\vec{R}_i, \vec{r}_j, t)$$

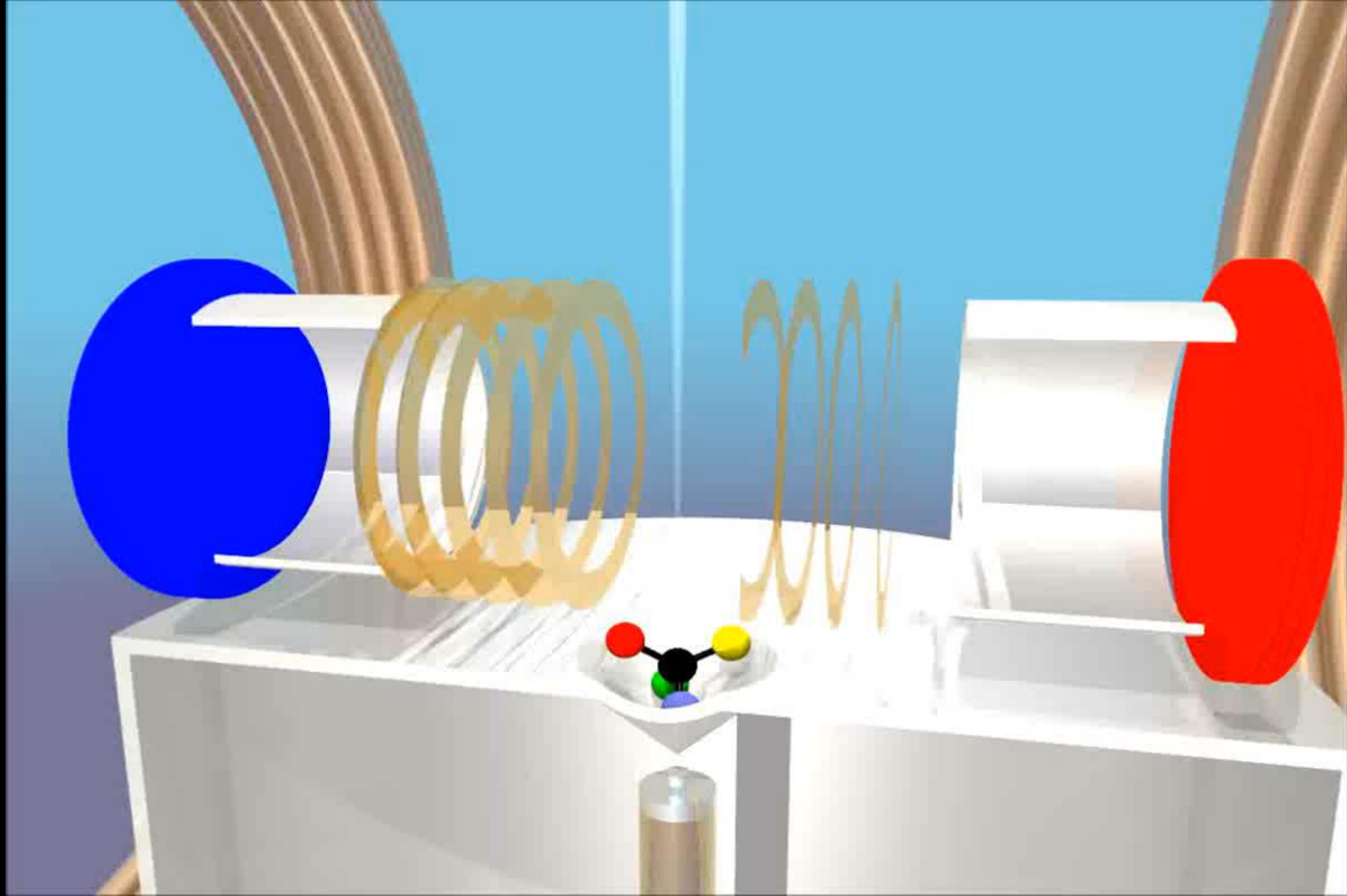
Imaging Molecules (nuclei) :

- Diffraction
- Coulomb Explosion Imaging



Imaging Molecules (electrons) :

- Image Electrons
- Momentum Space
- Correlations



Coincidence Imaging

$$\Psi(\vec{R}_i, \vec{r}_j, t)$$

Imaging Molecules (nuclei) :

- Diffraction
- Coulomb Explosion Imaging

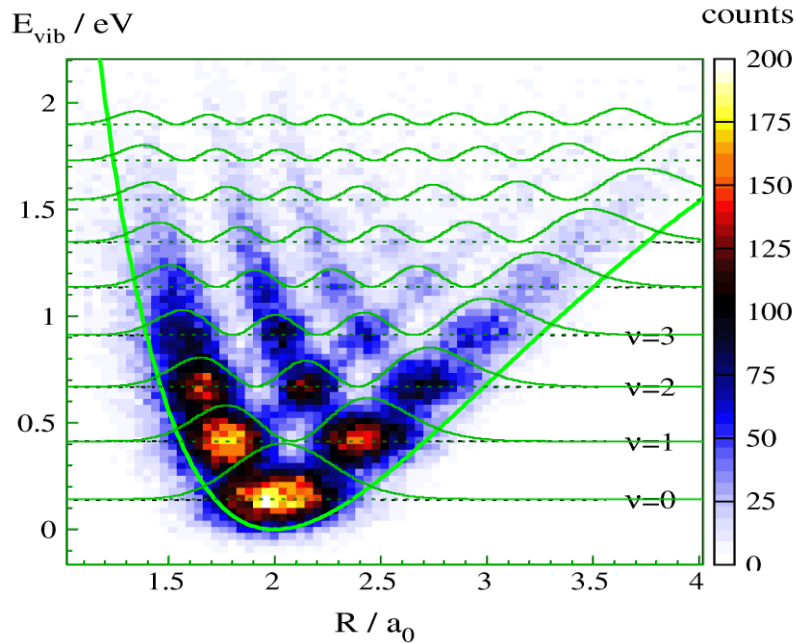
Imaging Molecules (electrons) :

- Image Electrons
- Real Space
- Momentum Space
- Correlations



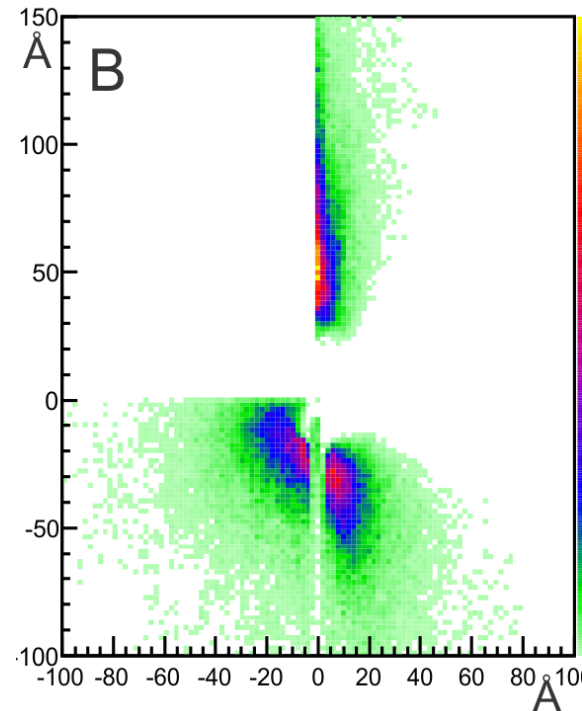
Coulomb Explosion Imaging: Frontiers

Frontier in Precision



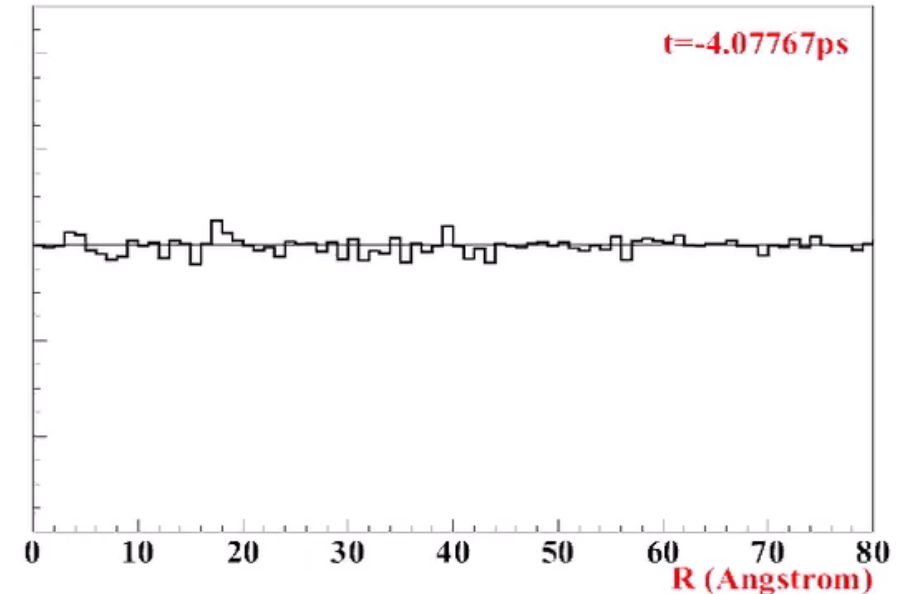
„Spatial Imaging at the quantum limit“
L. Schmidt et al
PRL 108, 73202 (2012)

Frontier in size (spatial extension)



“Efimov State of He_3 ”
Kunitiski et al.
Science, 348 (2015) 551

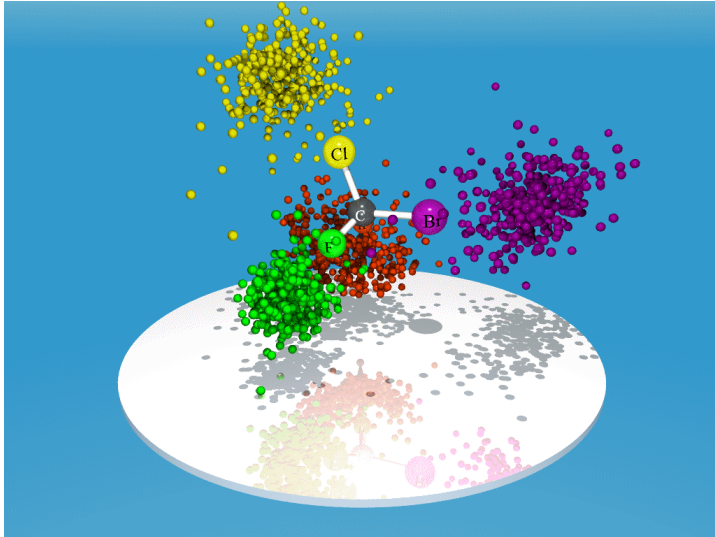
Frontier in size – time domain (spatial extension)



Kunitiski et al.
Nat. Phys. **17**, 174–178 (2021)

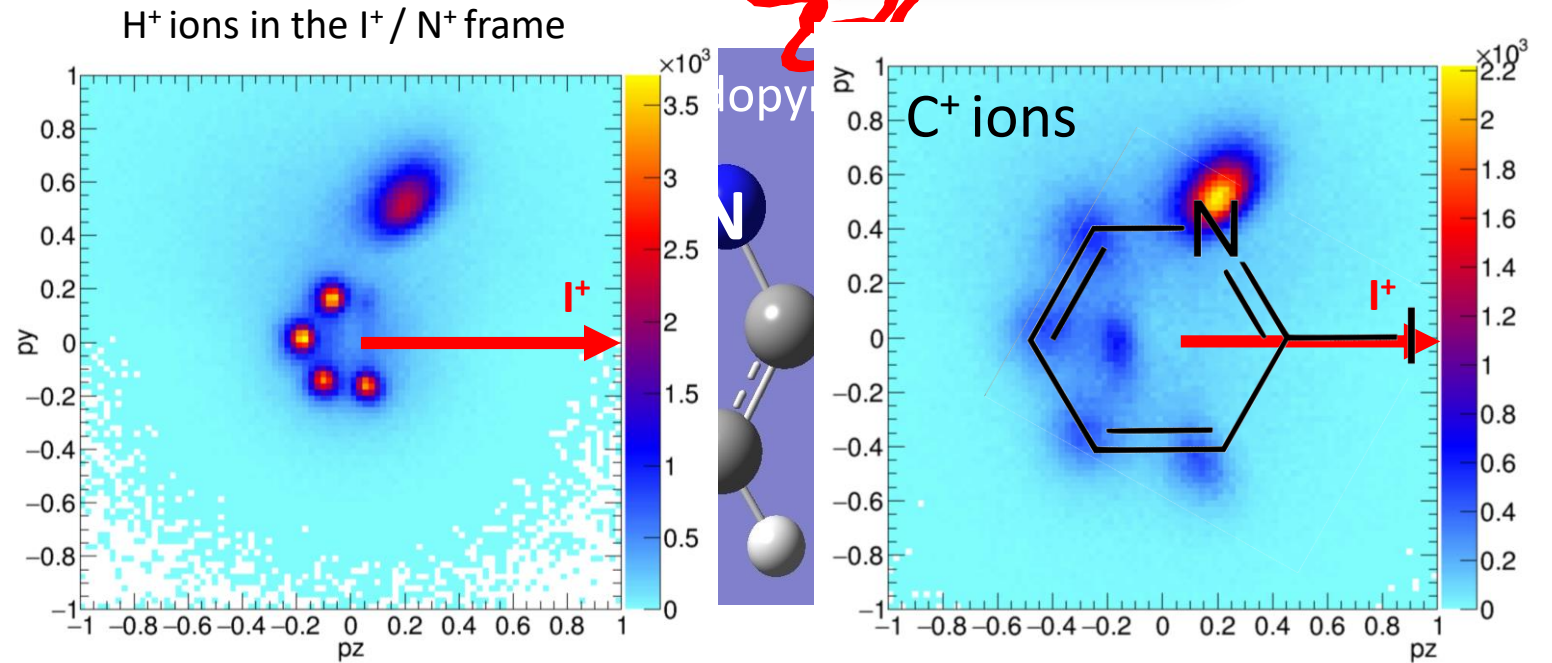
Coulomb Explosion Imaging: Frontiers

Frontier in Dimensions



Chiral Molecules
Pitzer et al.
Science, 341 (2013) 1096

Frontier in size
(number of atoms)



Experiment: Boll, Jahnke, ...
REMI @SQS EuXFEL (2019)

$$\Psi(\vec{R}_i, \vec{r}_j, t)$$

Imaging Molecules (nuclei) :

- Diffraction
- Coulomb Explosion Imaging

Imaging Molecules (electrons) :

- Image Electrons
- Real Space
- Momentum Space
- Correlations

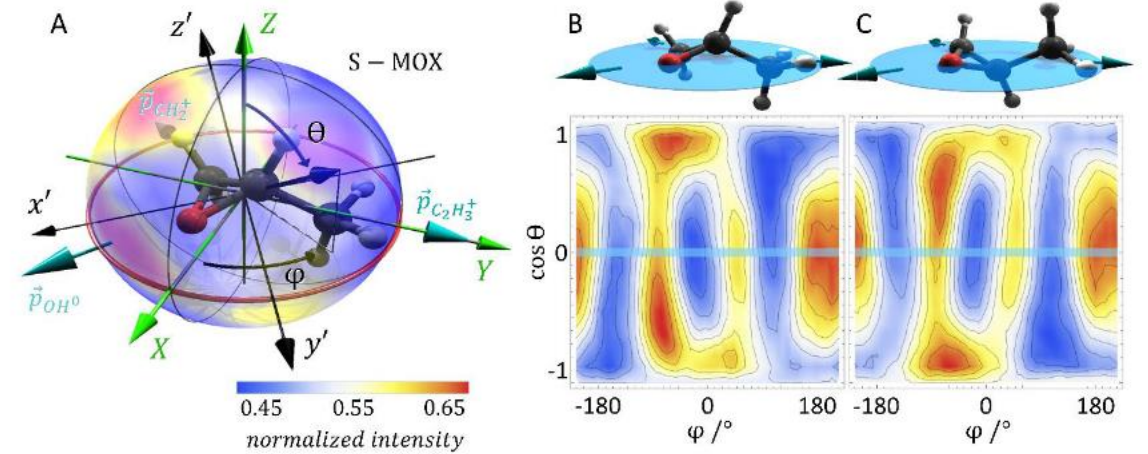
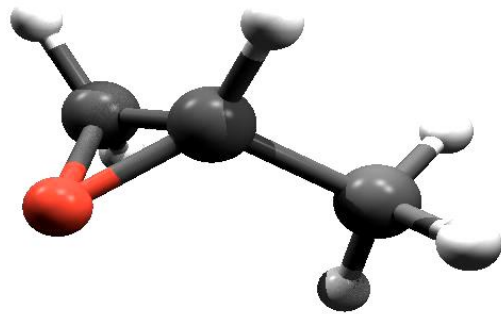


Photoelectron Diffraction: „Molecules Illuminated from Within“ (Landers et al PRL 87 (2001) 013002)

[Fehre et al. arXiv:2101.03375](https://arxiv.org/abs/2101.03375)

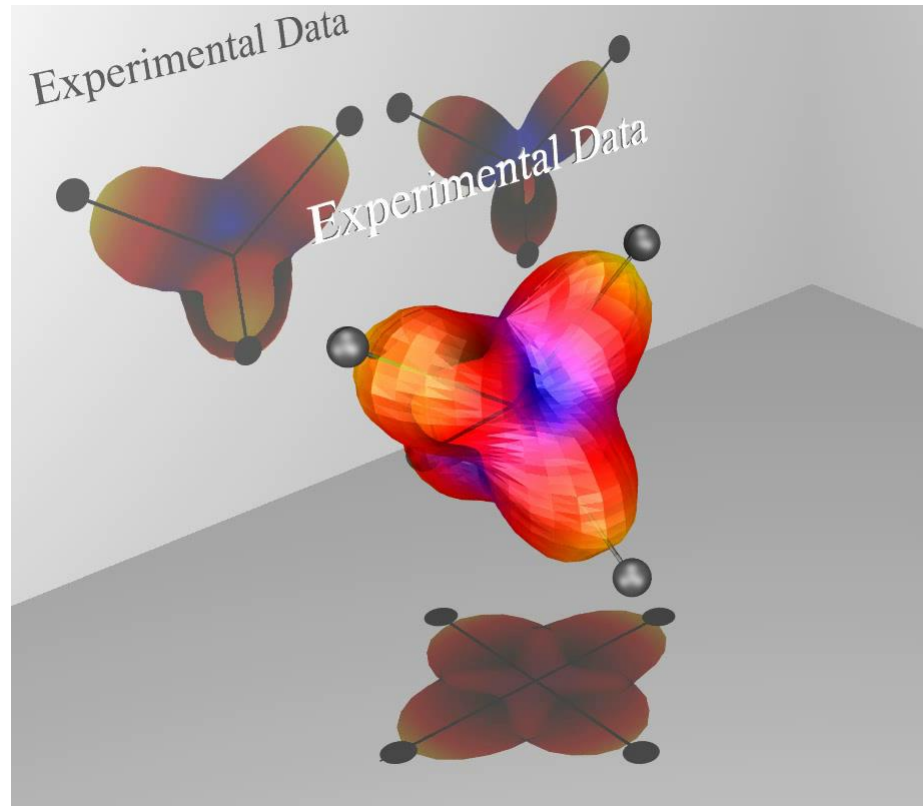
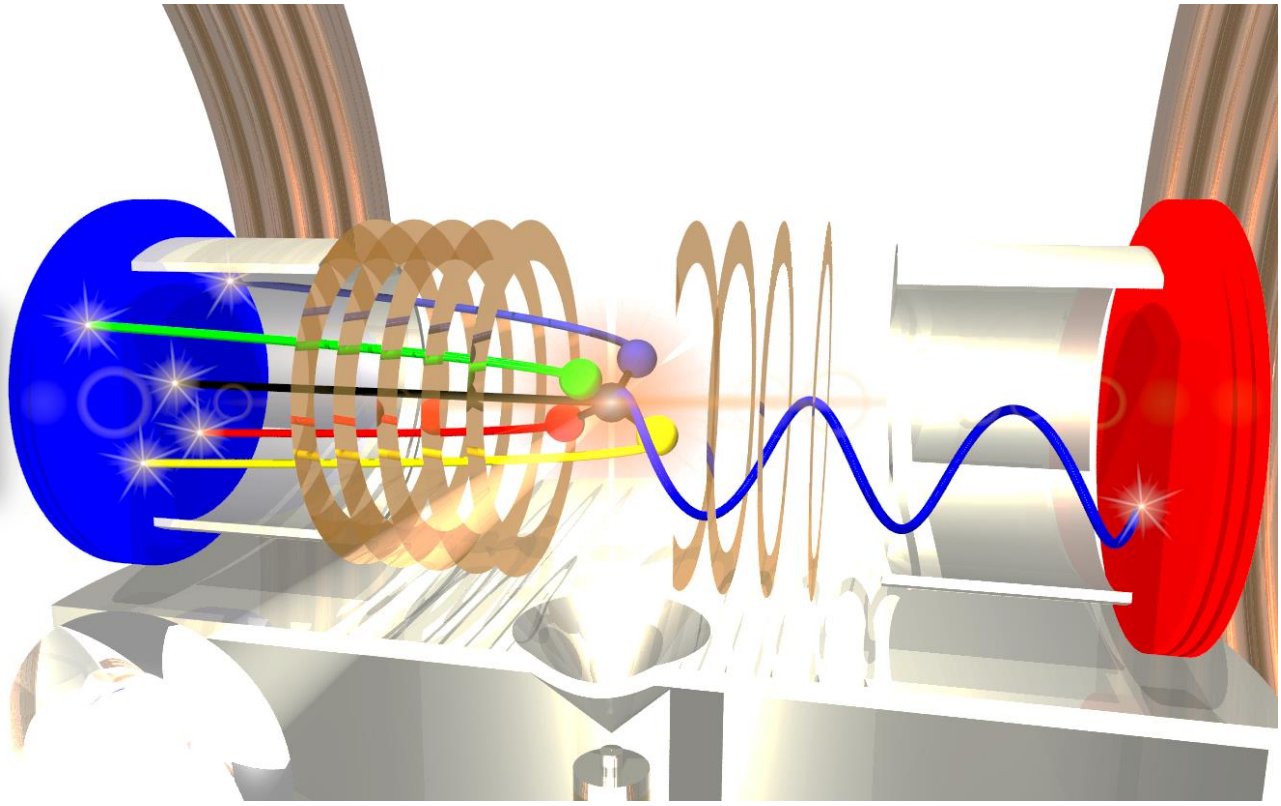
Experiment: Methoxyrane

- Enantio selective
- Image also protons
- Bond length accuracy 5%

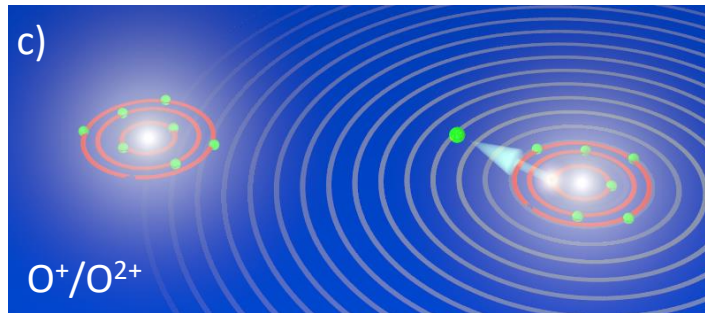
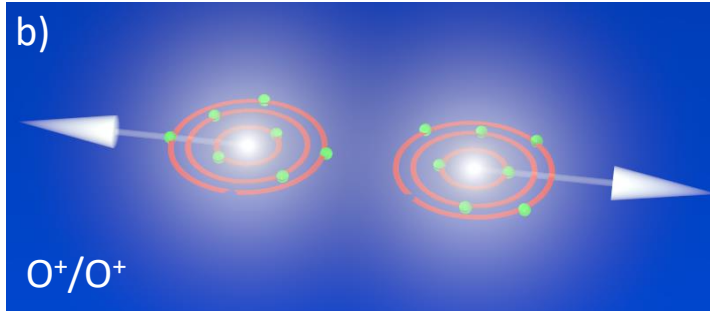
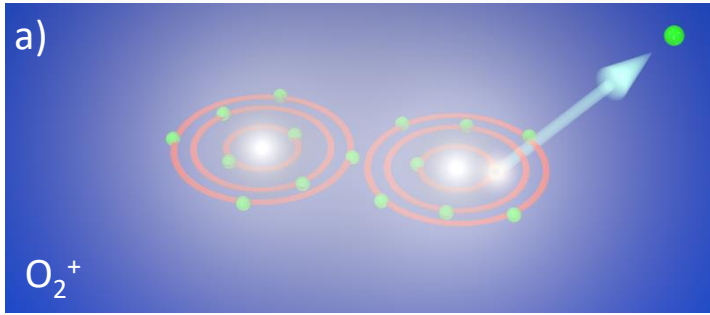


Photoelectron Diffraction: „Molecules Illuminated from Within“ (Landers et al PRL 87 (2001) 013002)

Select
molecular
orientation

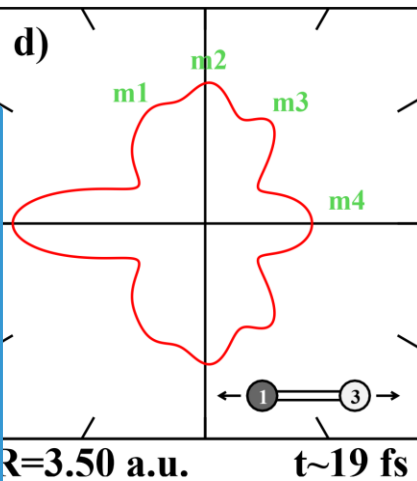
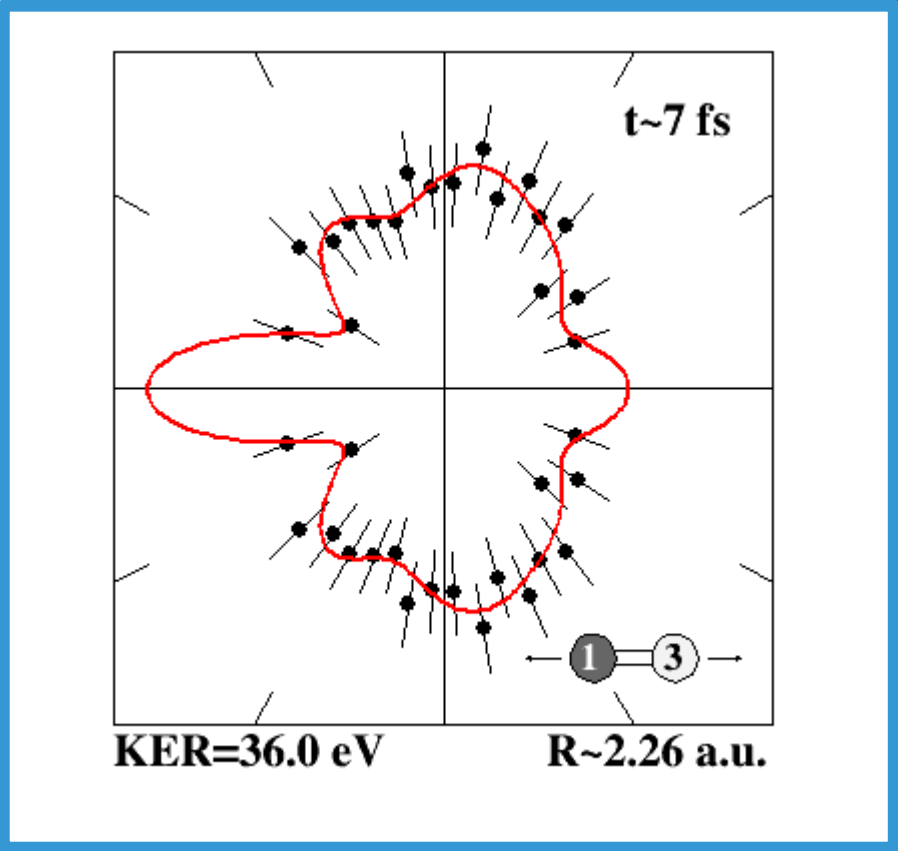
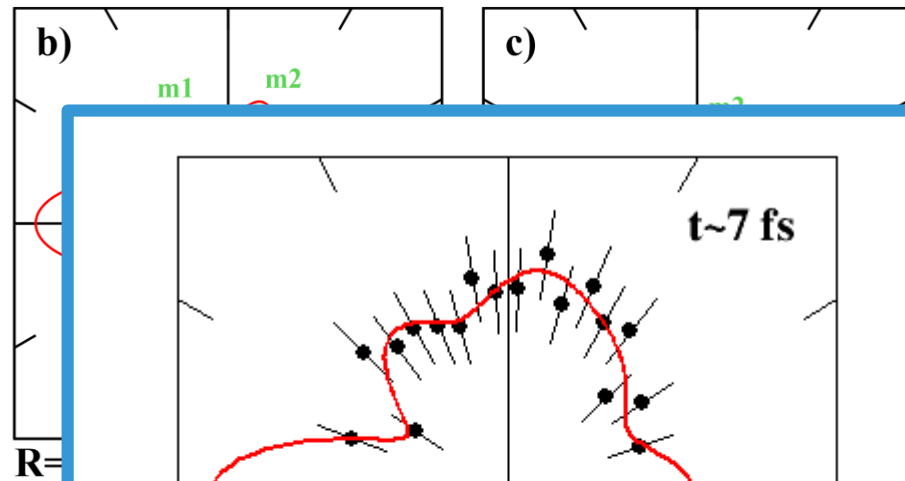
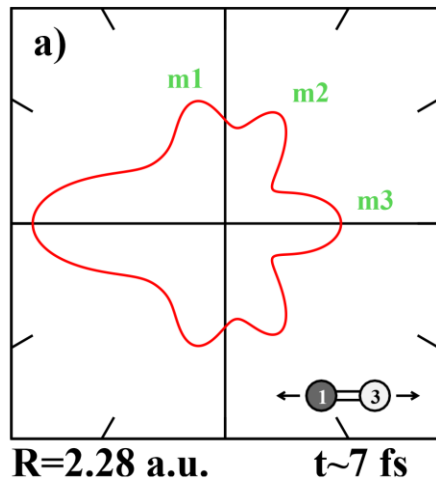


Photoelectron Diffraction: „Molecules Illuminated from Within“ (Landers et al PRL 87 (2001) 013002)

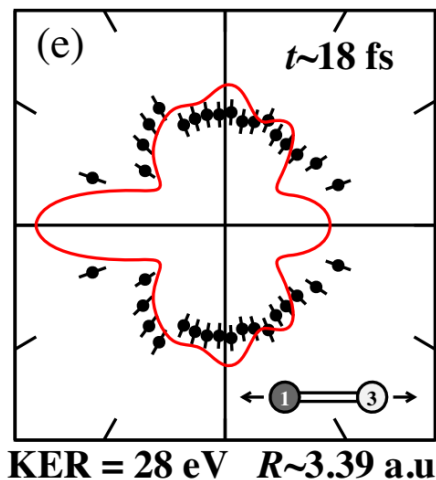
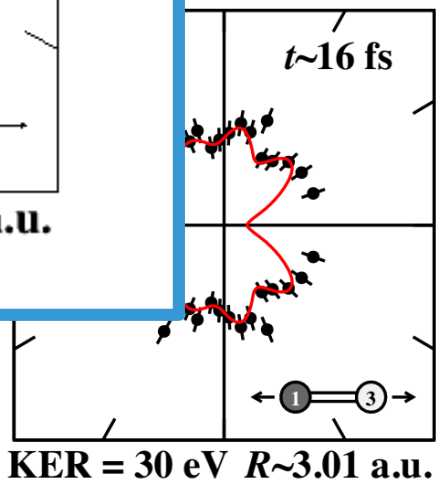
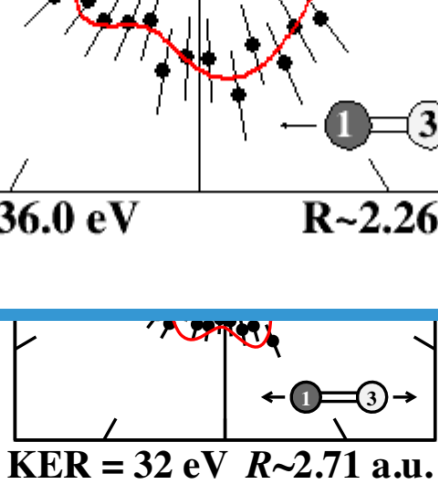
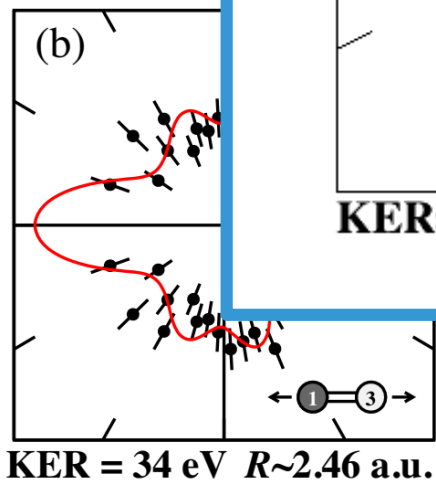
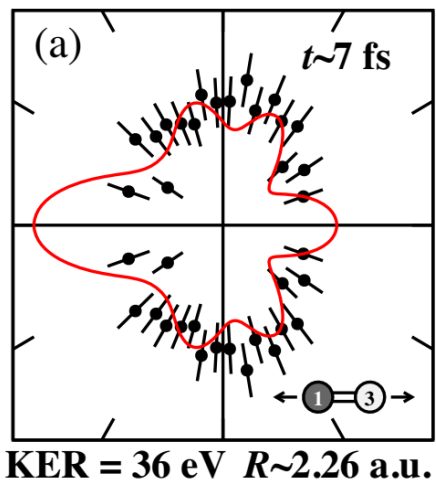


COLTRIMS Reaction Microscope @SQS
EuXFEL
Courtesy Till Jahnke

G. Kastirke et al., PRX 10, 021052 (2020)



Theory:
Ph. Demekhin,
Kassel



G. Kastirke et al., PRX 10, 021052 (2020)

Examining electron dynamics in real time at SQS!

Wolfram Helm
Till Jahnke
Markus Ilchen



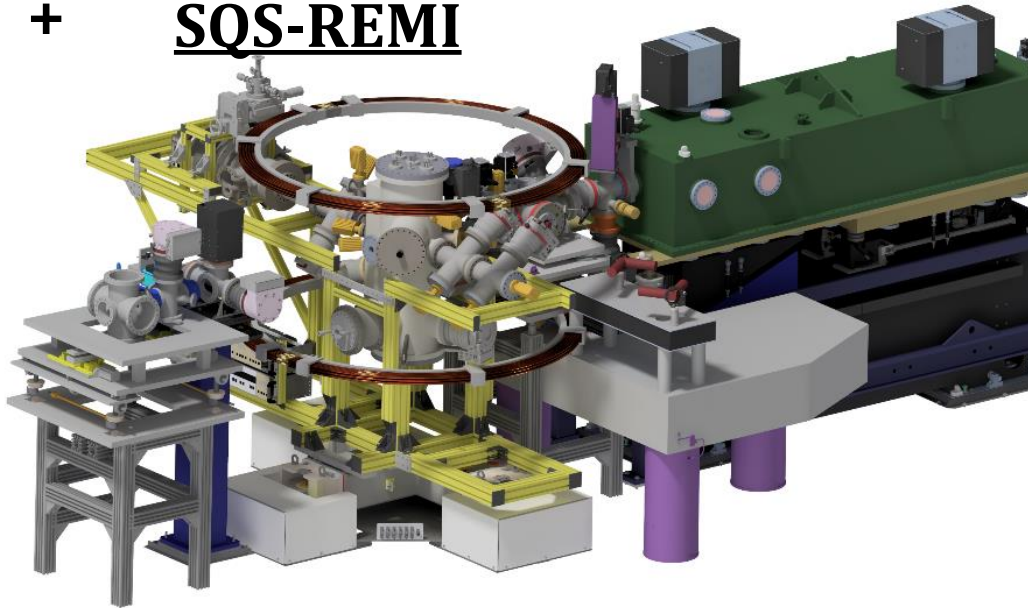
tu technische universität dortmund



Atto-Streaking

+

SQS-REMI

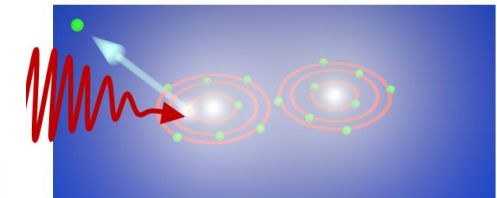
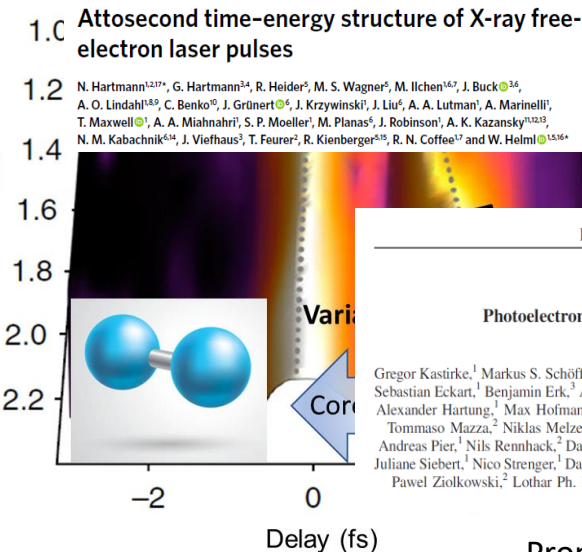


Technical Background

- Simultaneously capturing electrons and ions in coincidence with COLTRIMS in COMBINATION with non-invasive pulse diagnostics.
- Two-spike lasing operation for attosecond X-ray pump X-ray probe (few-fs pulses sorted by angular streaking).

Intra-SASE Pump-Probe Scheme

nature photonics ARTICLES
<https://doi.org/10.1038/s41566-018-0107-6>



PHYSICAL REVIEW X 10, 021052 (2020)

Photoelectron Diffraction Imaging of a Molecular Breakup Using an X-Ray Free-Electron Laser

Gregor Kastirke,¹ Markus S. Schöffler,¹ Miriam Weller,¹ Jonas Rist,¹ Rebecca Boll,² Nils Anders,¹ Thomas M. Baumann,² Sebastian Eckart,¹ Benjamin Erk,³ Alberto De Faniis,³ Kilian Fehre,¹ Averell Gattton,¹ Sven Grundmann,¹ Patrik Grychtol,² Alexander Hartung,¹ Max Hofmann,¹ Markus Ilchen,^{2,5} Christian Janke,¹ Max Kircher,¹ Maksim Kunitski,¹ Xiang Li,⁶ Tommaso Mazza,² Niklas Melzer,¹ Jacobo Montano,^{2,5} Valerija Music,^{2,5} Giammarco Nalin,¹ Yevheniy Ovcharenko,² Andreas Pier,¹ Nils Rennhack,² Daniel E. Rivas,² Reinhard Dörner,¹ Daniel Rolles,⁶ Artem Rudenko,⁶ Philipp Schmidt,^{2,5} Juliane Siebert,¹ Nico Strenger,¹ Daniel Trabert,¹ Isabel Vela-Perez,¹ Rene Wagner,² Thorsten Weber,⁷ Joshua B. Williams,⁸ Pawel Ziolkowski,² Lothar Ph. H. Schmidt,¹ Achim Czasch,¹ Florian Trinter,^{3,9} Michael Meyer,² Kiyoshi Ueda,¹⁰ Philipp V. Demekhin,^{3,5} and Till Jahnke^{1,1}

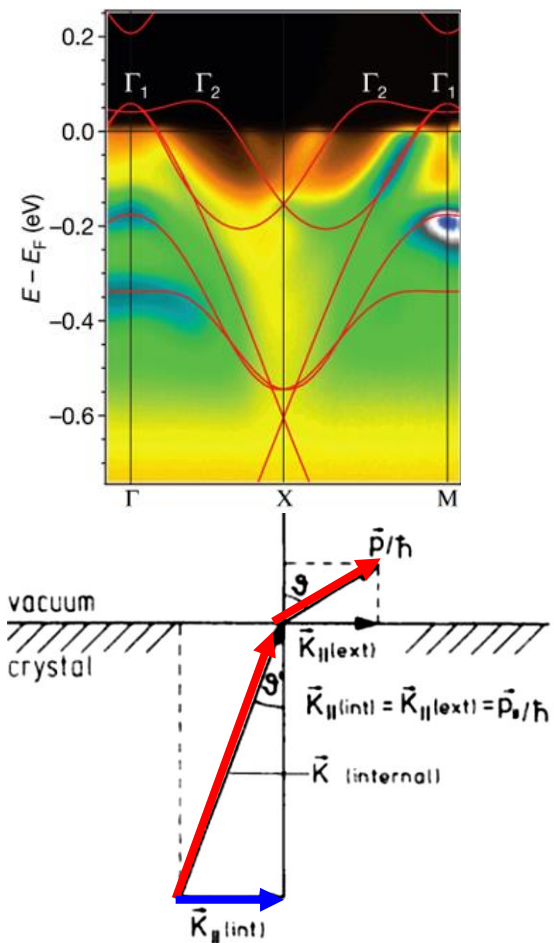
Scientific Goals

- Follow electron dynamics in real time with an intra-pulse attosecond X-ray pump X-ray probe scheme (see figure).
- Monitor the electronic evolution during structural molecular changes site-specifically with sub-fs resolution.
- Correlate molecular structure and function during their ultrafast time evolution at their very origin, close to and entering the attosecond regime.

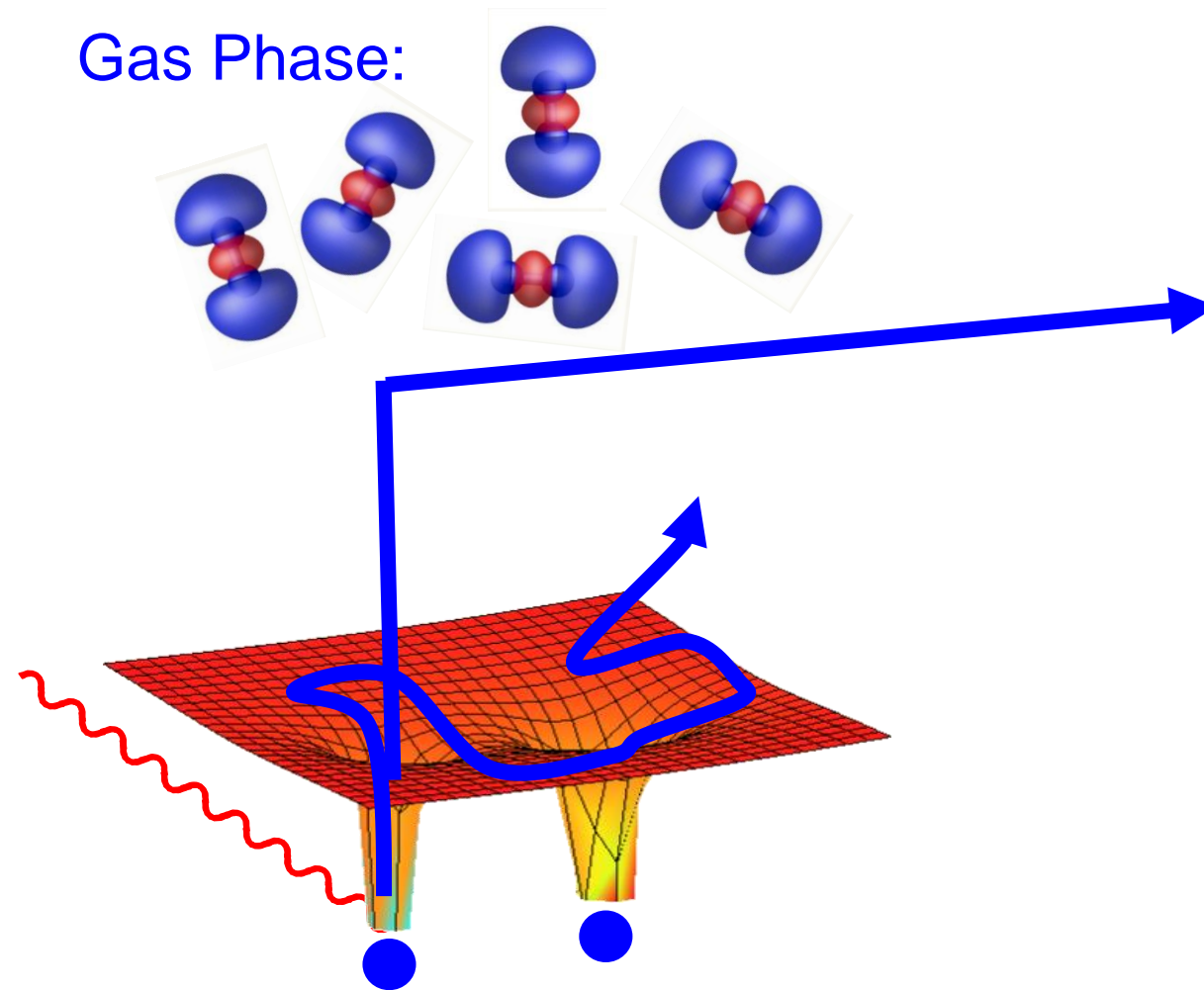
Photoelectron diffraction versus ARPES in molecules

$$\Psi(\vec{R}_i, \vec{r}_j, t)$$

Solid Phase:

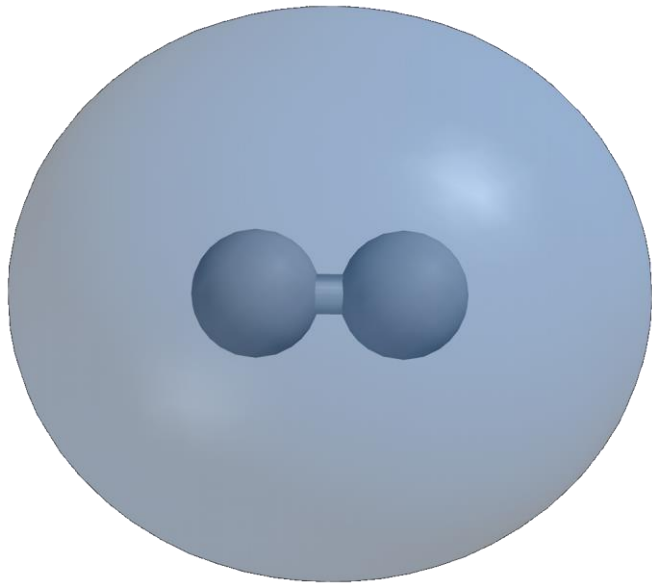


Gas Phase:

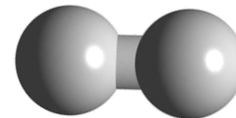
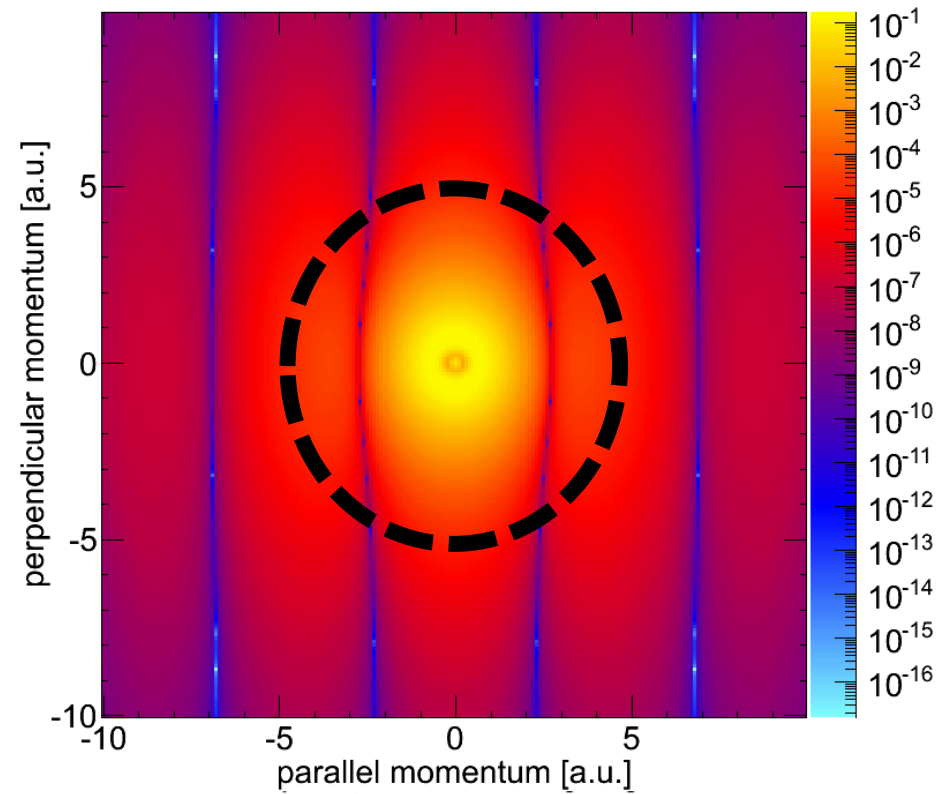


Correlations: H_2 wavefunction

Coordinate Space

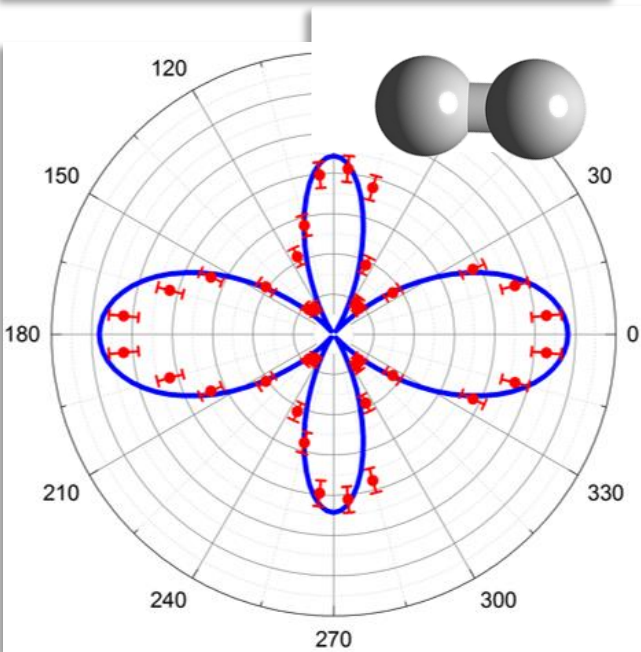
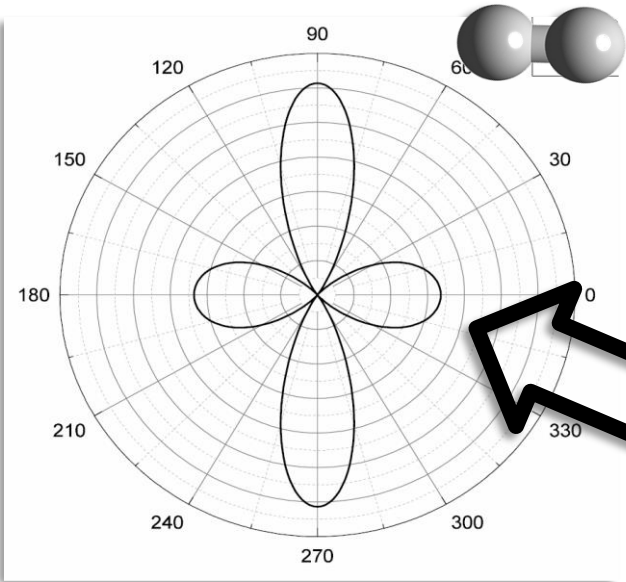


Momentum Space

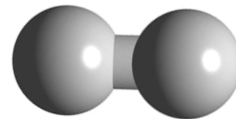
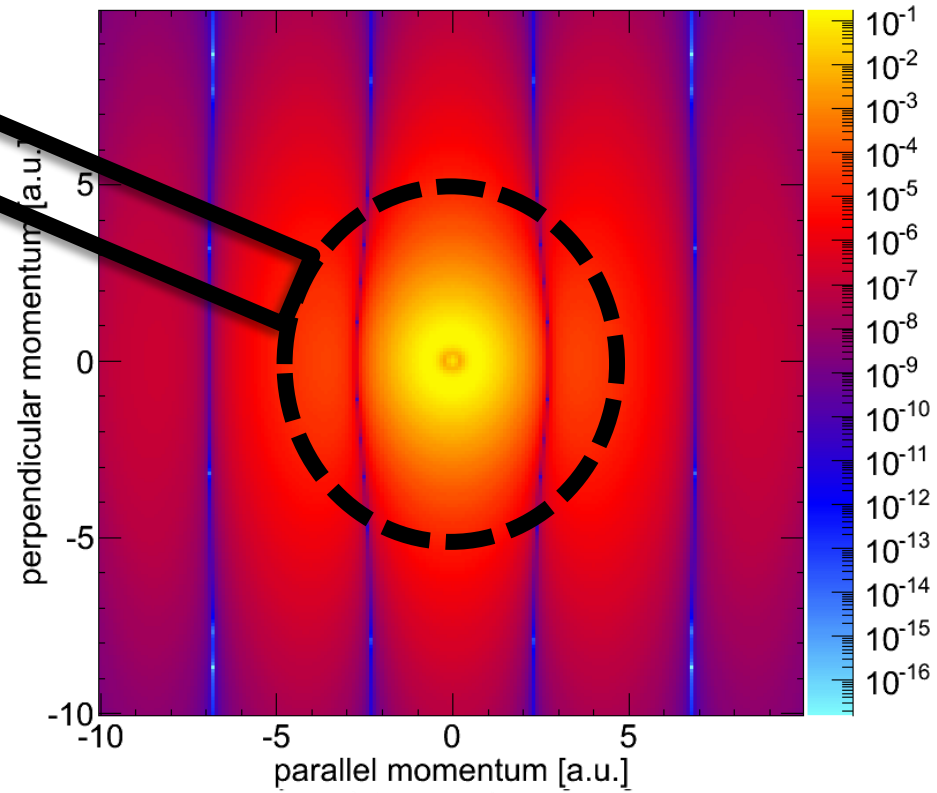


Collaboration:
Fernando Martin
Waitz et al.
Nat. Commun., 8 (2017) 2266

Correlations: H₂ wavefunction



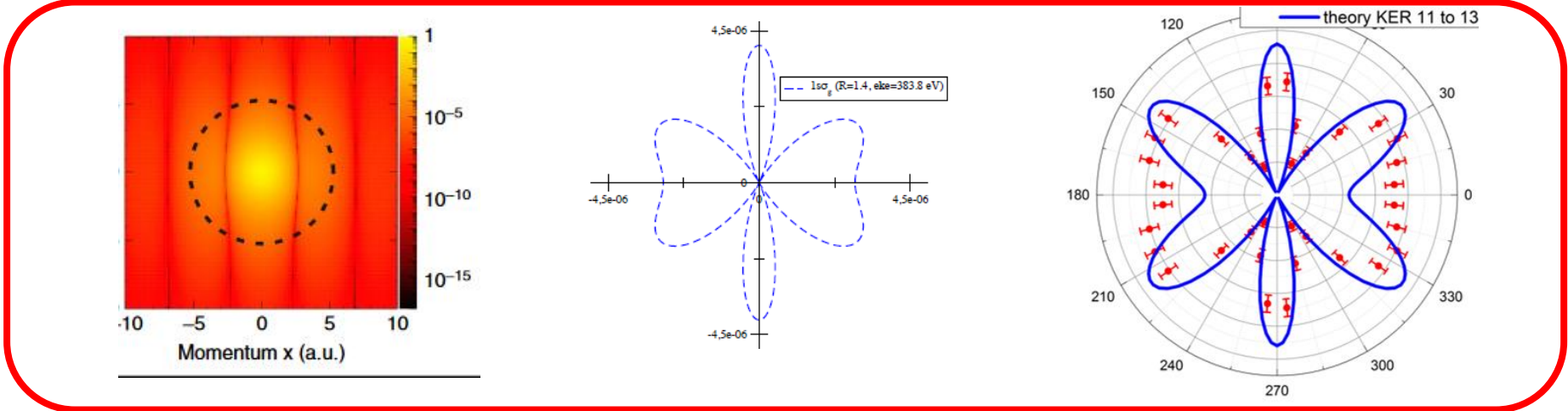
Momentum Space



$$\Psi_0^{H_2}(k_A, k_B) =$$

$$A |\varphi^{HF}(1s\sigma_g)\rangle |\varphi^{HF}(1s\sigma_g)\rangle +$$

Hartree Fock wavefunction



Hartree Fock wavefunction

$$\Psi(\vec{R}_i, \vec{r}_j, t)$$

At the horizon

2 Photons (<50as), 2 electrons
Movies of correlated wavefunctions

- Cookibox tagging
- >10kHz rep rate
- <50as spikes
- Smallest possible focus (<1 μm , >1 $\mu\text{J}/50\text{as}$ spike)
- COLTRIMS Reaction Microscope

Frankfurt:

Markus Schöffler

Martin Pitzer

Markus Waitz

Sven Grundmann

Isabell Vela Perez

Giammarco Nalin

Lothar Schmidt

Jörg Voigtsberger

Maksim Kunitski

Stefan Zeller

Jonas Rist

Gregor Kastirke

Miriam Weller

Alexander Hartung

Daniel Trabert

Sebastian Eckart

R.D.



LBNL:



Thorsten Weber

Madrid

Fernando Martin

Alicia Palacios



Canberra

Anatoli Kheifets



Kassel:

Philipp Demekhin



Reno:

Joshua Williams

University of Oklahoma:

Dörte Blume

Qingze Guan



EuXFEL:

Till Jahnke

Michael Meyer

Rebecca Boll



UPMC

Marc Simon

Maria Novella Piancastelli

Sendai:

Kiyoshi Ueda

Petra 3, Hamburg

Jens Viefhaus

Florian Trinter