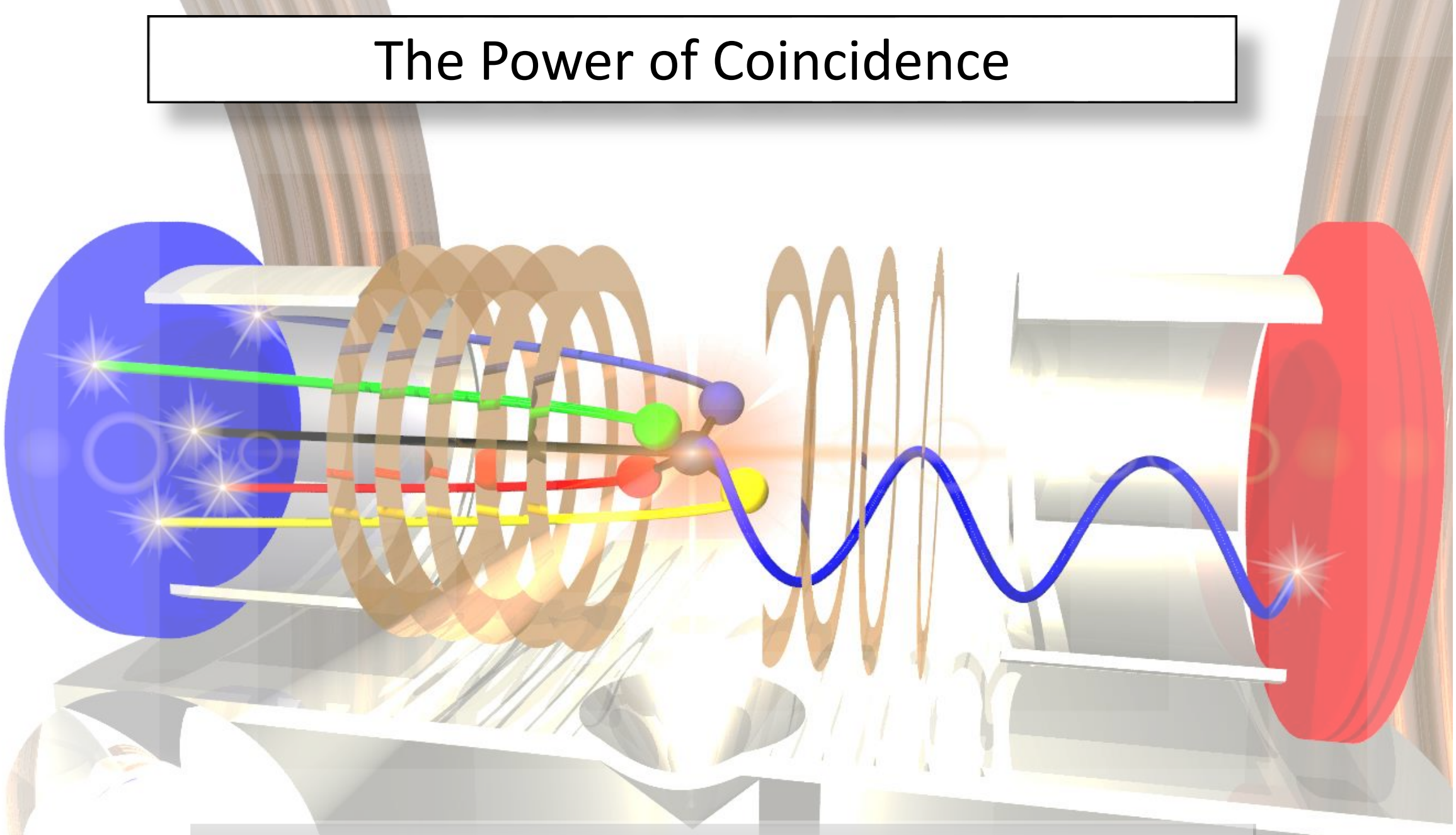
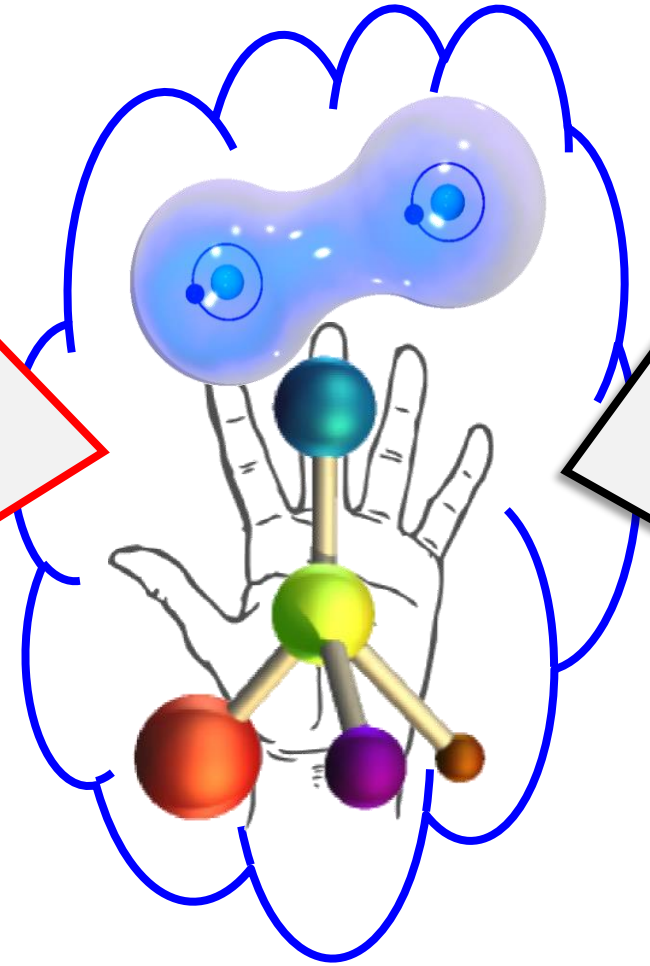
The Power of Coincidence



Reinhard Dörner, Goethe University Frankfurt

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



Imaging Molecules (nuclei) :

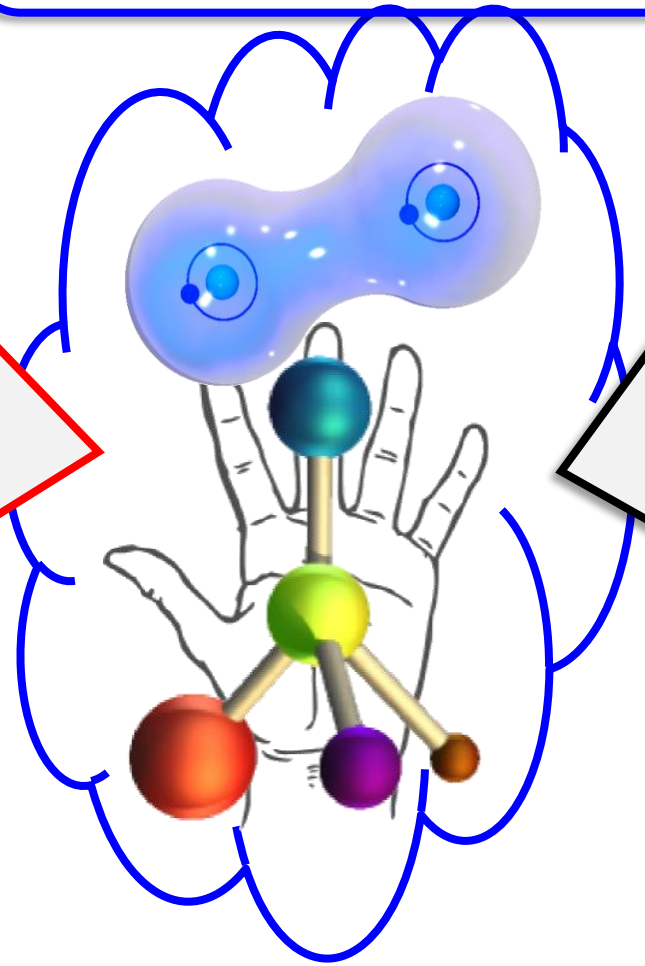
- Diffraction
- Coulomb Explosion Imaging

Imaging Molecules (electrons) :

- Image Electrons
- Momentum Space
- Correlations

Coincidence Imaging

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



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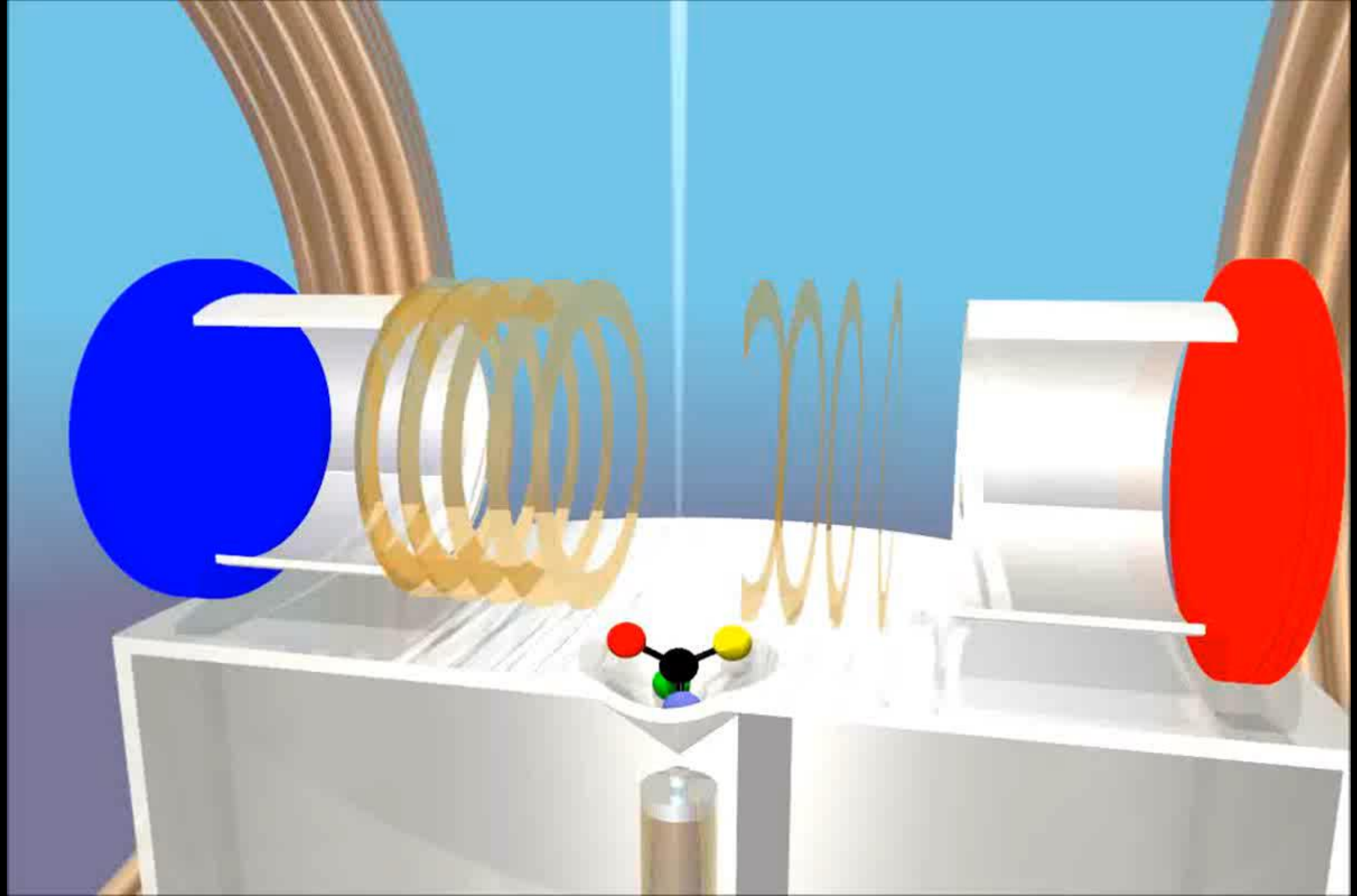


Coincidence Imaging

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

Imaging Molecules (nuclei) :

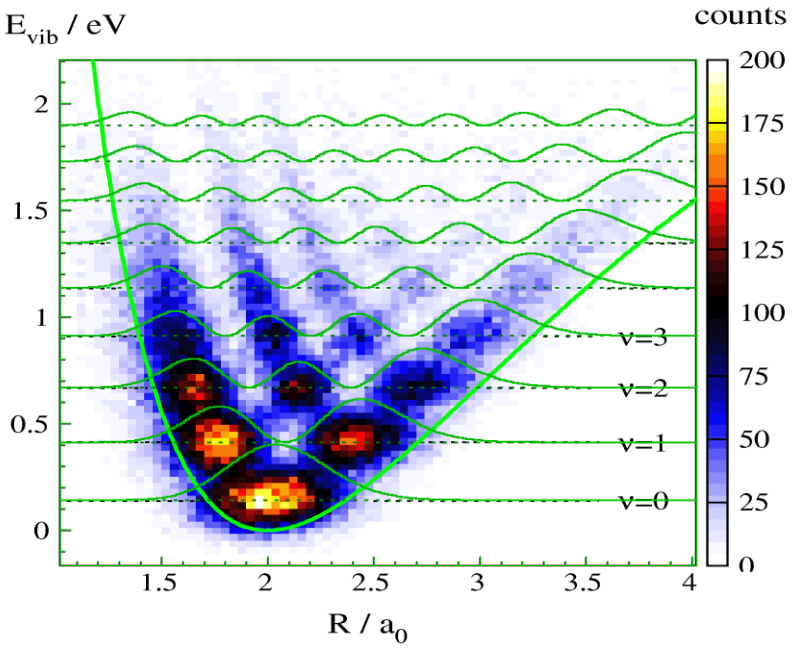
- Diffraction
- Coulomb Explosion Imaging

Imaging Molecules (electrons) :

- Image Electrons
- Real Space
- Momentum Space
- Correlations

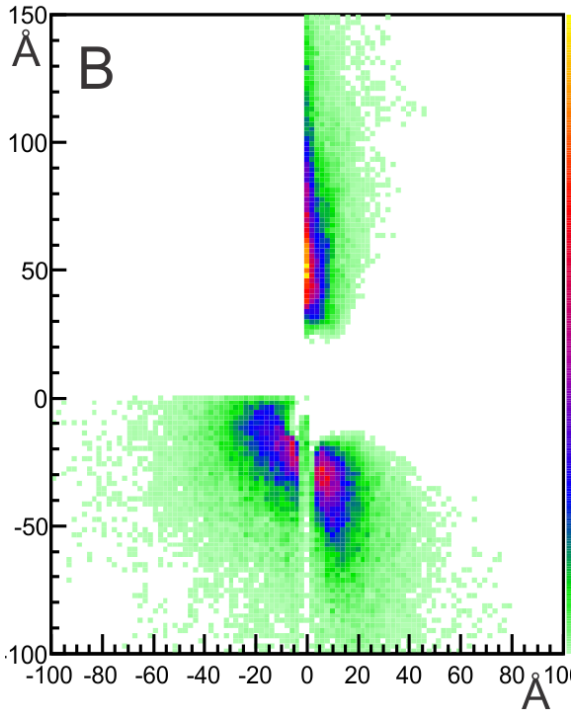
Coulomb Explosion Imaging: Frontiers

Frontier in Precision



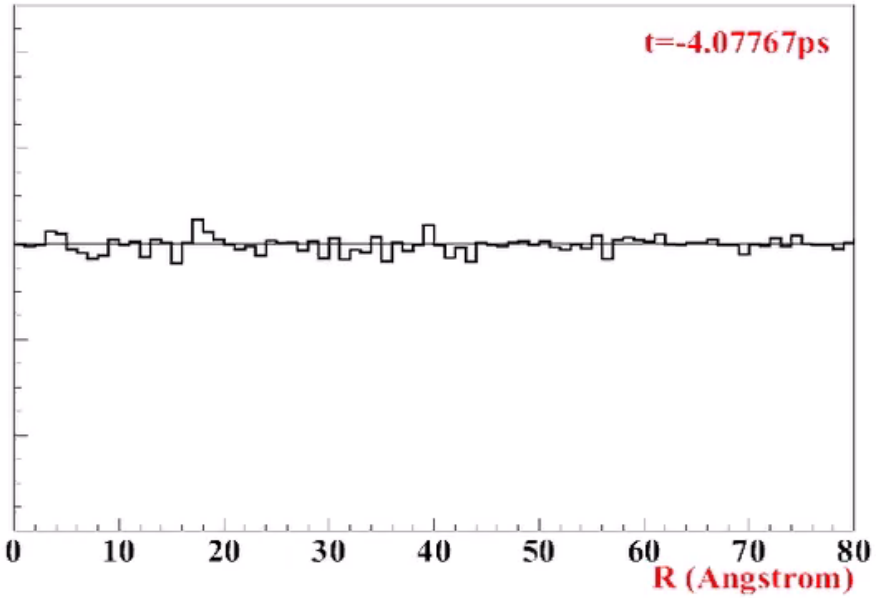
„Spatial Imaging at the quantum limit“
L. Schmidt et al
PRL 108, 73202 (2012)

Frontier in size (spatial extension)



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

Frontier in size – time domaine (spatial extension)

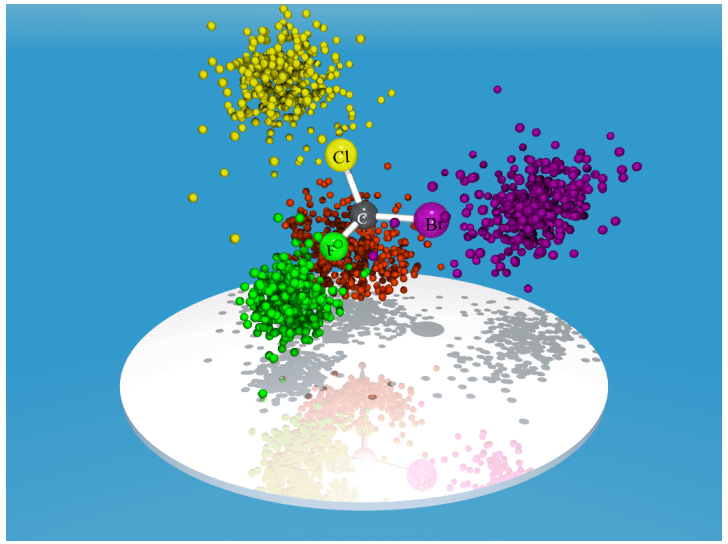


Kunitiski et al.
Nat. Phys. **17,** 174–178 (2021)

Coulomb Explosion Imaging: Frontiers

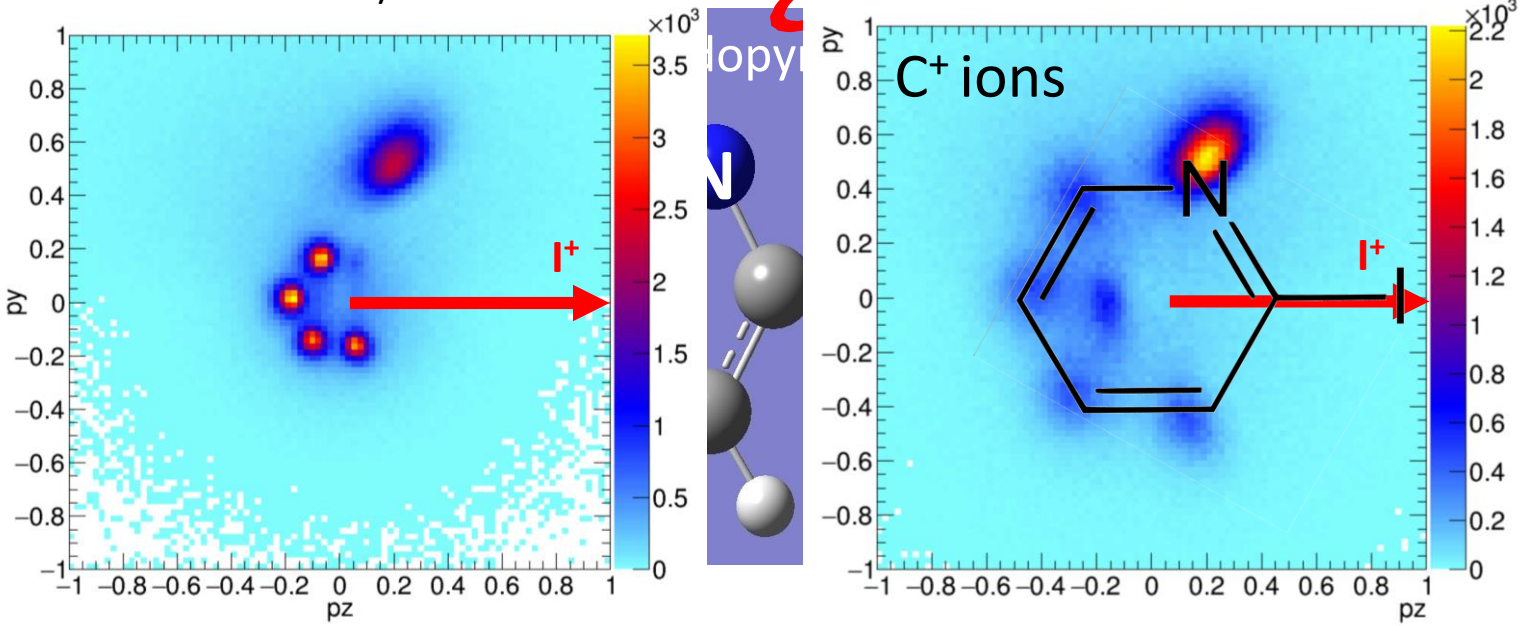
Frontier in Dimensions

Frontier in size
(number of atoms)



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

H^+ ions in the I^+ / N^+ frame



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

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

Imaging Molecules (nuclei) :

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Imaging Molecules (electrons) :

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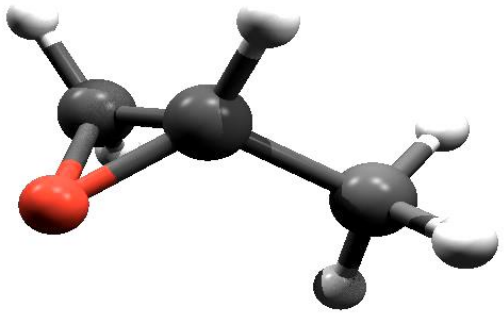
- Real Space
- Momentum Space
- Correlations

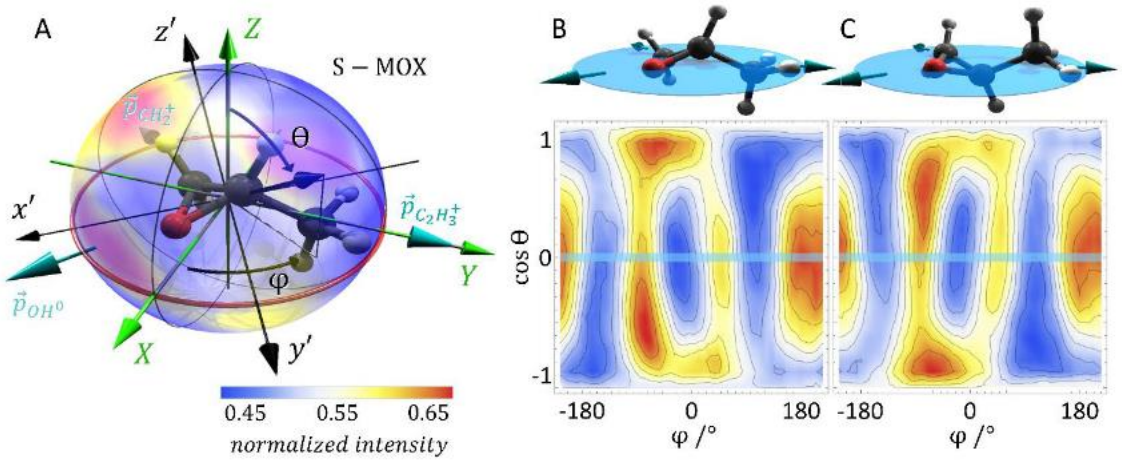
Photoelecton Diffraction: „Molecules Illuminated from Within“ (Landers et al PRL 87 (2001) 013002



Fehre et al. arXiv:2101.03375

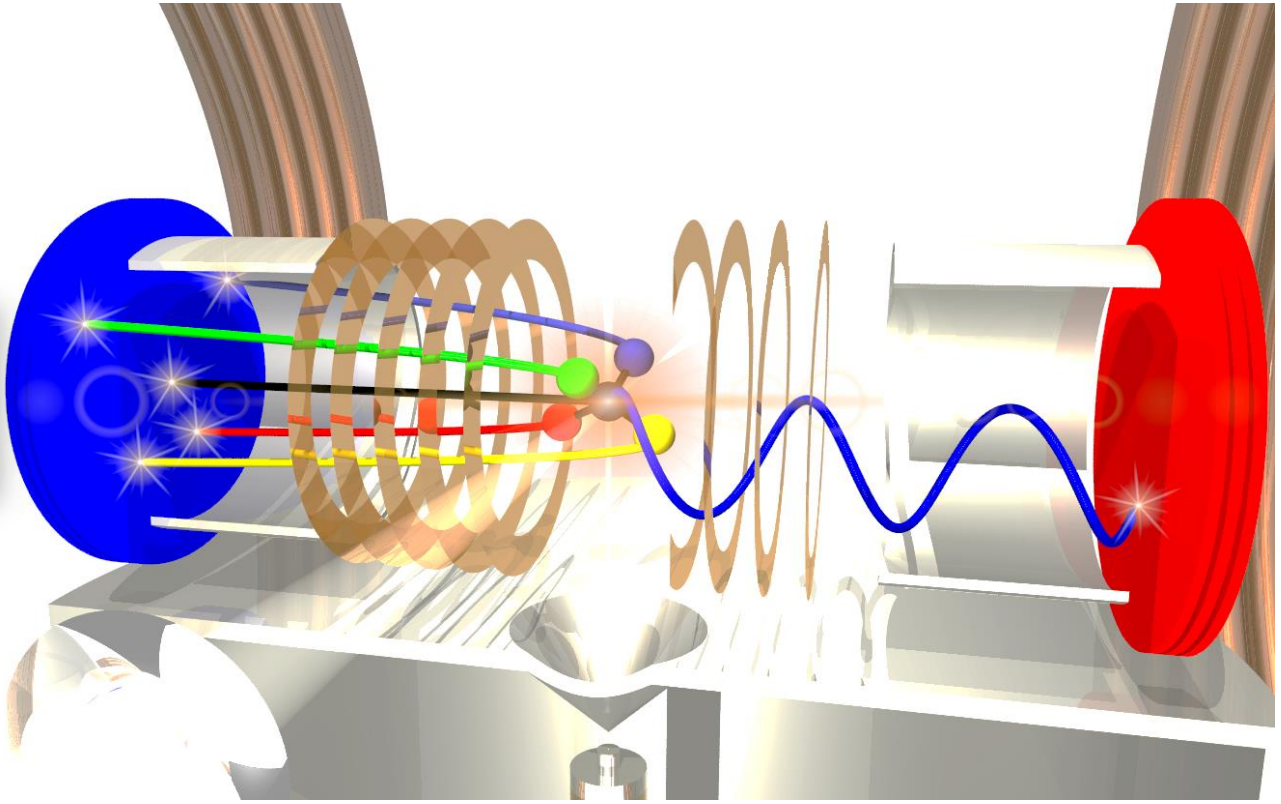
Experiment: Methyloxyrane

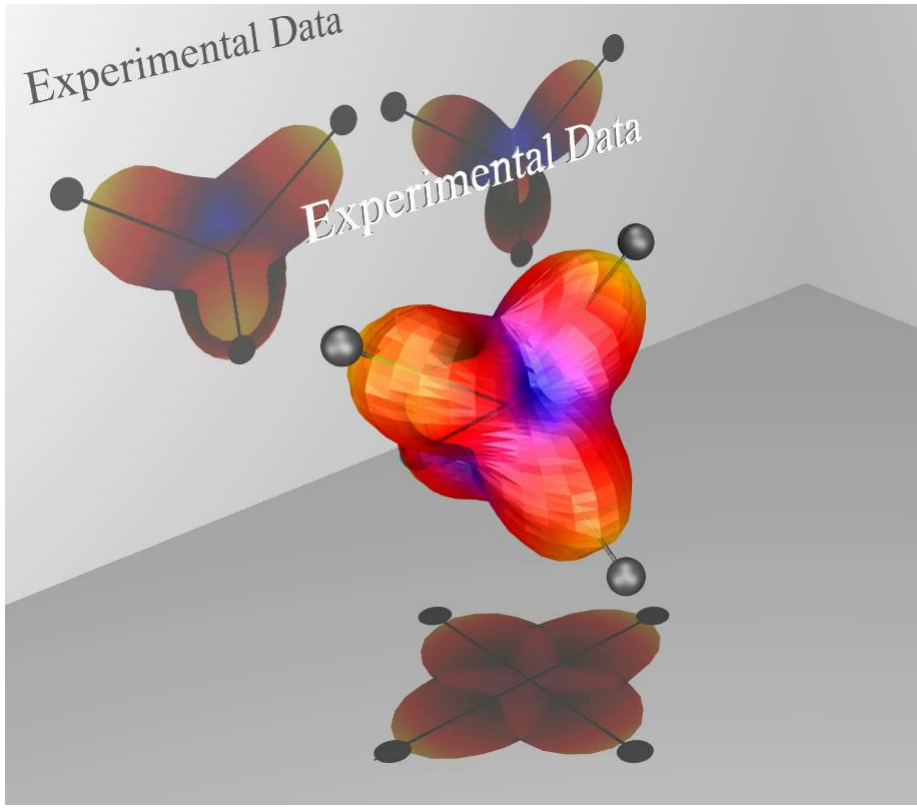
- Enantio selective
- Image also protons
- Bond length accuracy 5%



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

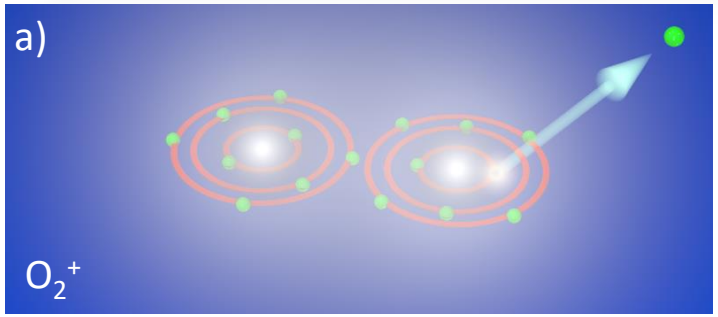
Select molecular orientation

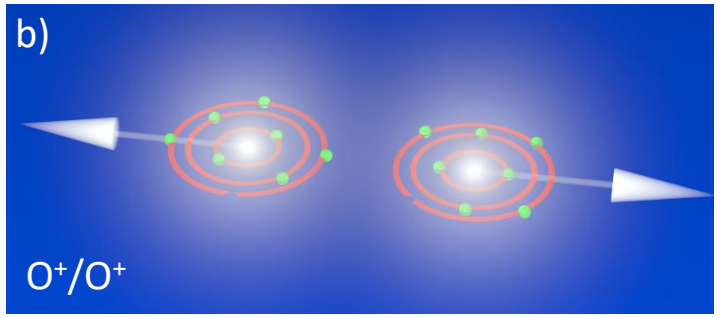


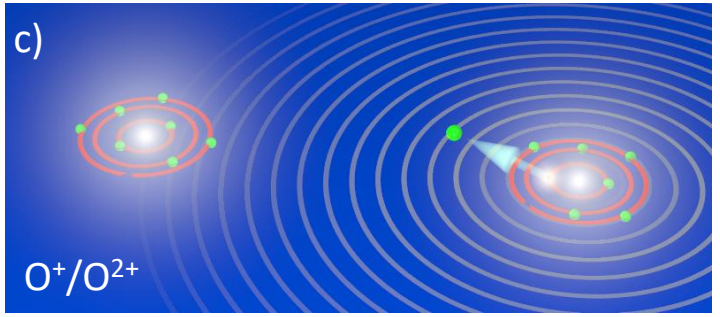


Williams et al PRL 108, 233002 (2012)

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



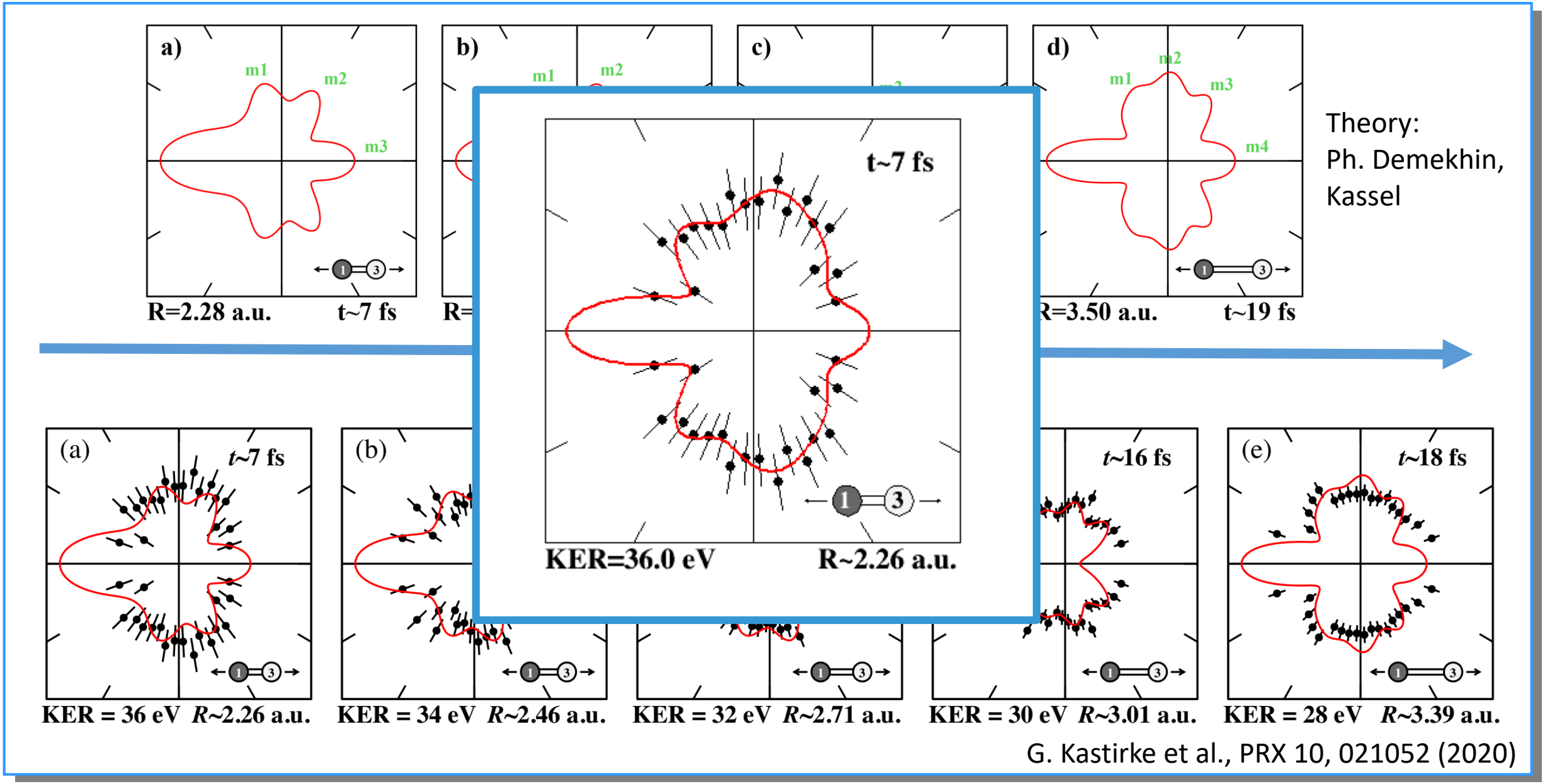




COLTRIMS Reaction Microscope @SQS EuXFEL

Courtesy Till Jahnke

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



Courtesy Till Jahnke

Examining electron dynamics in real time at SQS!

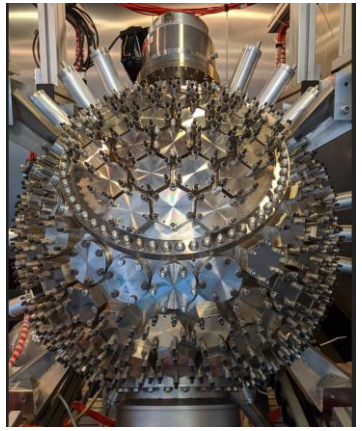
Wolfram Helm
Till Jahnke
Markus Ilchen

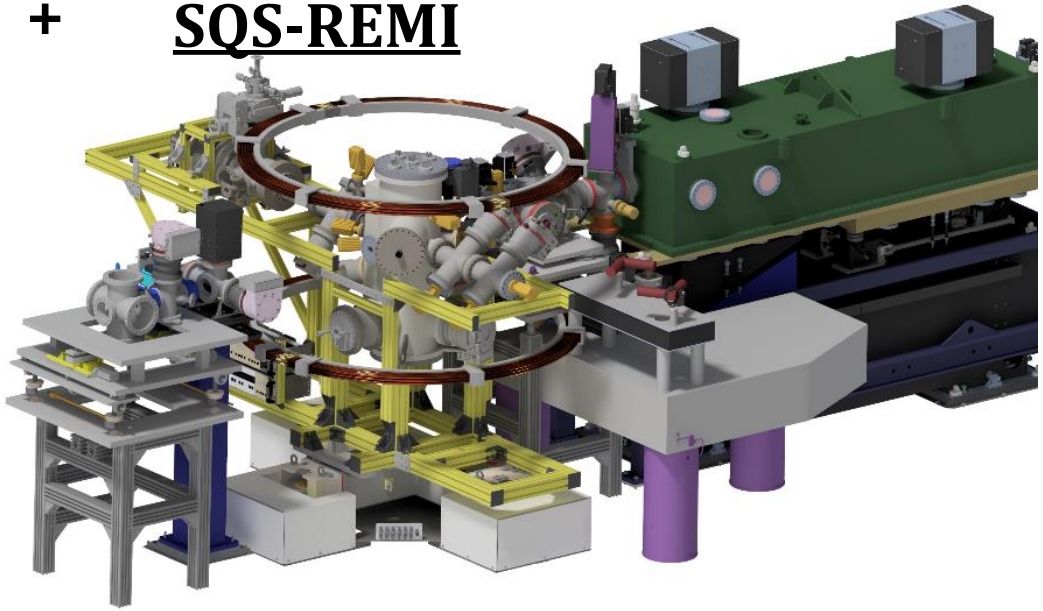
technische universität dortmund

European XFEL

DESY

Atto-Streaking + SQS-REMI

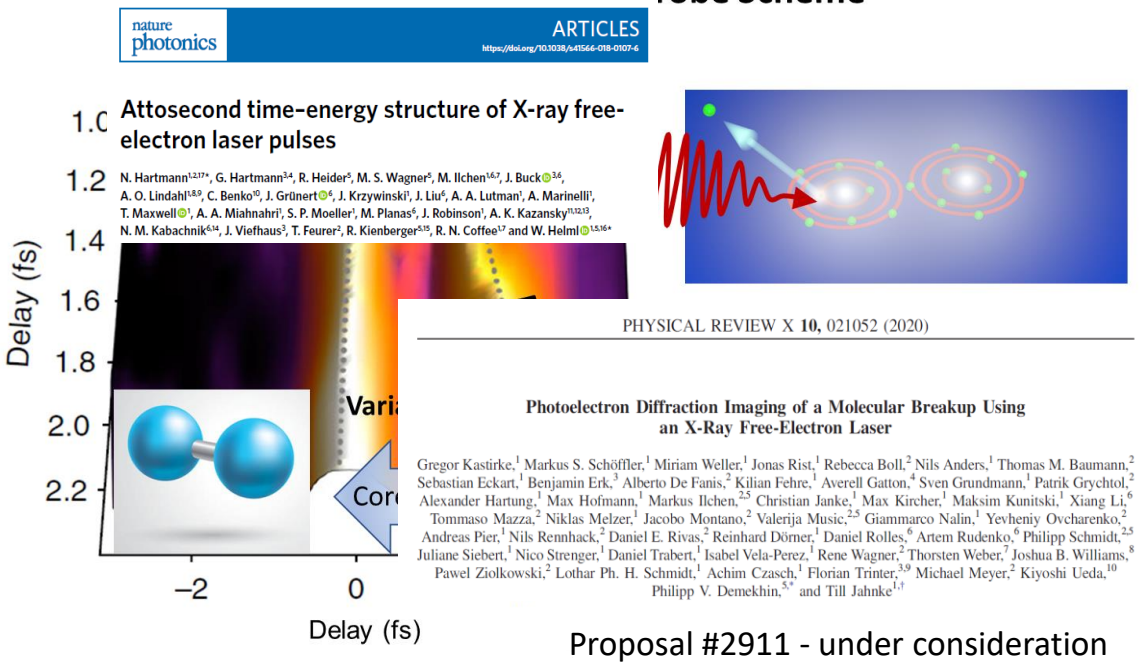




Technical Background

- **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



Scientific Goals

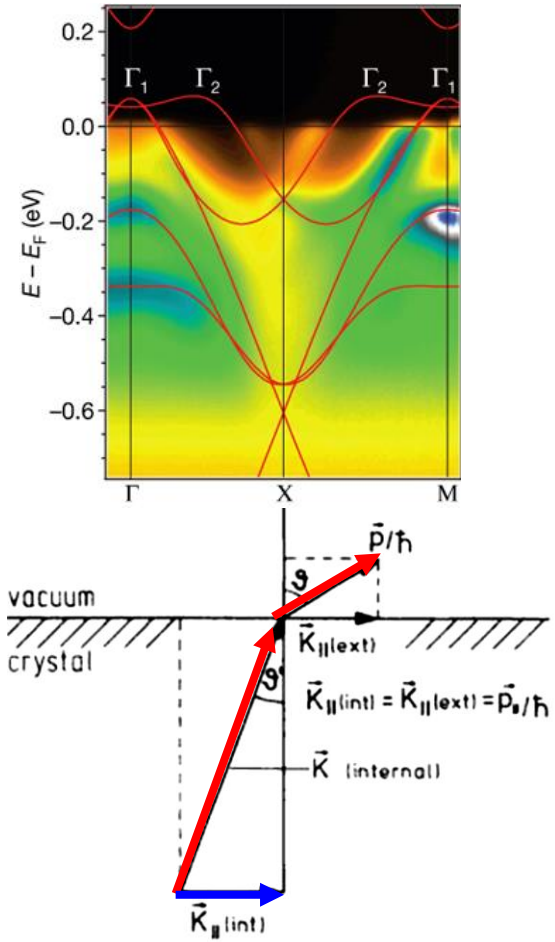
- **Follow electron dynamics in real time with an intra-pulse attosecond X-ray pump X-ray probe scheme (see figure).**
- **Monitor the electronic evolution during structural molecular changes site-specifically with sub-fs resolution.**
- **Correlate molecular structure and function during their ultrafast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

Proposal #2911 - under consideration

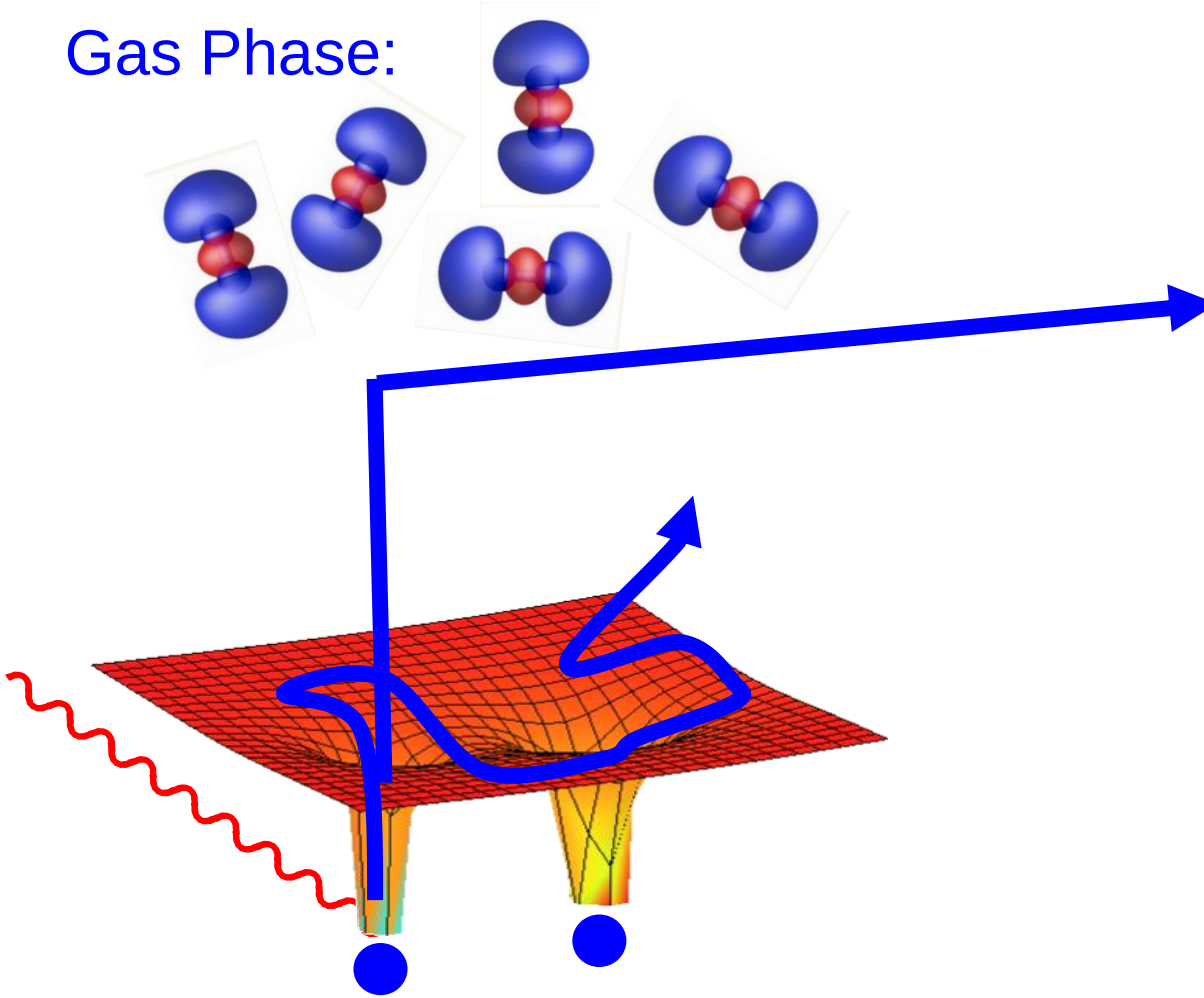
Photoelectron diffraction versus ARPES in molecules

$$\Psi\left(\overrightarrow{R_i}, \overrightarrow{r_j}, t\right)$$

Solid Phase:

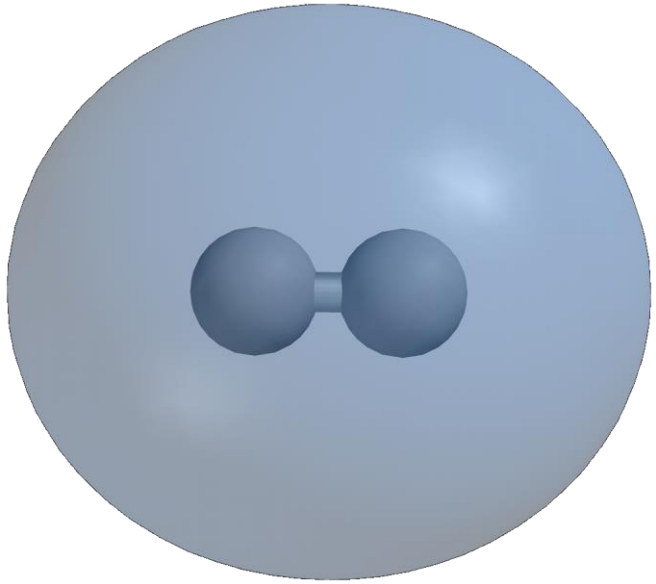


Gas Phase:

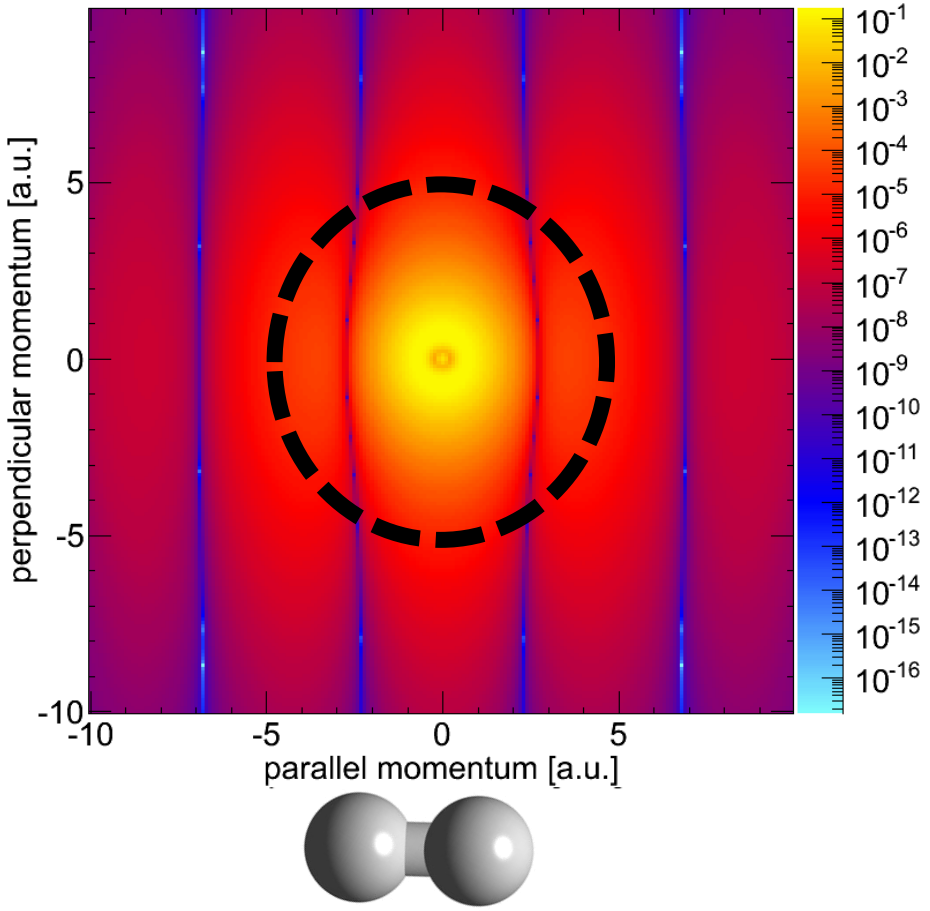


Correlations: H_2 wavefunction

Coordinate Space

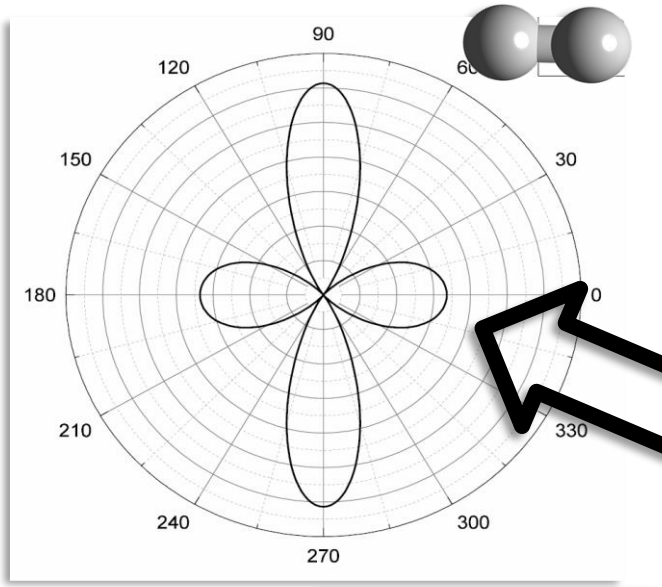


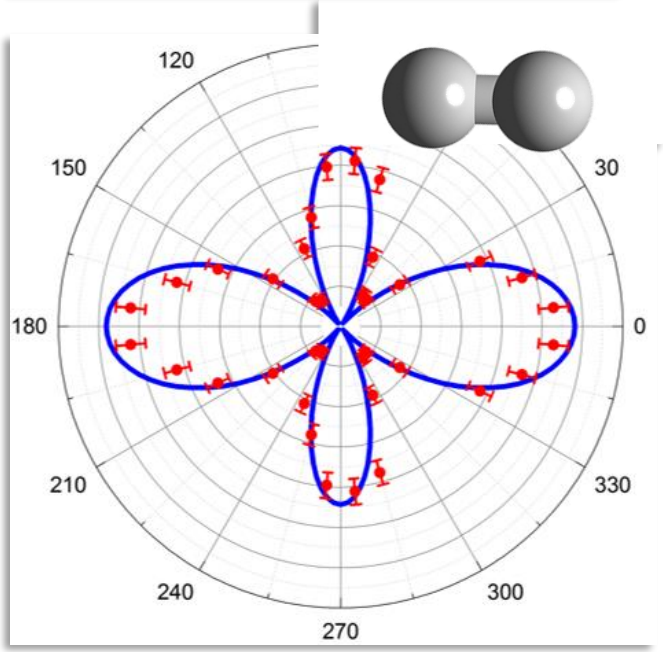
Momentum Space



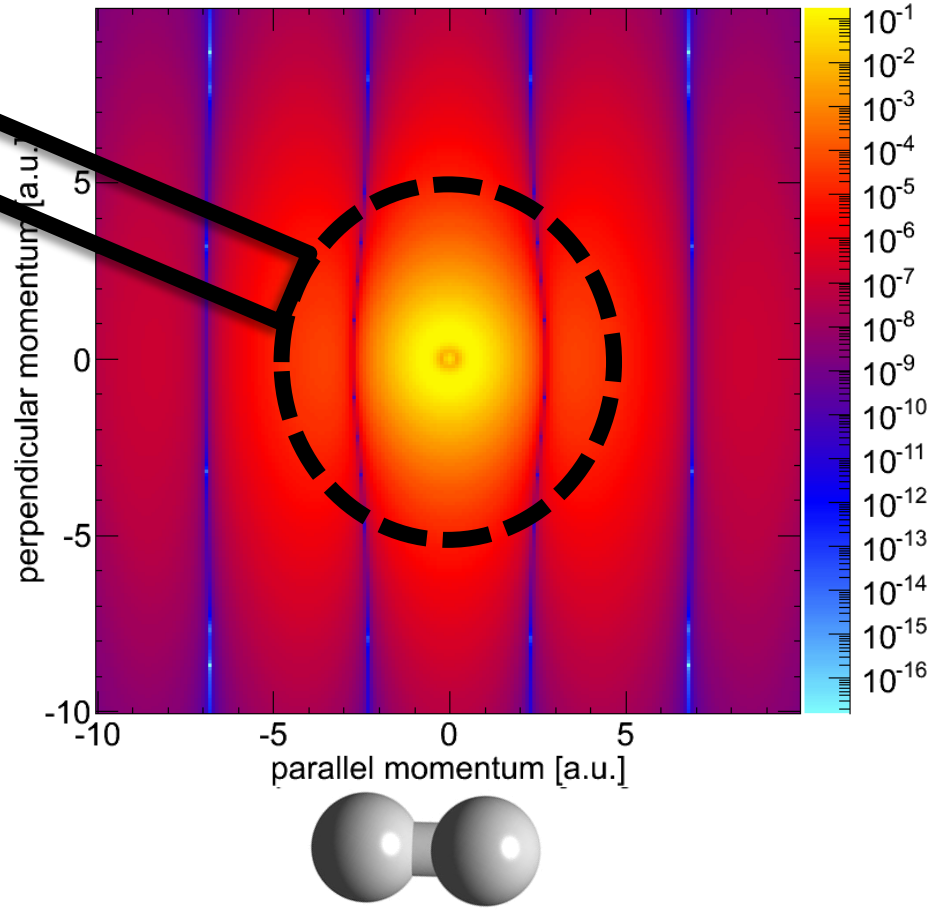
Collaboration:
Fernando Martin
Waitz et al.
Nat. Commun., 8 (2017) 2266

Correlations: H_2 wavefunction



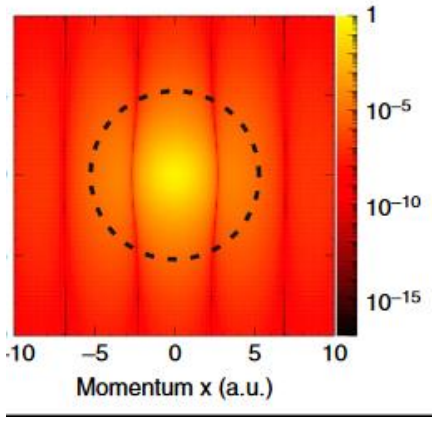


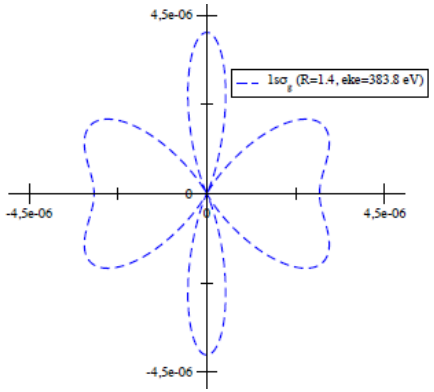
Momentum Space

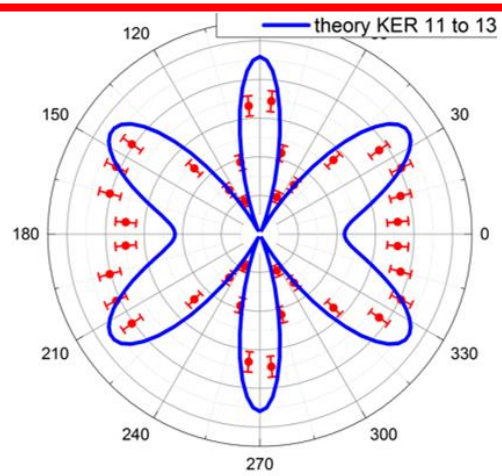


$$\Psi_0^{H_2}(k_A, k_B) = A\left|\varphi^{HF}(1s\sigma_g)\right\rangle\left|\varphi^{HF}(1s\sigma_g)\right\rangle +$$

Hartree Fock wavefunction







Hartree Fock wavefunction

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

At the horizon

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

- Cookibox tagging
- >10kHz rep rate
- <50as spikes
- Smallest possible focus (<1μm, >1 μJ/50as 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
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Kassel:

Philipp Demekhin

Reno:

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Till Jahnke
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