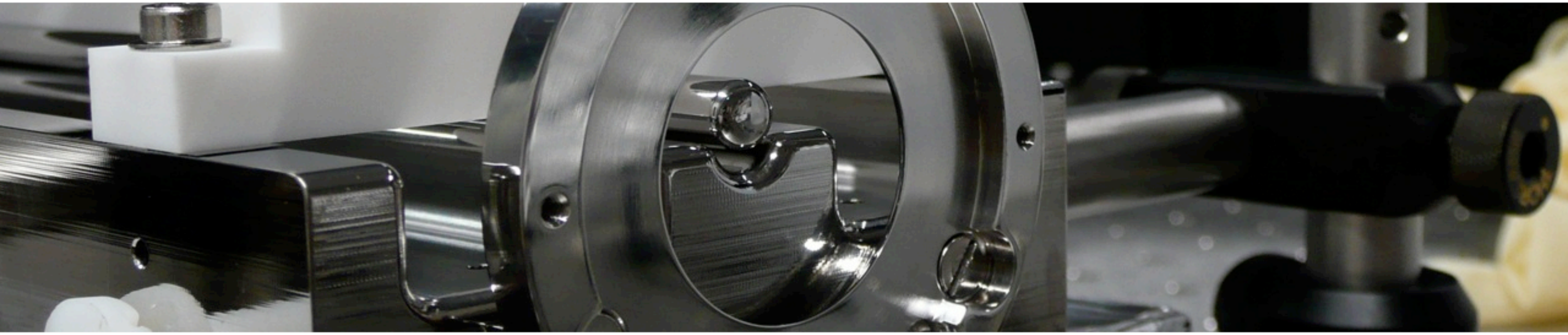
Cold and Controlled Molecules for Hot Experiments

Jochen Küpper

Center for Free-Electron Laser Science (CFEL)
DESY, Hamburg, Germany

The Hamburg Center for Ultrafast Imaging (CUI) and Department of Physics,
University of Hamburg, Germany



Cold and Controlled *large molecules* for Hot Experiments

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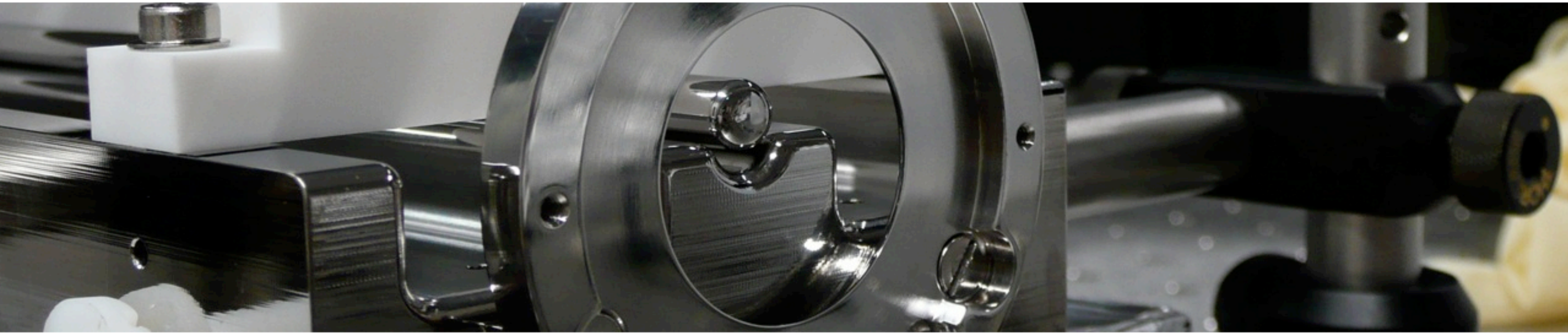
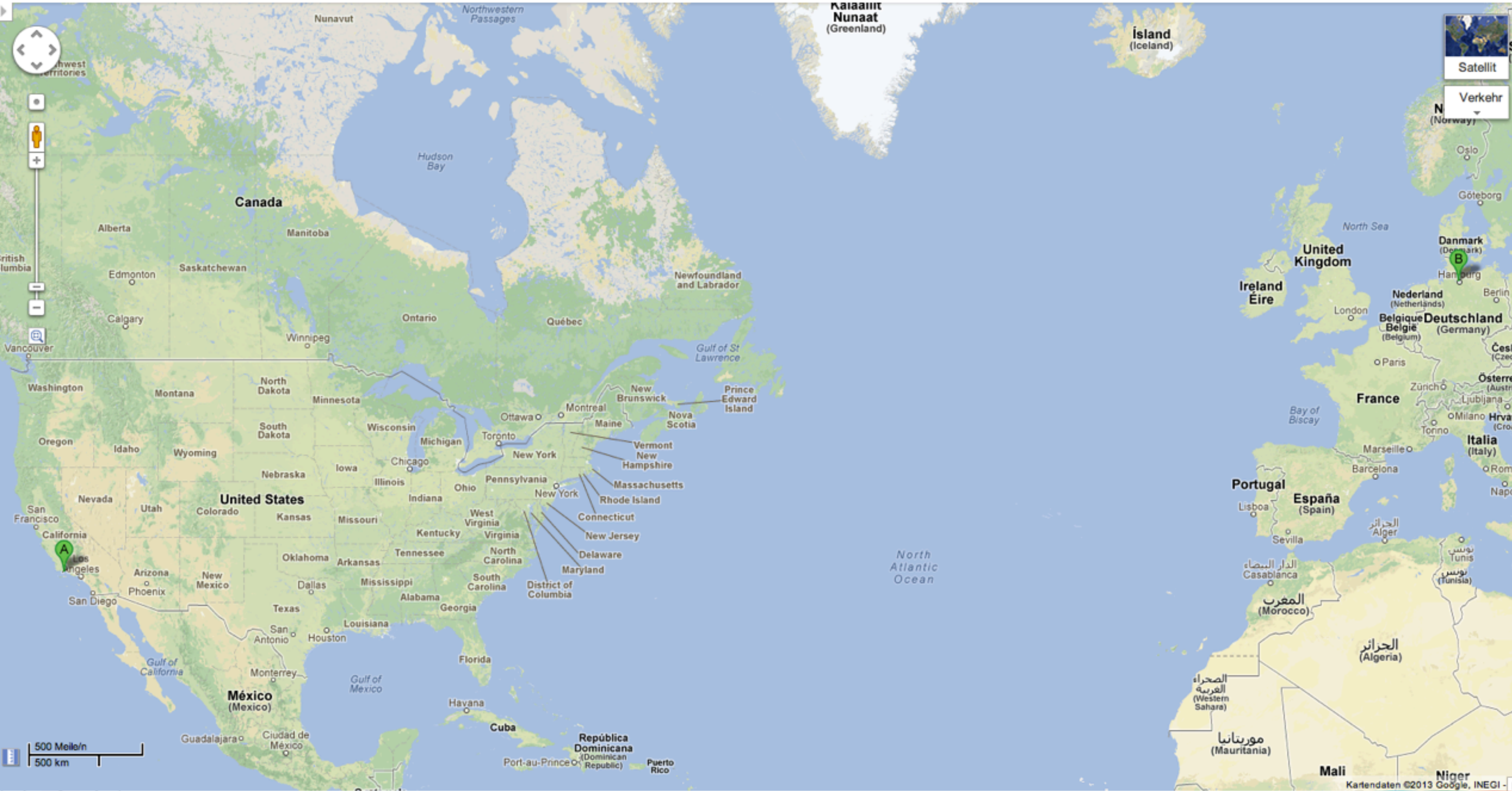


Controlled Molecule Imaging

Center for Free Electron Laser Science
Controlled Molecule Imaging Group

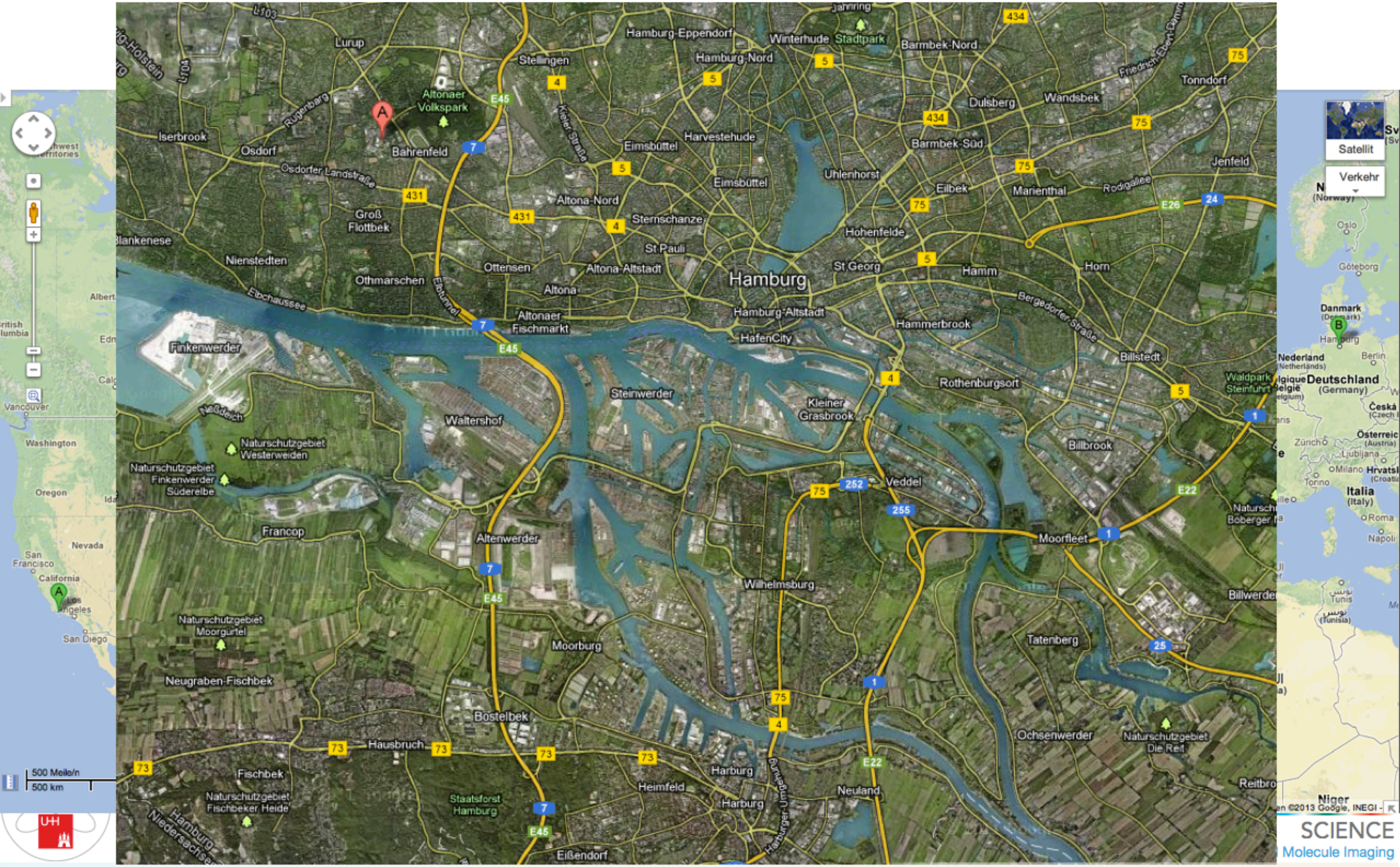


SCIENCE
Controlled Molecule Imaging

We are looking for motivated colleagues! See *http://desy.cfel.de/cid/cmi* for details.

Center for Free Electron Laser Science

Controlled Molecule Imaging Group



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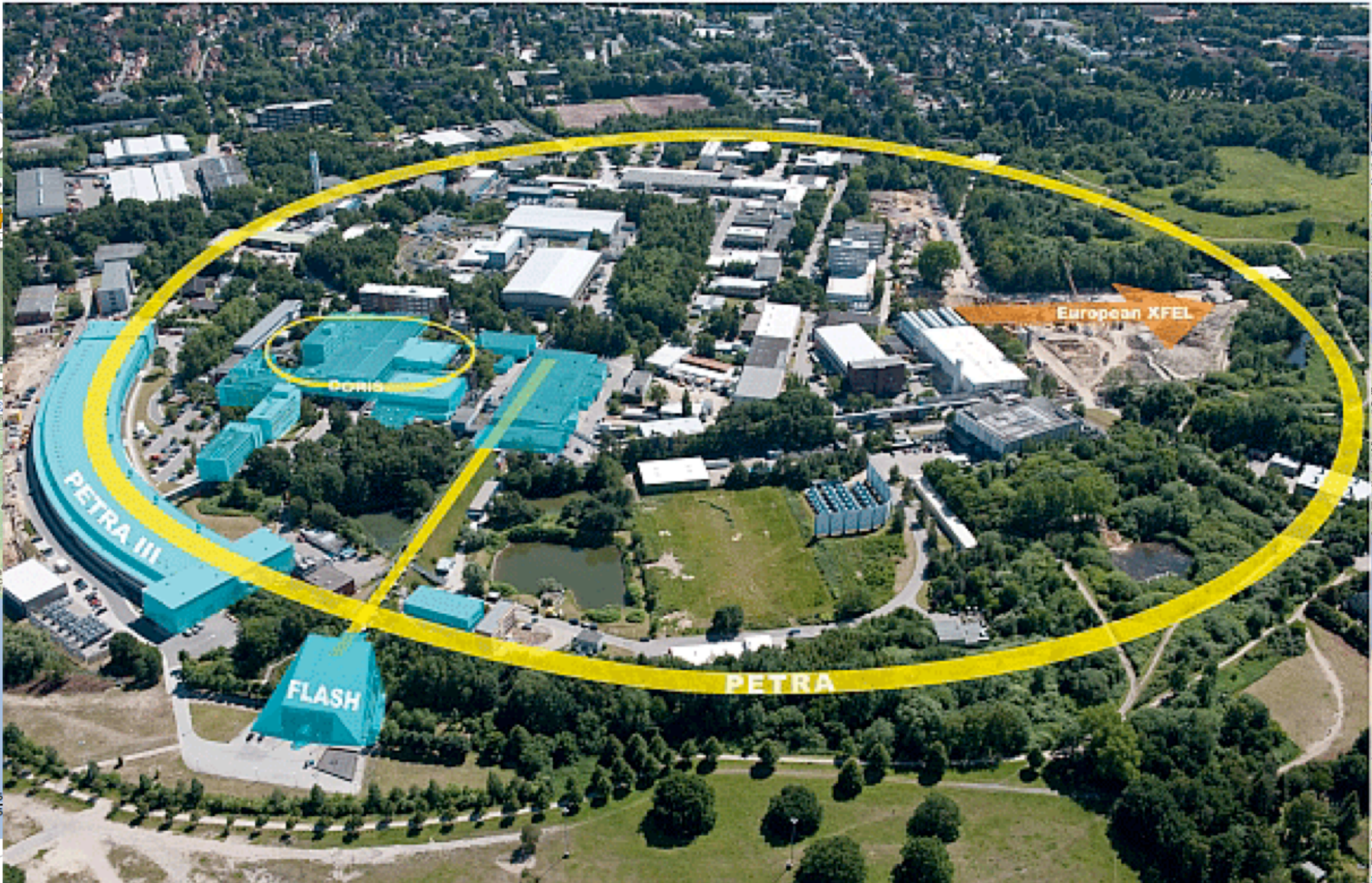
Center for Free Electron Laser Science
Controlled Molecule Imaging Group



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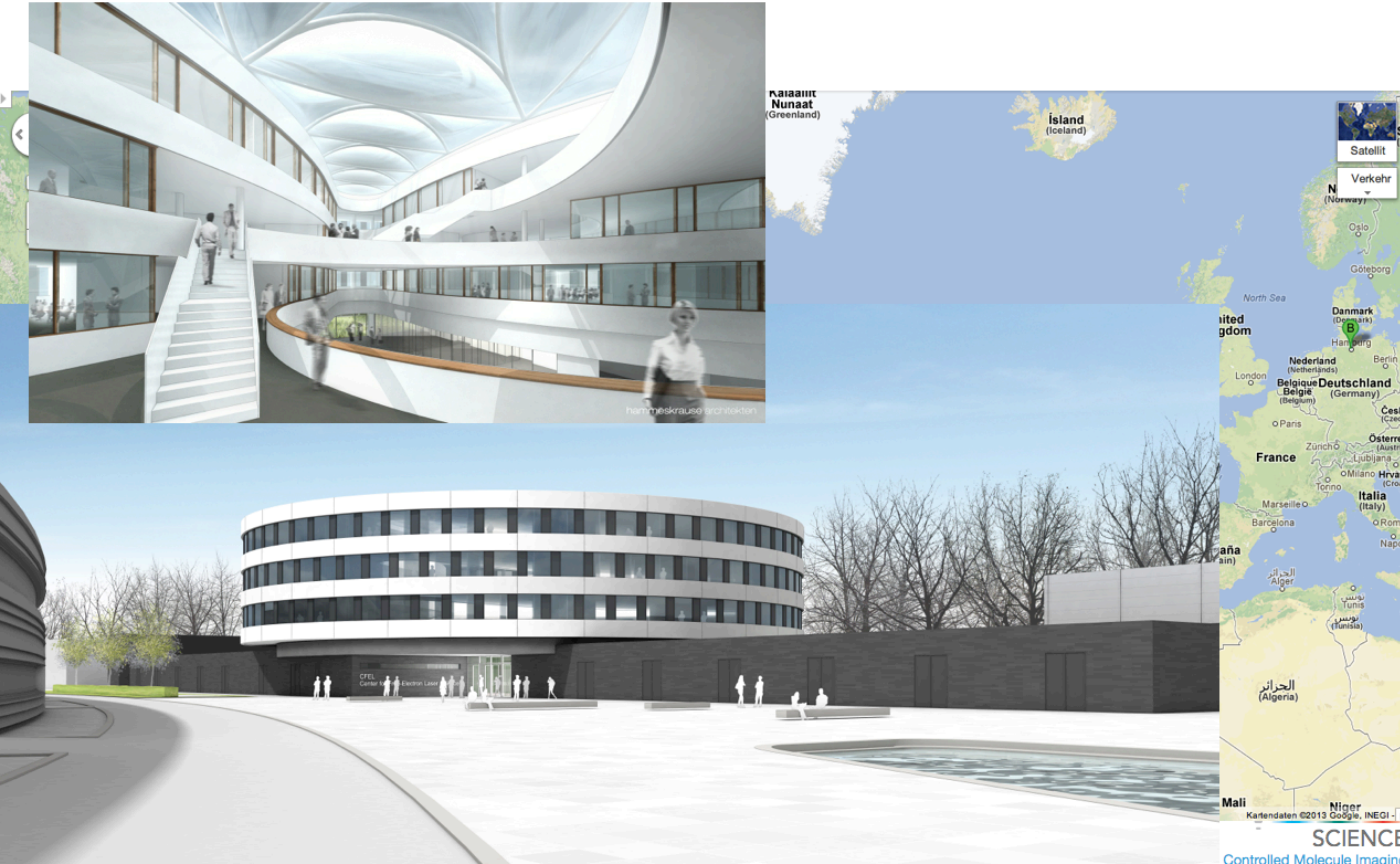

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Center for Free Electron Laser Science

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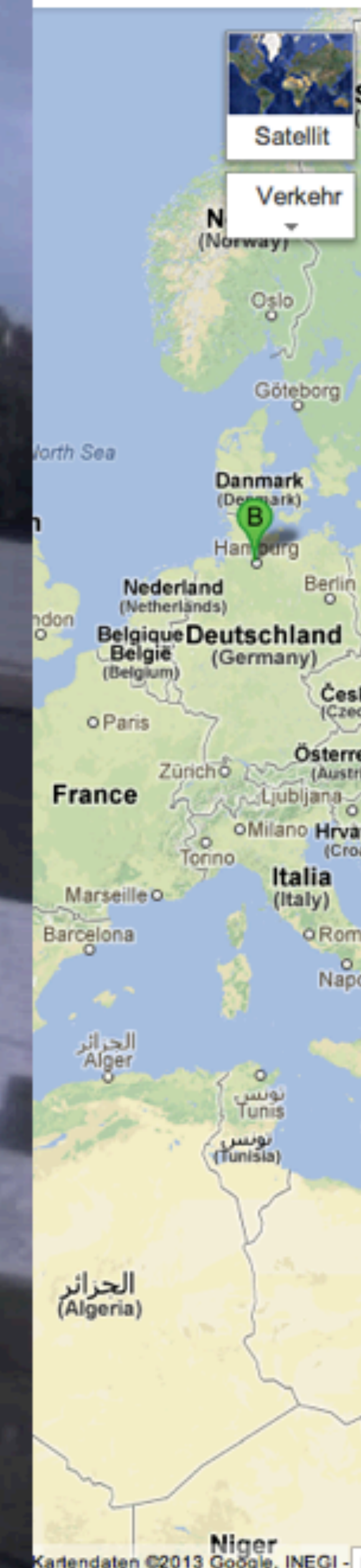

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Controlled Molecule Imaging

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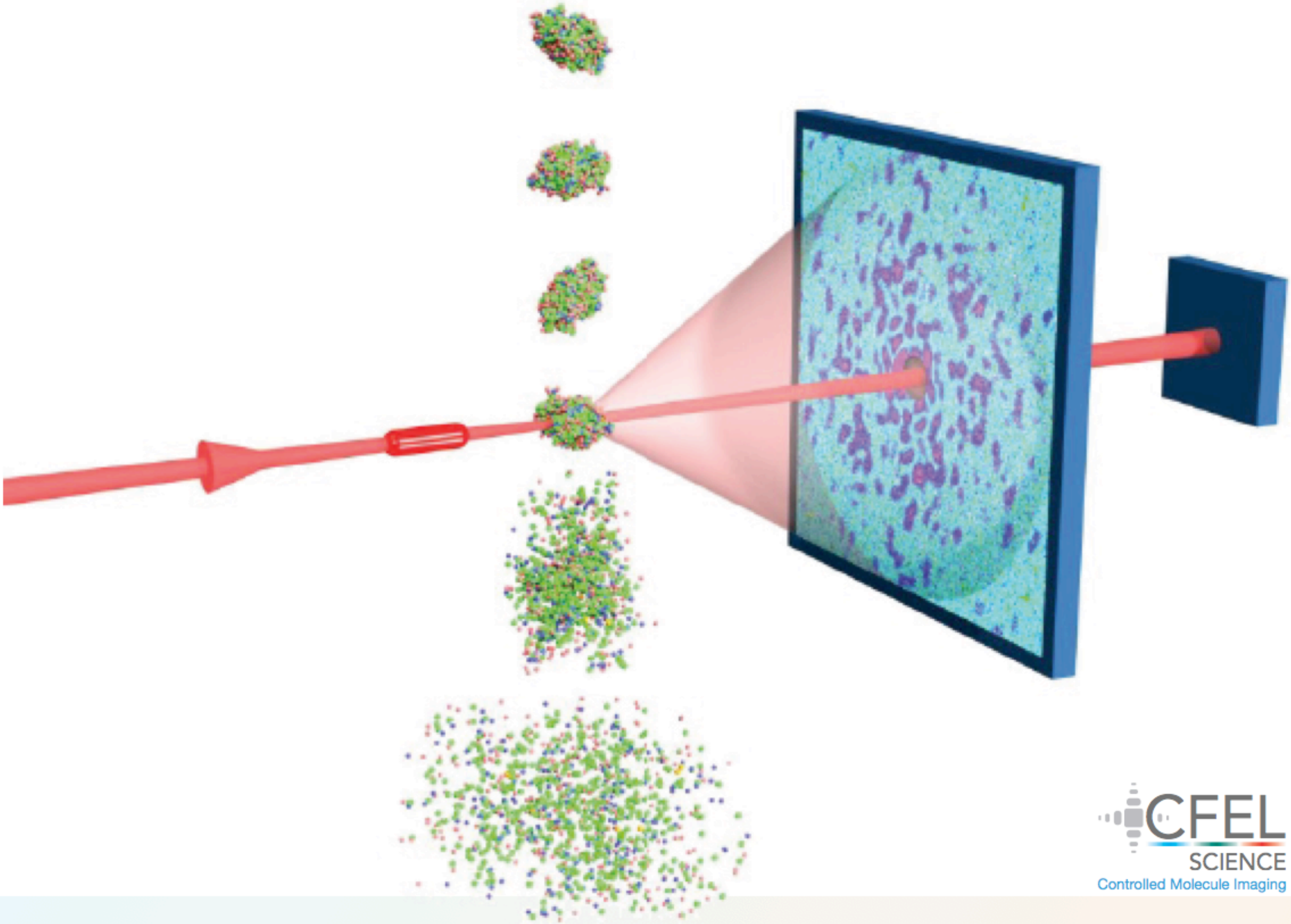
Center for Free Electron Laser Science
"our mission"

The *Center for Free-Electron Laser Science* is a novel joint enterprise of **DESY**, the **Max Planck Society (MPG)**, and the **University of Hamburg**. CFEL is designed to **advance science with next generation light sources**.

The three partners join forces to ***explore structural changes of atoms, molecules, condensed, biological, or warm dense matter on femtosecond time scales***. CFEL envisions uncovering the "dynamics of matter" by uniting expertise from various disciplines and research institutions into a new interdisciplinary and synergistic effort.

Center for Free Electron Laser Science
"our mission"



CFEL
SCIENCE
Controlled Molecule Imaging

Acknowledgments – FEL experiments

X-ray diffraction from isolated and strongly aligned gas-phase molecules with a free-electron laser

Jochen Küpper,[1,2,3,4,5,*] Stephan Stern,[1,2] Lotte Holmegaard,[1,6] Frank Filsinger,[4] Arnaud Rouzée,[7,8] Daniel Rolles,[5,9,†] Artem Rudenko,[5,10,‡] Per Johnsson,[11] Andrew V. Martin,[1,§] Marcus Adolph,[12] Andrew Aquila,[1] Saša Bajt,[1] Anton Barty,[1] Christoph Bostedt,[13] John Bozek,[13] Carl Caleman,[1] Ryan Coffee,[13] Nicola Coppola,[1] Tjark Delmas,[1] Sascha Epp,[5,10] Benjamin Erk,[5,10,†] Lutz Foucar,[5,9] Tais Gorkhover,[12] Lars Gumprecht,[1] Andreas Hartmann,[14] Robert Hartmann,[14] Günter Hauser,[15,16] Peter Holl,[14] Andre Hömke,[5,10] Nils Kimmel,[15] Faton Krasniqi,[5,9] Kai-Uwe Kühnel,[10] Jochen Maurer,[6] Marc Messerschmidt,[13] Robert Moshammer,[10,5] Christian Reich,[14] Benedikt Rudek,[5,10,†] Robin Santra,[1,2,3] Ilme Schlichting,[9,5] Carlo Schmidt,[5] Sebastian Schorb,[12] Joachim Schulz,[1,¶] Heike Soltau,[14] Lothar Strüder,[17] Jan Thøgersen,[6] Marc Vrakking,[7,8] Georg Weidenspointer,[15,16] Thomas A. White,[1] Cornelia Wunderer,[18] Gerard Meijer,[4,**] Joachim Ullrich,[10,5,††] Henrik Stapelfeldt,[6,19] and Henry N. Chapman[1,2,3]

[1] *Center for Free-Electron Laser Science, DESY, Notkestrasse 85, 22607 Hamburg, Germany*
[2] *University of Hamburg, Department of Physics, Luruper Chaussee 149, 22761 Hamburg, Germany*
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[4] *Fritz Haber Institute of the MPG, Faradayweg 4–6, 14195 Berlin, Germany*
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[9] *Max Planck Institute for Medical Research, 69120 Heidelberg, Germany*
[10] *Max Planck Institute for Nuclear Physics, 69117 Heidelberg, Germany*
[11] *Lund University, 22100 Lund, Sweden*
[12] *Technical University of Berlin, 10623 Berlin, Germany*
[13] *SLAC National Accelerator Laboratory, Menlo Park, CA 94025, USA*
[14] *PNSensor GmbH, 81739 Munich, Germany*
[15] *Max Planck Semiconductor Laboratory, 81739 Munich, Germany*
[16] *Max Planck Institute for Extraterrestrial Physics, 85741 Garching, Germany*
[17] *University of Siegen, Emmy-Noether Campus, Walter Flex Str.3, 57068 Siegen,Germany*[‡‡]
[18] *Deutsches Elektronen-Synchrotron (DESY), 22607 Hamburg, Germany*
[19] *Aarhus University, Interdisciplinary Nanoscience Center (iNANO), 8000 Aarhus C, Denmark*

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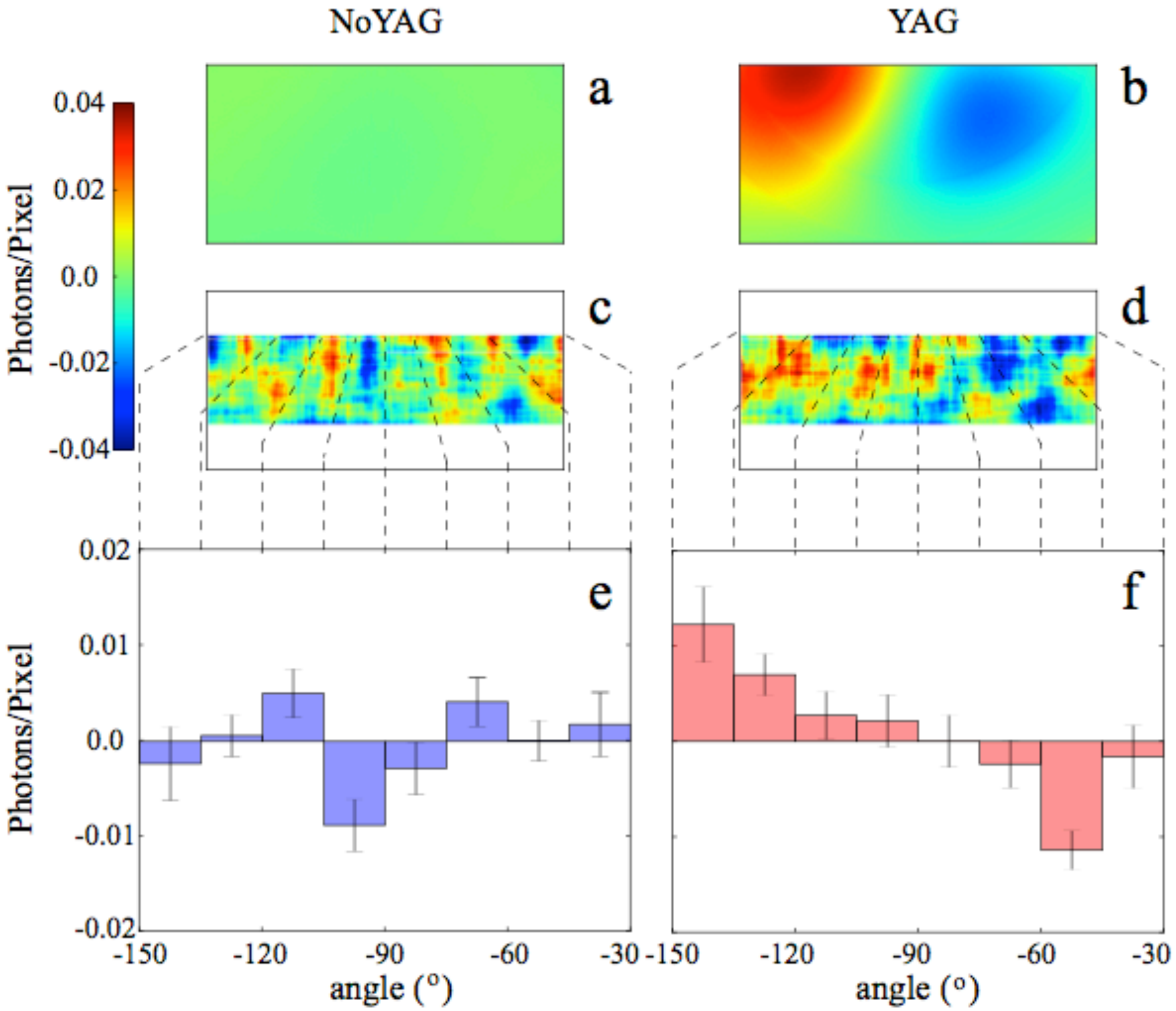
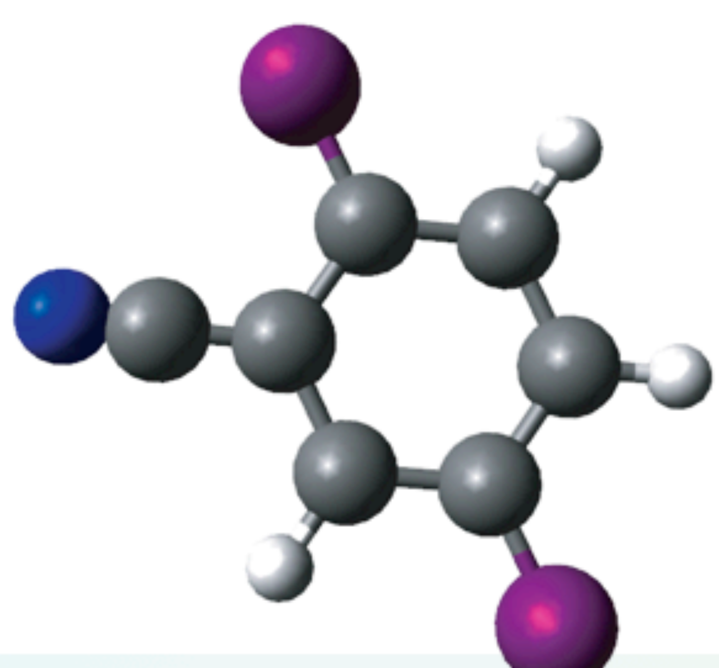
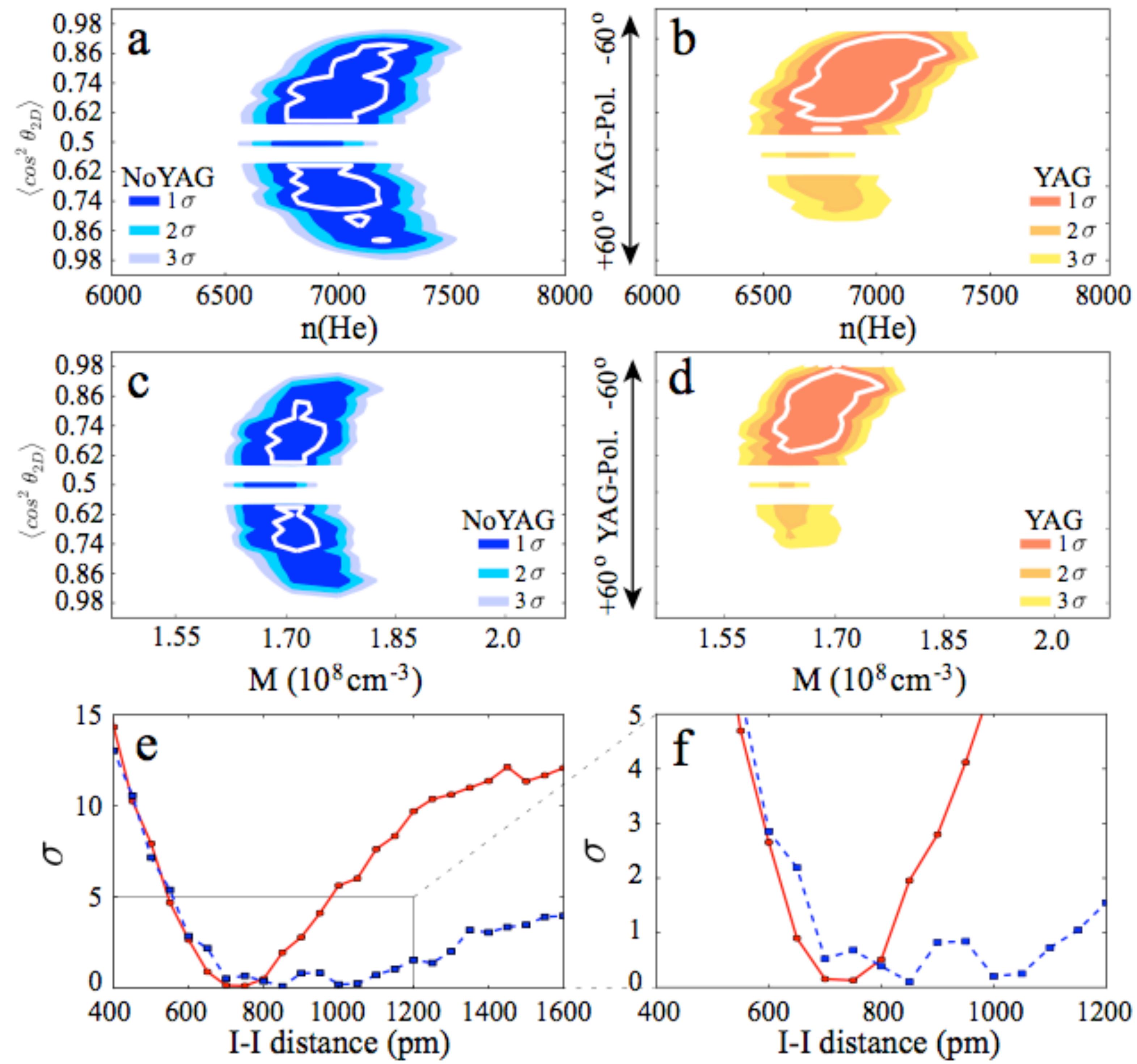
Controlled Molecule Imaging

under internal review

Coherent (fs) X-ray diffractive imaging of 2,6-diiodobenzonitrile
Analysis of anisotropic part of molecular x-ray diffraction pattern







JK, Stern, Holmegaard, et al, *under internal review*

Large/complex molecules in the gas phase
“Molecular building blocks of life” – tryptophan

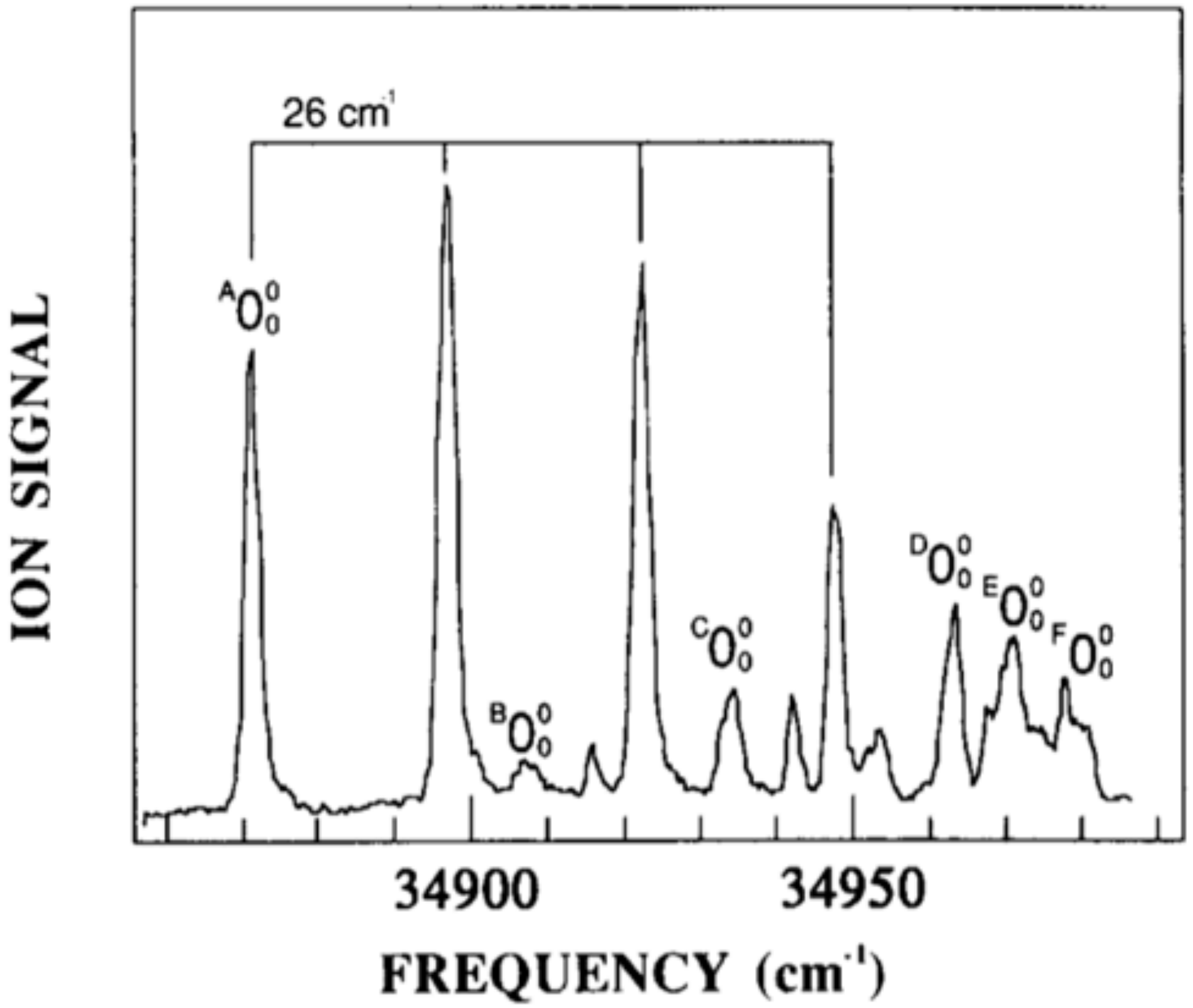
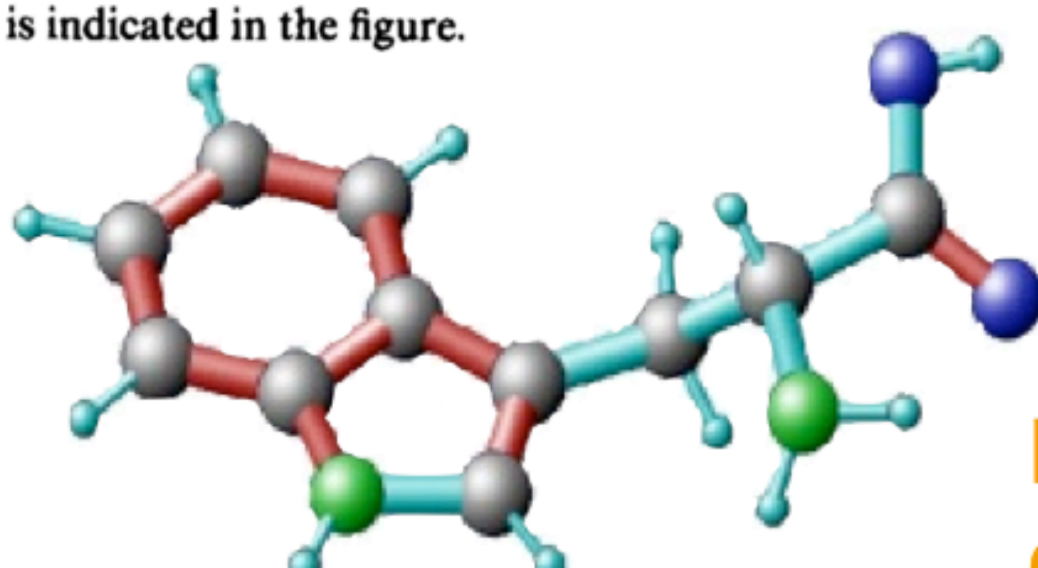


FIG. 1. Resonantly enhanced two-photon ionization spectrum of tryptophan in a molecular beam using the pulsed source described in Ref. 12. Electronic origin transitions due to conformers A–F were determined by the power saturation technique described in Ref. 12 and are labeled $^{A}0^0_0$–$^{F}0^0_0$. A nearly harmonic 26 cm $^{-1}$ vibrational progression built on the origin of comformer A is indicated in the figure.

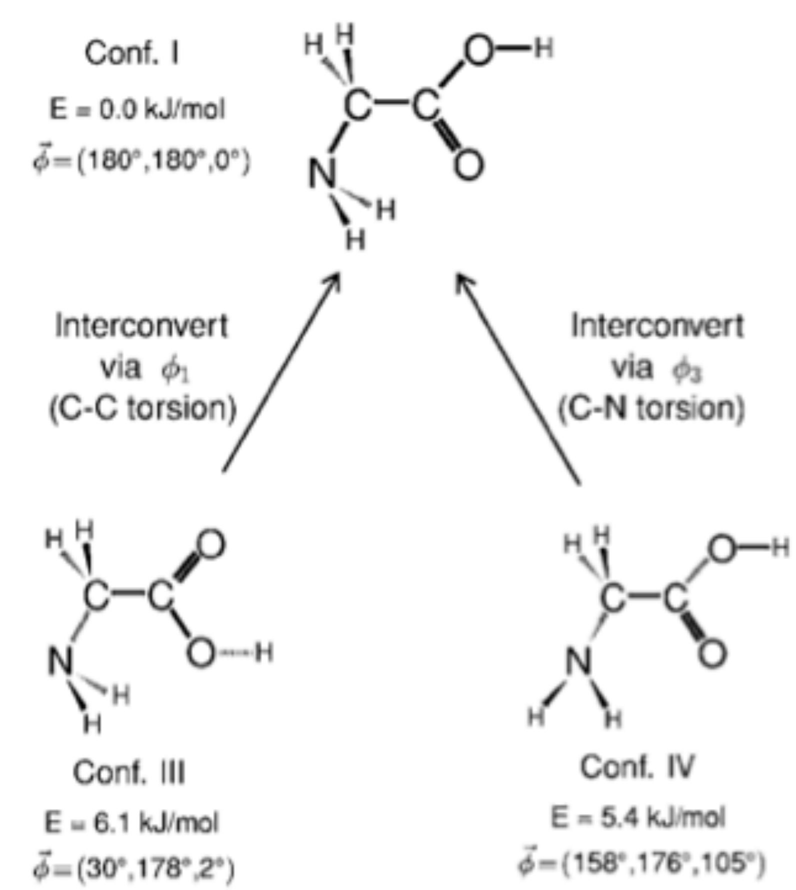


Millimeter Wave Spectrum of Glycine. A New Conformer

R. D. Suenram* and F. J. Lovas

Contribution from the Molecular Spectroscopy Division, National Bureau of Standards, Washington, D.C. 20234. Received June 13, 1980

J. Chem. Phys. **122**, 244323 (2005)



Multiple conformers present, even at low temperatures (1 K)

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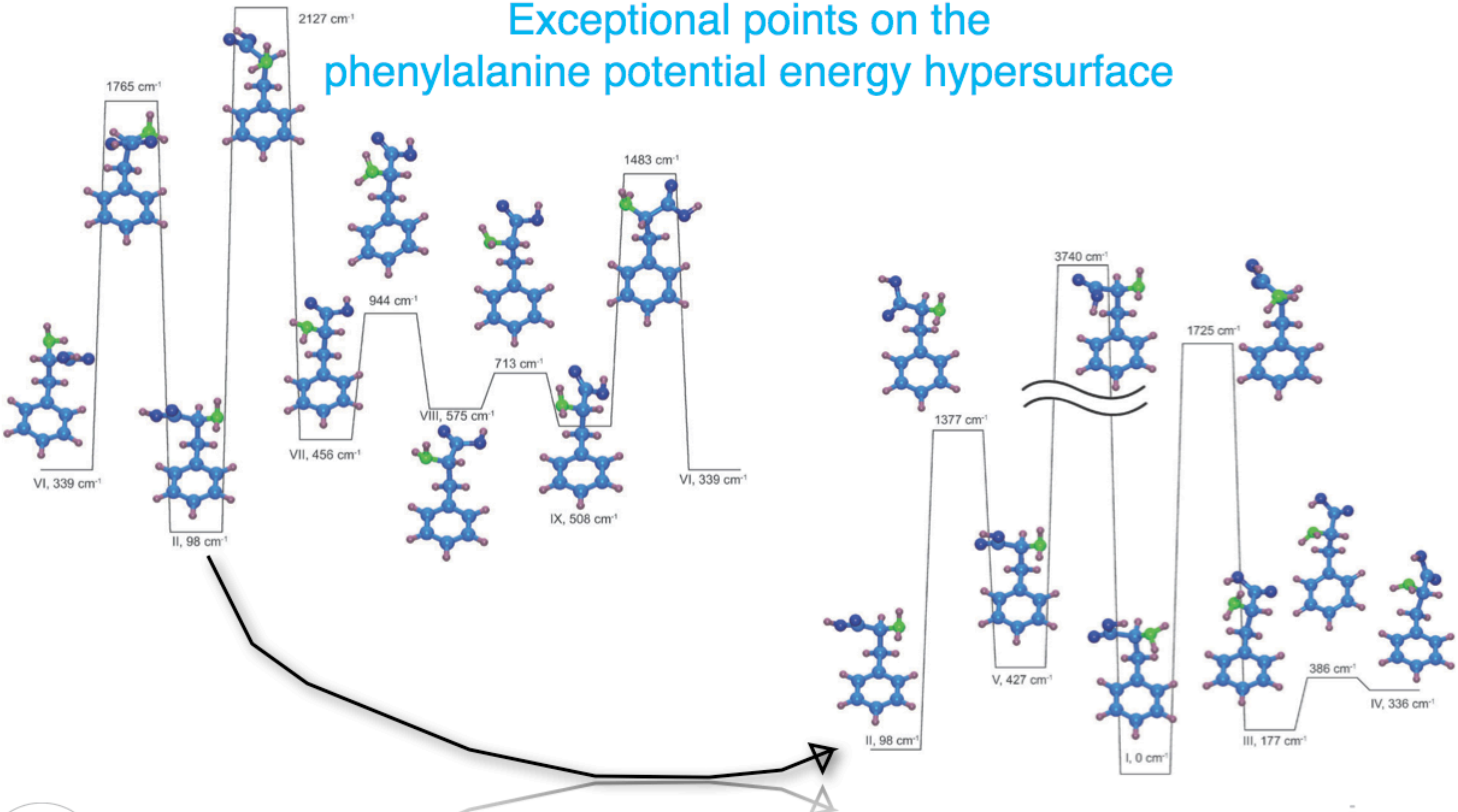
Controlled Molecule Imaging

Rizzo, Park, Peteanu, Levy, J. Chem. Phys. 83, 4819 (1985)

Complexity of molecules – "few body" quantum systems
Potential energy landscapes of isolated "floppy" molecules

Exceptional points on the phenylalanine potential energy hypersurface



$1\ cm^{-1}$ ≙ 30 GHz ≙ 12 J/mol ≙ 0.12 meV ≙ 4.6 µHartree ≙ 1.4 K

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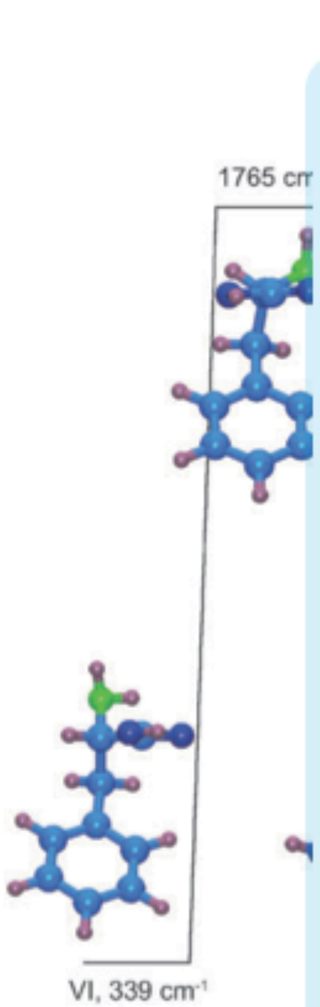
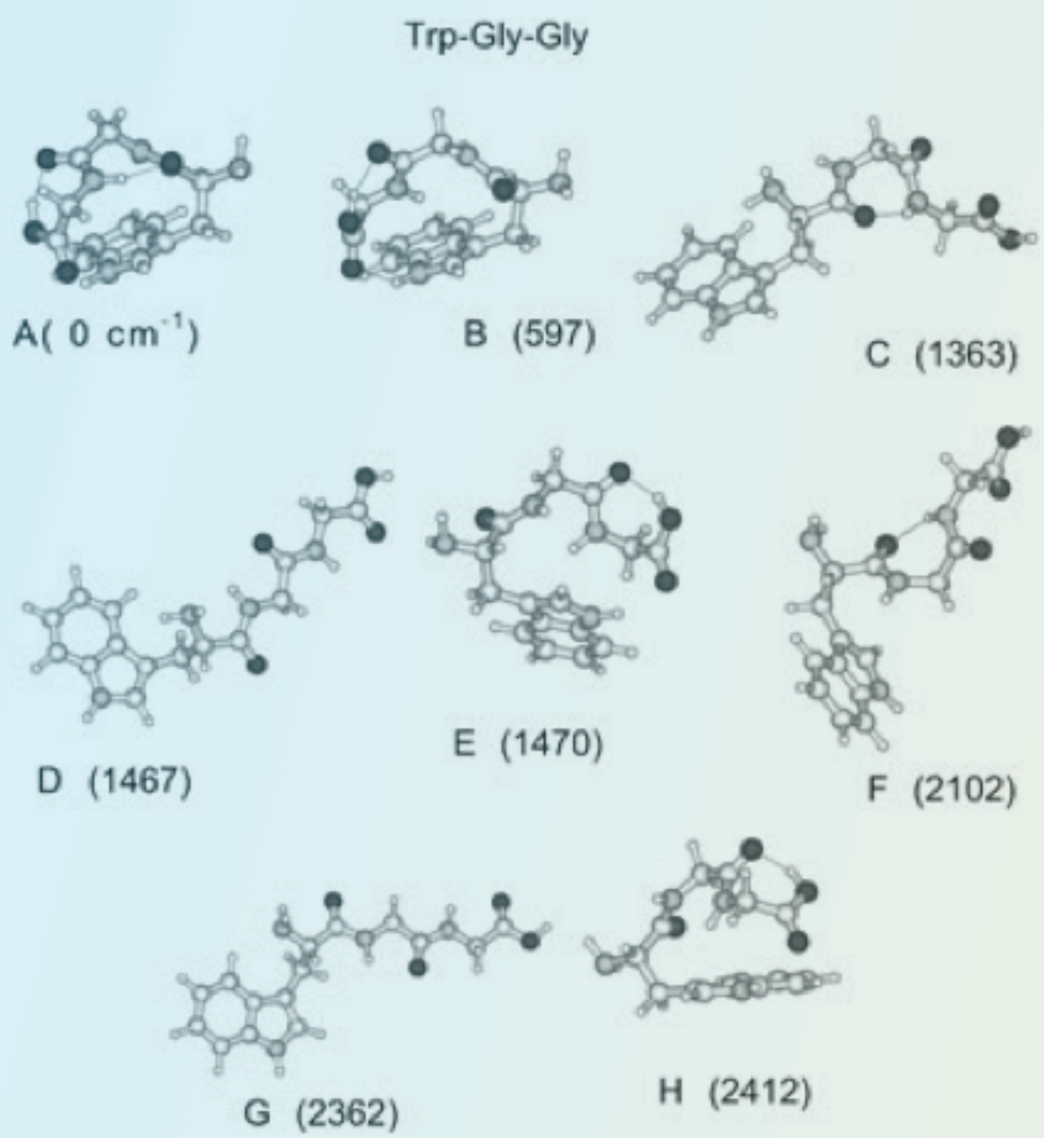
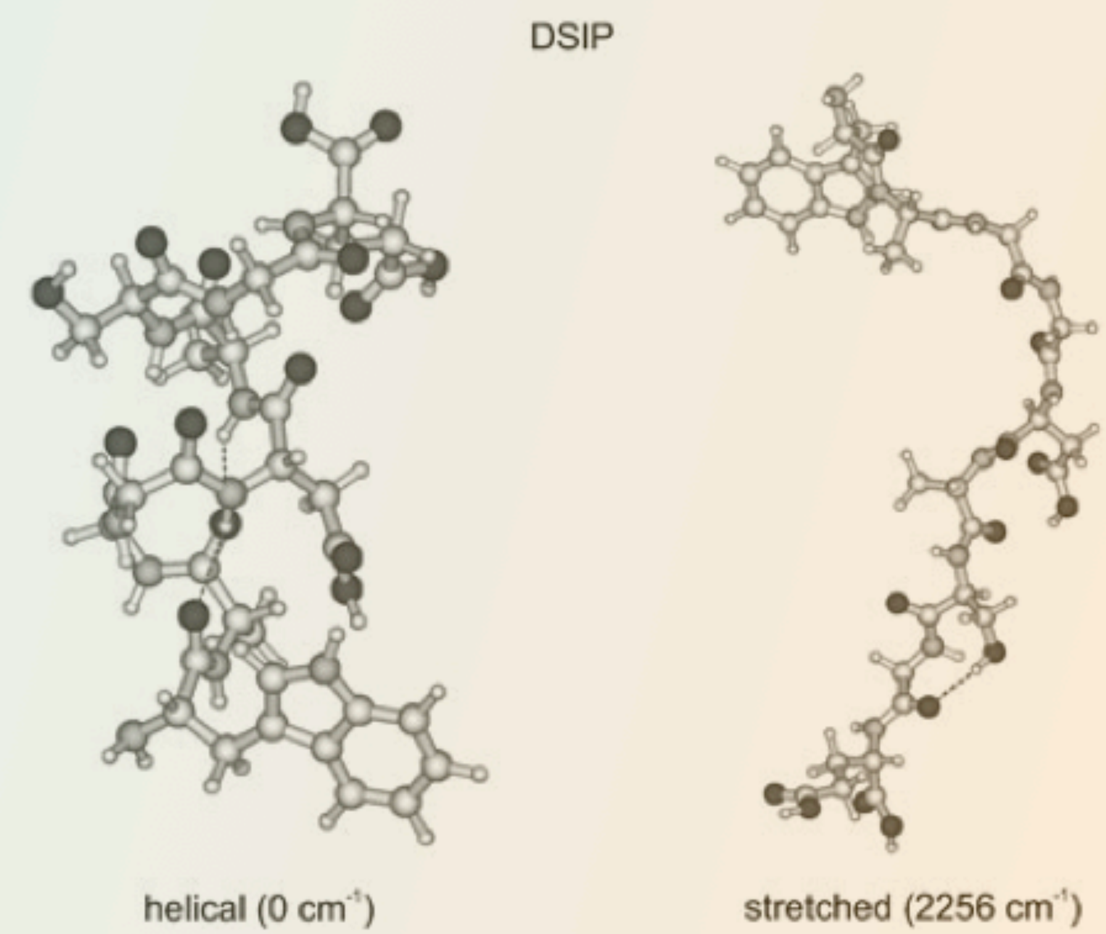
von Helden, Compagnon, Blom, Frankowski, Erlekam, Oomens, Brauer, Gerber, Meijer, *Phys. Chem. Chem. Phys.* **10**, 1248 (2008)

Complexity of molecules – "few body" quantum systems
Potential energy landscapes of isolated "floppy" molecules



and bigger....





1 cm^{-1} ≙ 30 GHz ≙ 12 J/mol ≙ 0.12 meV ≙ 4.6 μHartree ≙ 1.4 K

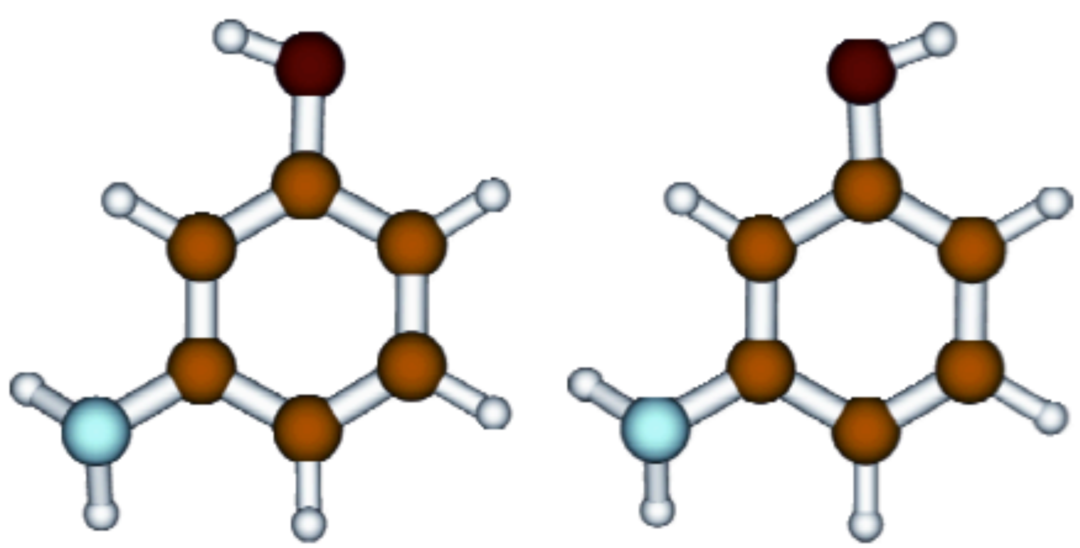
CFEL SCIENCE Controlled Molecule Imaging

von Helden, Compagnon, Blom, Frankowski, Erlekam, Oomens, Brauer, Gerber, Meijer, *Phys. Chem. Chem. Phys.* **10**, 1248 (2008)

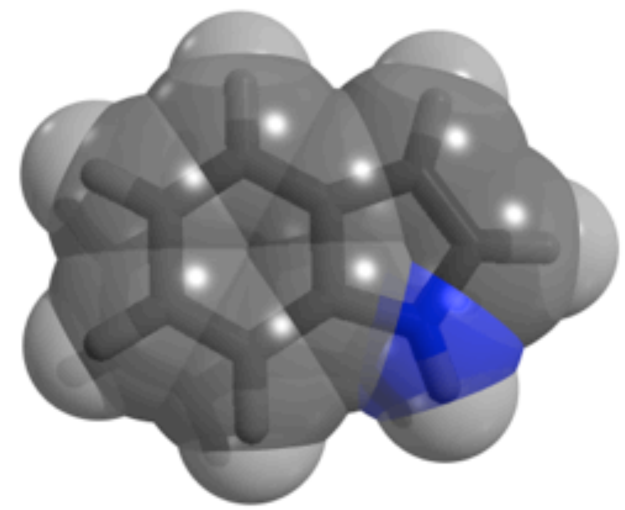
Large/complex molecules in the gas phase
our sample

3-aminophenol

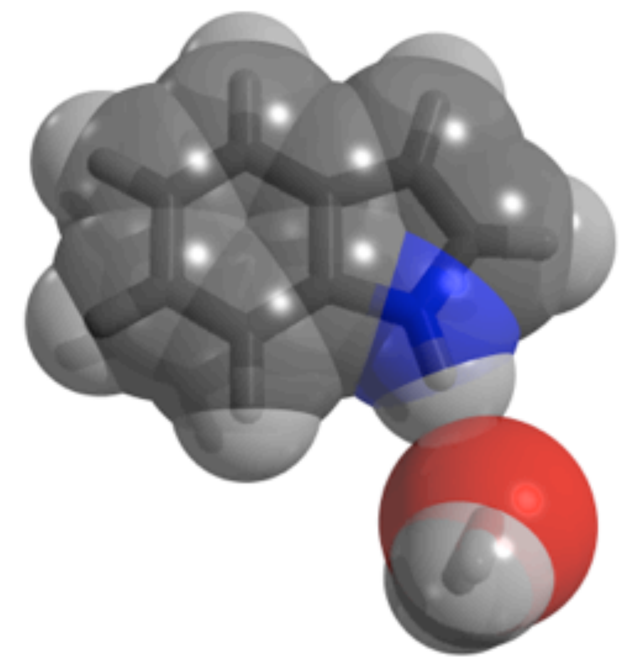
cis trans

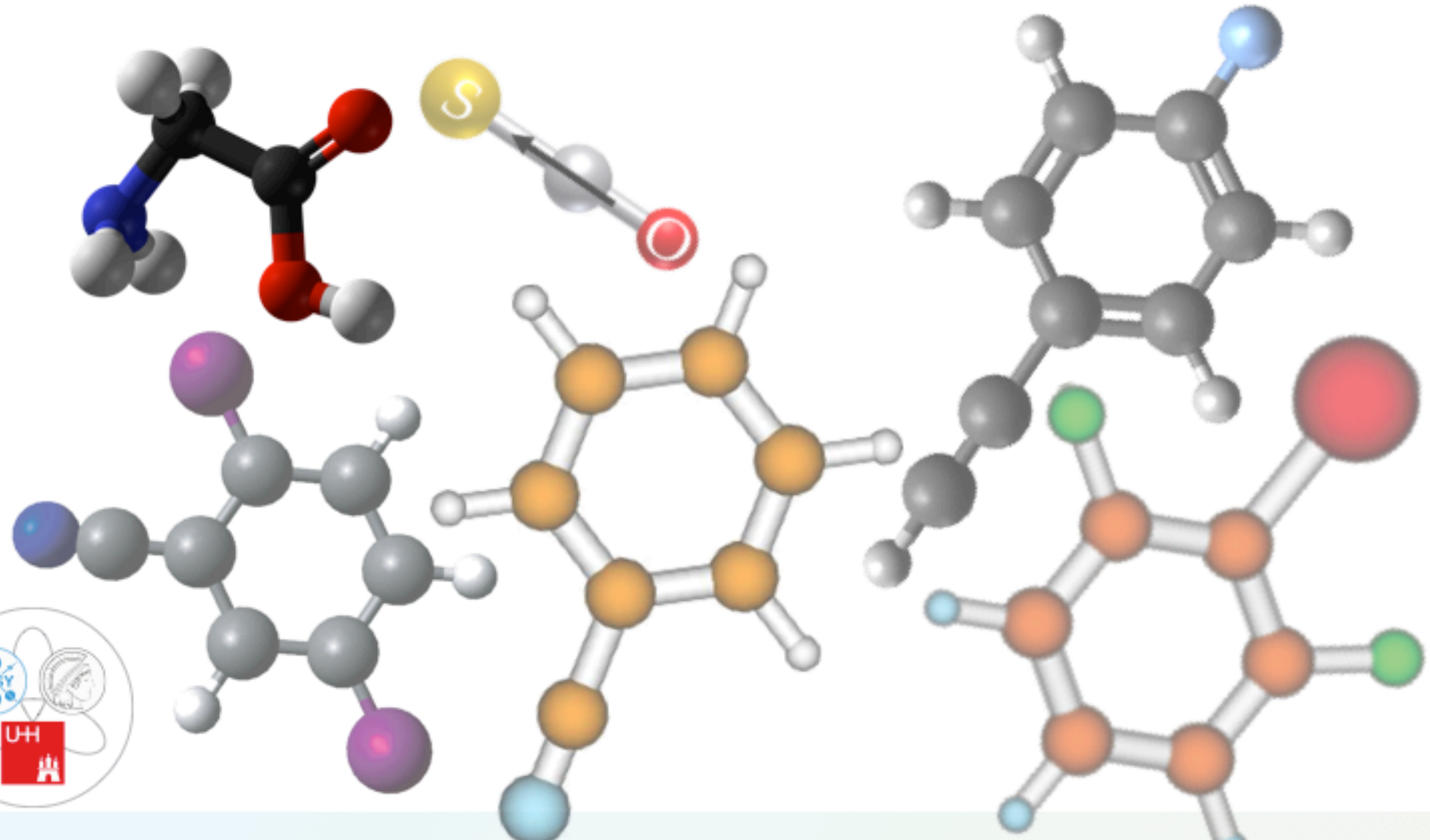


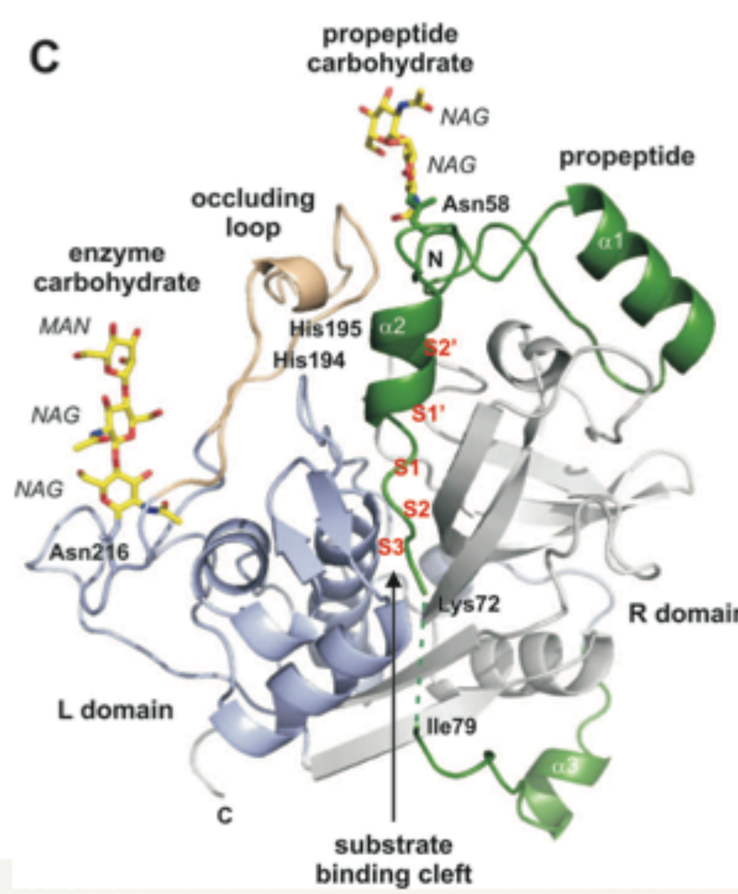
indole

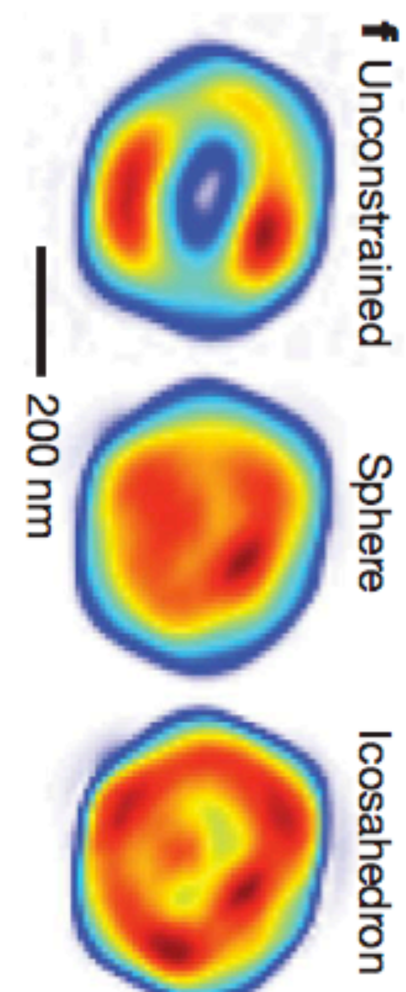


indole-water









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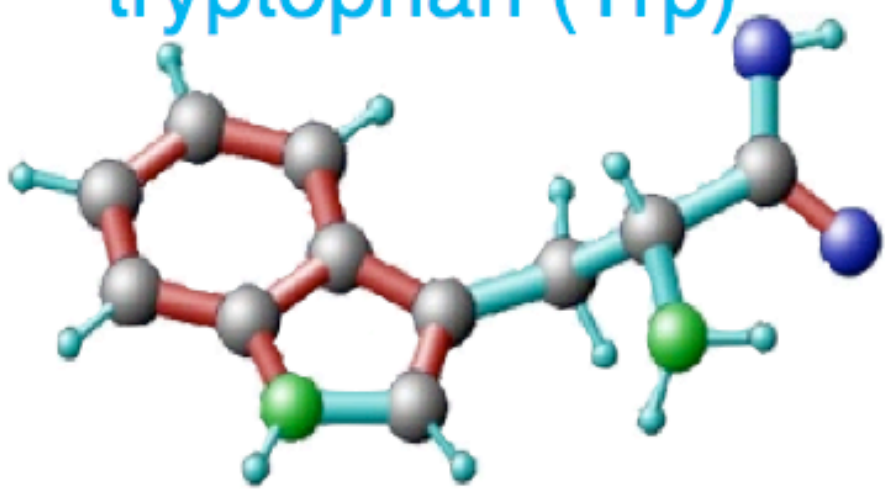
Large/complex molecules in the gas phase
How do “molecular building blocks of life” work?



- Multiple conformers present,
  even at the low temperatures (few K)
  in a supersonic jet
- Studying well-defined systems:
- Which structures (conformations) are (most) stable?
  - Why?
  - What are their intrinsic properties?
- How can we investigate these stable structures?
- How can we study the (chemical) dynamics on their corrugated potential energy surfaces? → *classical* “molecular movie”
- How do structure and dynamics depend on environment (solvation)?

How can we investigate these open questions
for complex molecules
without (intricate) **prior knowledge**?

An “electronic timescale” in chemistry
attosecond charge migration

vs

Born-Oppenheimer approximation

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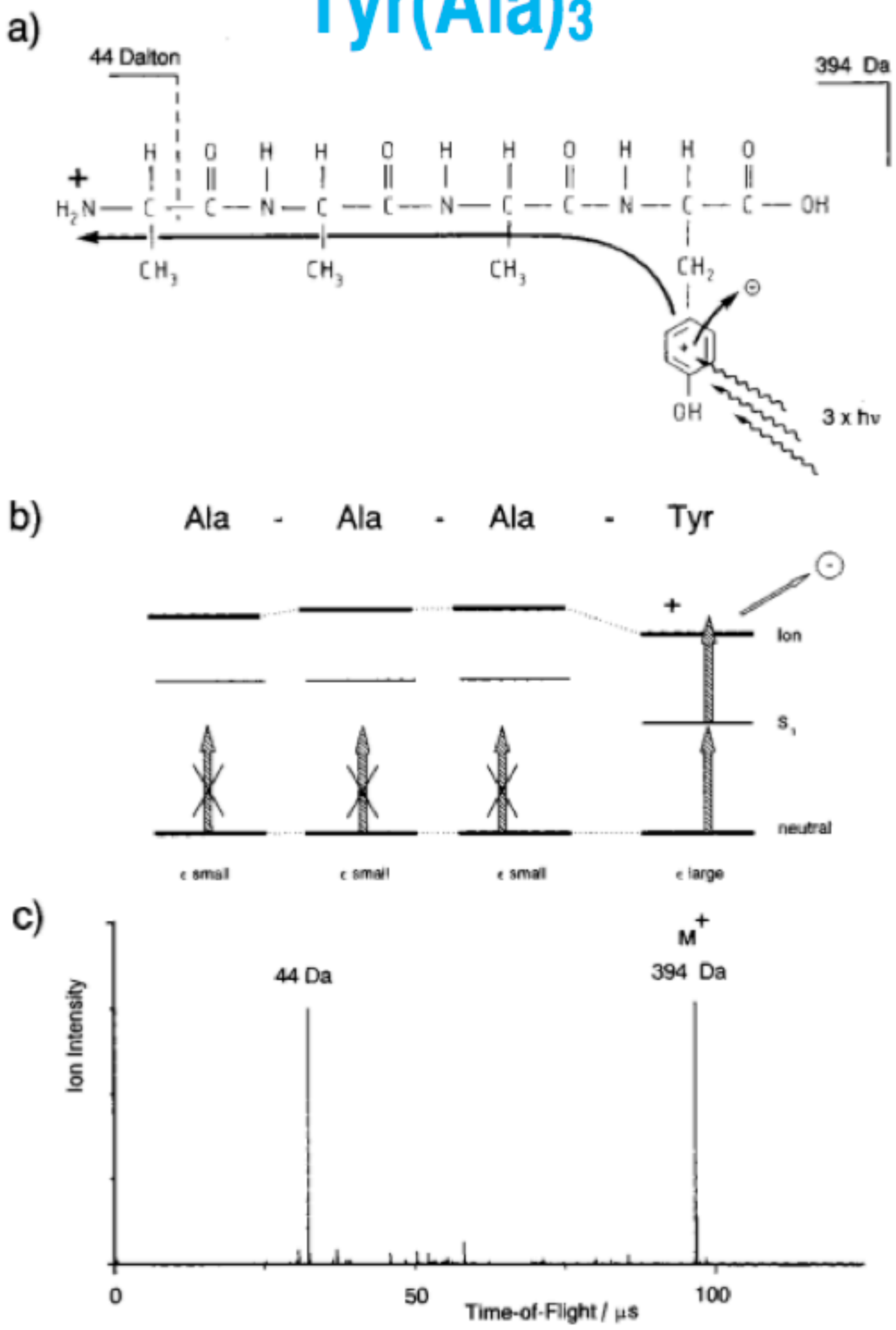
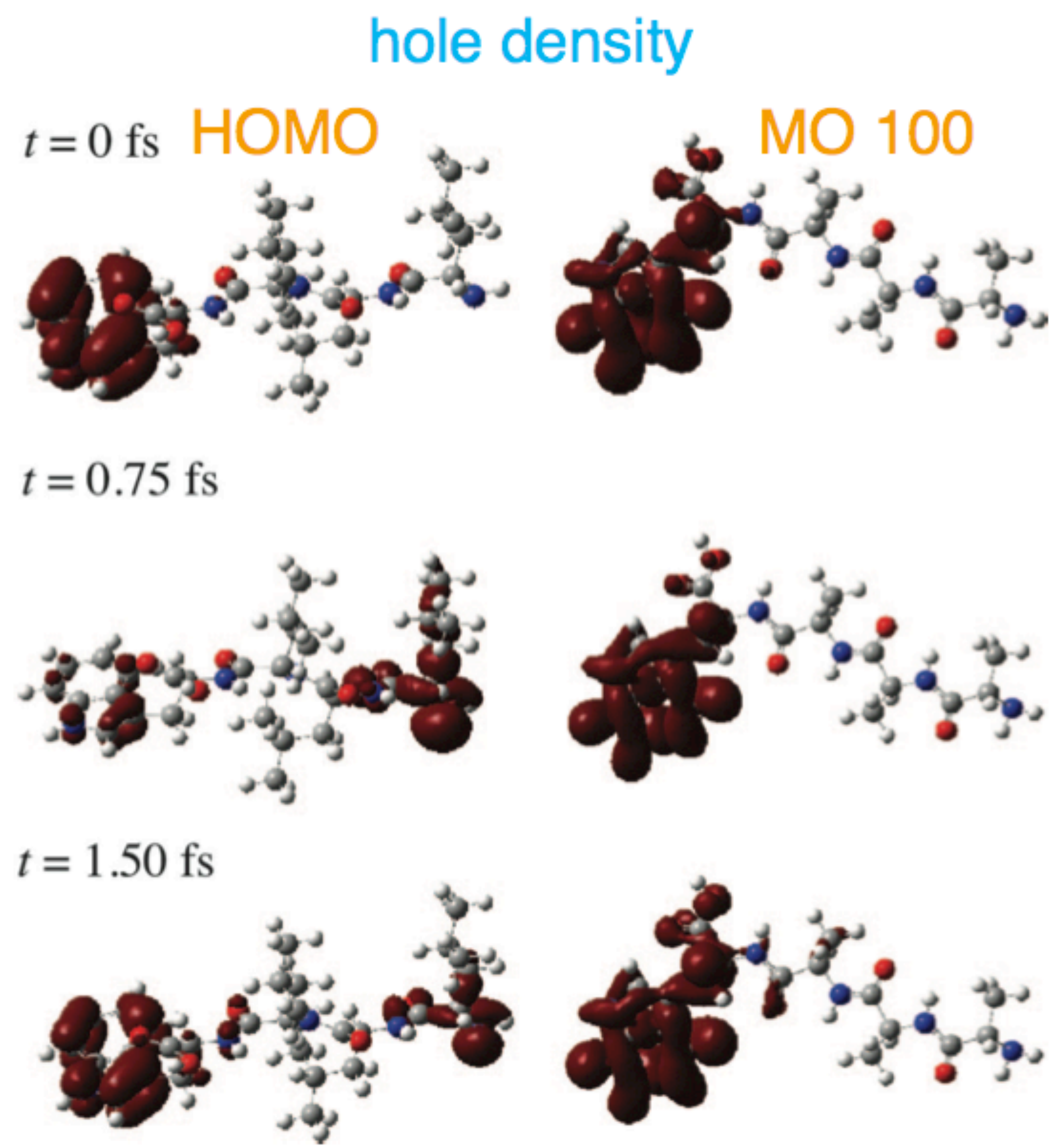
Controlled Molecule Imaging

An "electronic timescale" in chemistry
attosecond charge migration

Tyr(Ala)3



Trp(Leu)3



Weinkauf, Schanen, Metsala, Schlag, Bürgle, Kessler, *J. Phys. Chem.* **100**, 18567 (1996)

Remacle, Levine, *PNAS* **103**, 6793 (2006)

The "small molecule" case (Ultrafast charge) migration in glycine

Millimeter Wave Spectrum of Glycine. A New Conformer

R. D. Suenram* and F. J. Lovas

Contribution from the Molecular Spectroscopy Division, National Bureau of Standards, Washington, D.C. 20234.

Received June 13, 1980

Abstract: More sensitive observations on the vapor of glycine, the smallest amino acid, have been made in the millimeter region. This work has led to the identification of a second conformer in the gas phase. This conformer has the amino hydrogens hydrogen bonded to the carbonyl oxygen and the hydroxyl hydrogen in the normal cis configuration with respect to the carbonyl group. This new conformer is ≃490 (150) cm^{-1} *lower* in energy than the conformer originally reported. Its spectrum is weaker due to the smaller dipole moment ($\mu_a \simeq 1.00$ (15) D). The distortion fit of 37 transitions yield $A'' = 10341.76$ (17) MHz, $B'' = 3876.195$ (9) MHz, and $C'' = 2912.361$ (10) MHz for the ground state. The excellent agreement between experiment and theory is discussed.

J. Am. Chem. Soc. **1980**, *102*, 7180–7184

INTERSTELLAR GLYCINE

Yi-Jehng Kuan,[1,2] Steven B. Charnley,[3] Hui-Chun Huang,[1] Wei-Ling Tseng,[1] and Zbigniew Kisiel[4]

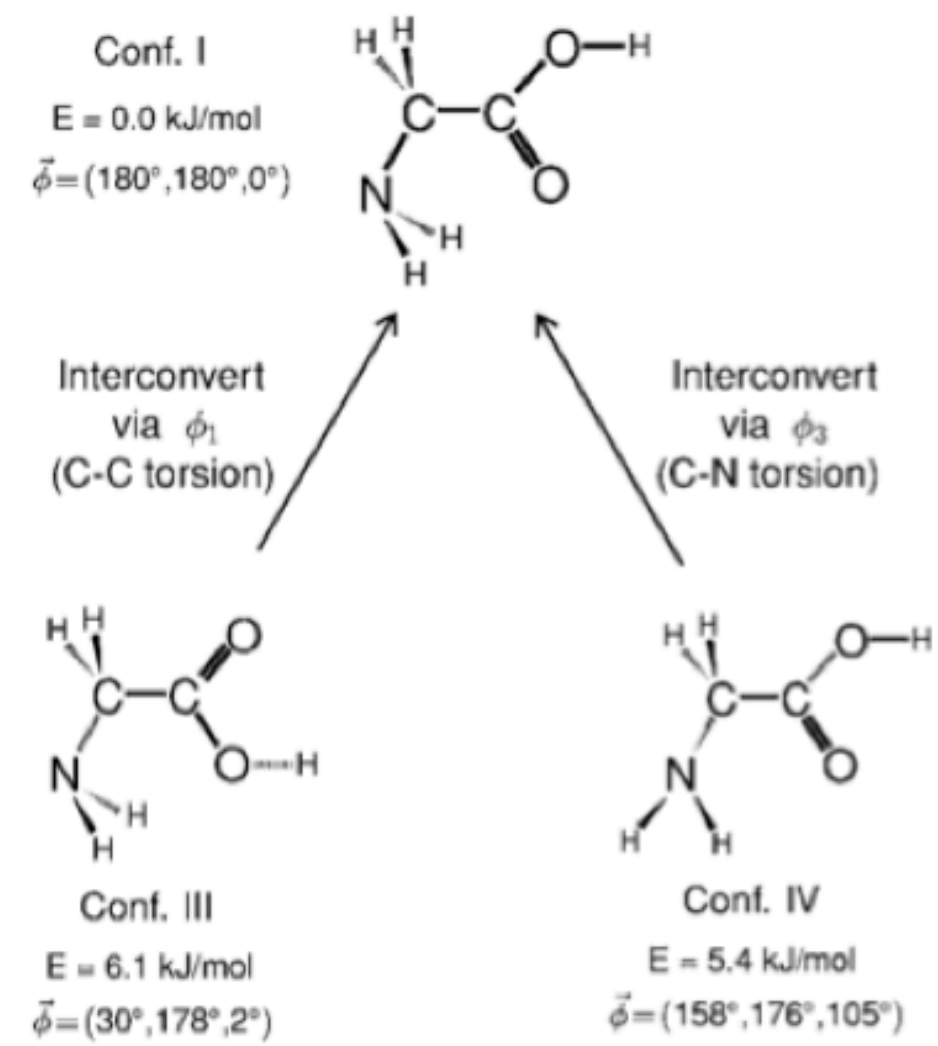
Received 2002 November 15; accepted 2003 April 1

ABSTRACT

We have searched for interstellar conformer I glycine (NH_2CH_2COOH), the simplest amino acid, in the hot molecular cores Sgr B2(N-LMH), Orion KL, and W51 e1/e2. An improved search strategy for intrinsically weak molecular lines, involving multisource observations, has been developed and implemented. In total, 82 spectral frequency bands, in the millimeter-wave region, were observed over a 4 yr period; 27 glycine lines were detected in 19 different spectral bands in one or more sources. The rotational temperatures derived from " rotation diagrams " are 75^{+29}_{-16} K for Sgr B2(N-LMH), 141^{+76}_{-37} K for Orion KL, and 121^{+71}_{-32} K for W51 e1/e2. The total column densities inferred are $4.16^{+3.22}_{-1.82} \times 10^{14}$ cm^{-2} for Sgr B2, $4.37^{+1.79}_{-1.27} \times 10^{14}$ cm^{-2} for Orion, and $2.09^{+1.22}_{-0.77} \times 10^{14}$ cm^{-2} for W51. Production of interstellar glycine by both gas-phase ion-molecule reactions and by ultraviolet photolysis of molecular ices is briefly discussed. The discovery of interstellar glycine strengthens the thesis that interstellar organic molecules could have played a pivotal role in the prebiotic chemistry of the early Earth.

The Astrophysical Journal, 593:848–867, 2003 August 20

J. Chem. Phys. **122**, 244323 (2005)



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Controlled Molecule Imaging

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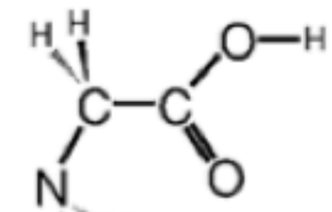
Conf. I
E = 0.0 kJ/mol
$\vec{\phi}$=(180°,180°,0°)



Charge migration in different conformers of glycine: The role of nuclear geometry

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Theoretische Chemie, PCI, Universität Heidelberg, Im Neuenheimer Feld 229, 69120 Heidelberg, Germany

Received 5 February 2007; accepted 18 April 2007
Available online 25 April 2007

Chemical Physics 338 (2007) 320–328

Abstract

The migration of hole charge created via ionization of the main conformers of the gaseous amino acid glycine is studied. The migration is ultrafast and is mediated solely by many-electron effects, i.e. electronic correlation and relaxation. The influence of the nuclear geometry is investigated by studying the three most abundant conformers of glycine. It is shown that the electron dynamics following ionization can be dramatically different for the different conformers. To facilitate the discussion of the charge migration, the ionization spectra of the conformers are computed as well.

prebiotic chemistry of the early Earth.

The Astrophysical Journal, 593:848–867, 2003 August 20

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Kuleff, Cederbaum, *Chem. Phys.* 338, 321 (2007)

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J. Chem. Phys. **122**, 244323 (2005)

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Conf. I

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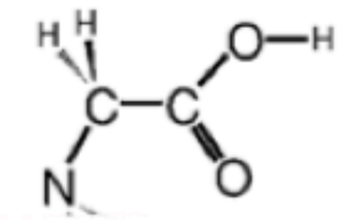

Charge migration in different conformers of glycine: The role of

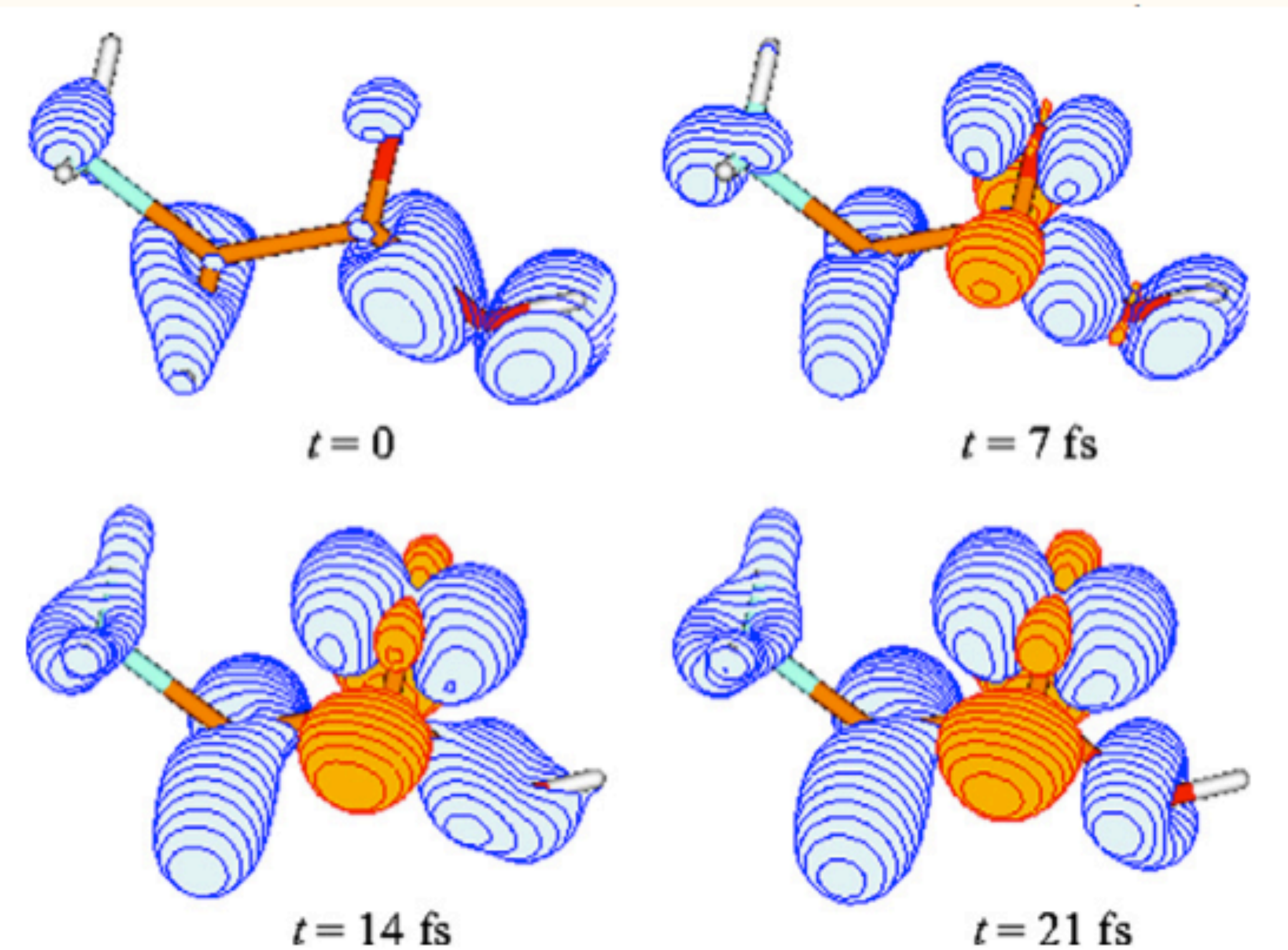

Fig. 5. Hole density $Q(\vec{r},t)$ of conformer Gly I at four different times

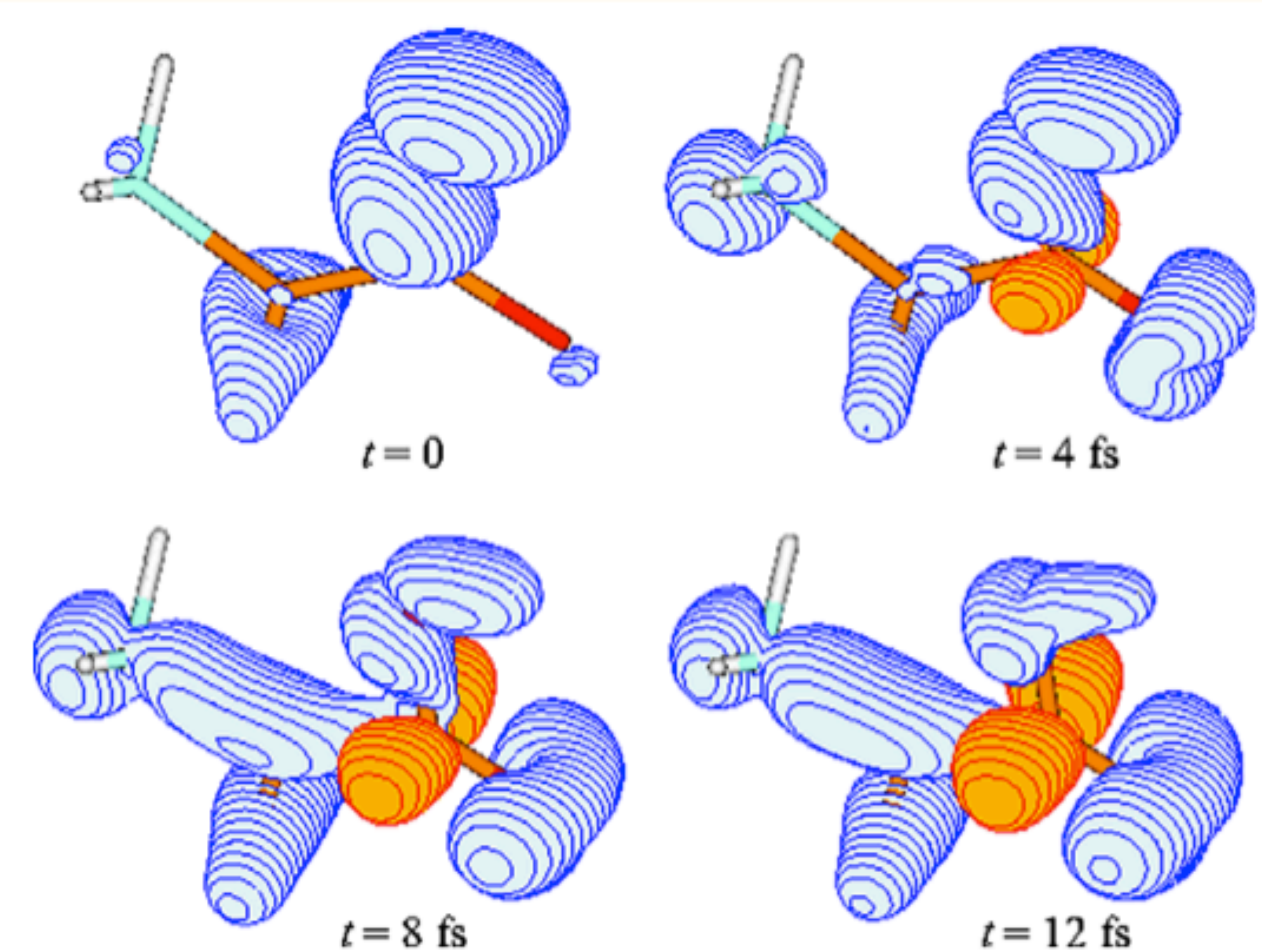

Fig. 6. Hole density $Q(\vec{r},t)$ of conformer Gly III at four different times

prebiotic chemistry of the early Earth.

THE ASTROPHYSICAL JOURNAL, 593:848–867, 2003 August 20

CFEL SCIENCE Controlled Molecule Imaging

Kuleff, Cederbaum, *Chem. Phys.* 338, 321 (2007)

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(Ultrafast charge) migration in glycine

Millimeter Wave Spectrum of Glycine. A New Conformer

J. Chem. Phys. **122**, 244323 (2005)

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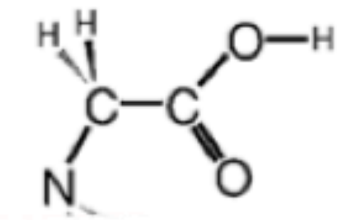


Charge migration in different conformers of glycine: The role of

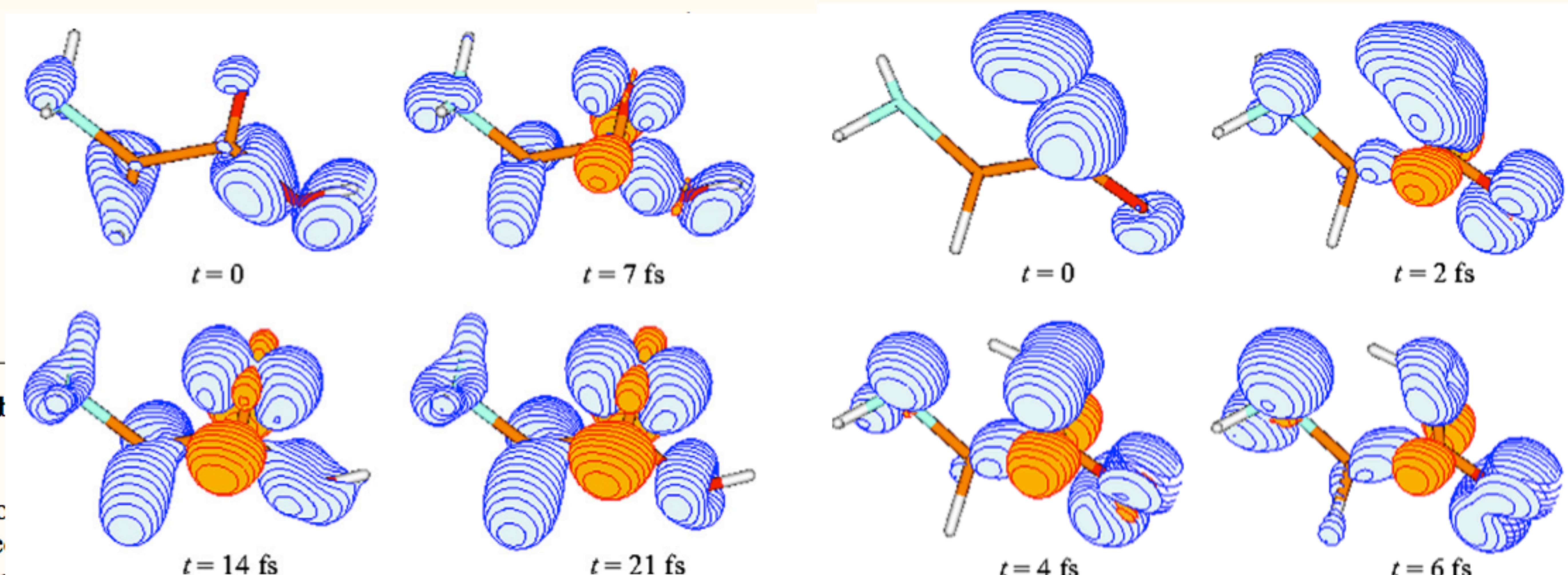


Fig. 5. Hole density $Q(\vec{r},t)$ of conformer Gly I at four different times

Fig. 7. Hole density $Q(\vec{r},t)$ of conformer Gly II at four different time

prebiotic chemistry of the early Earth.

THE ASTROPHYSICAL JOURNAL, 593:848–867, 2003 August 20

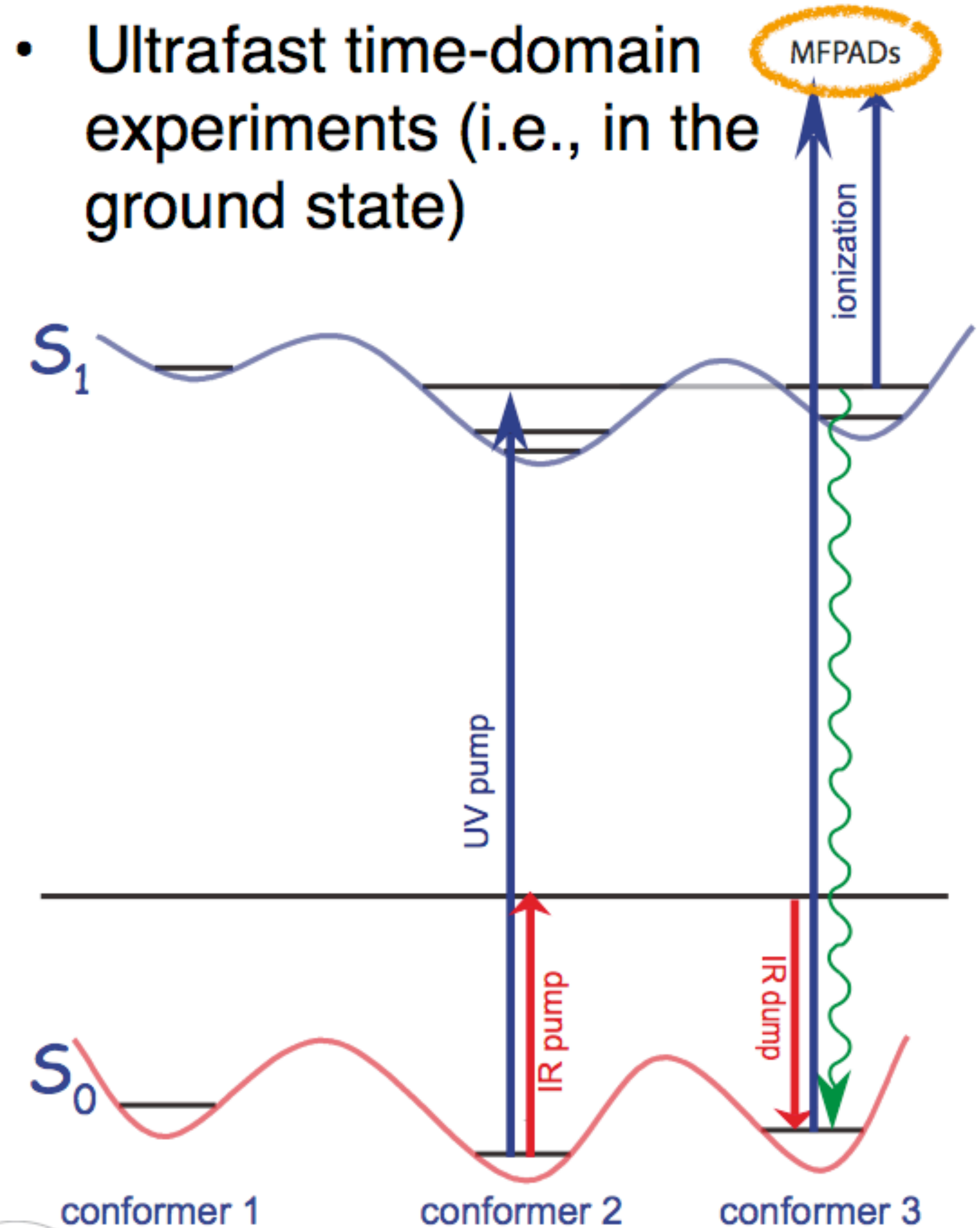
→ **real "*molecular* movie"**

CFEL SCIENCE Controlled Molecule Imaging

Kuleff, Cederbaum, *Chem. Phys.* 338, 321 (2007)

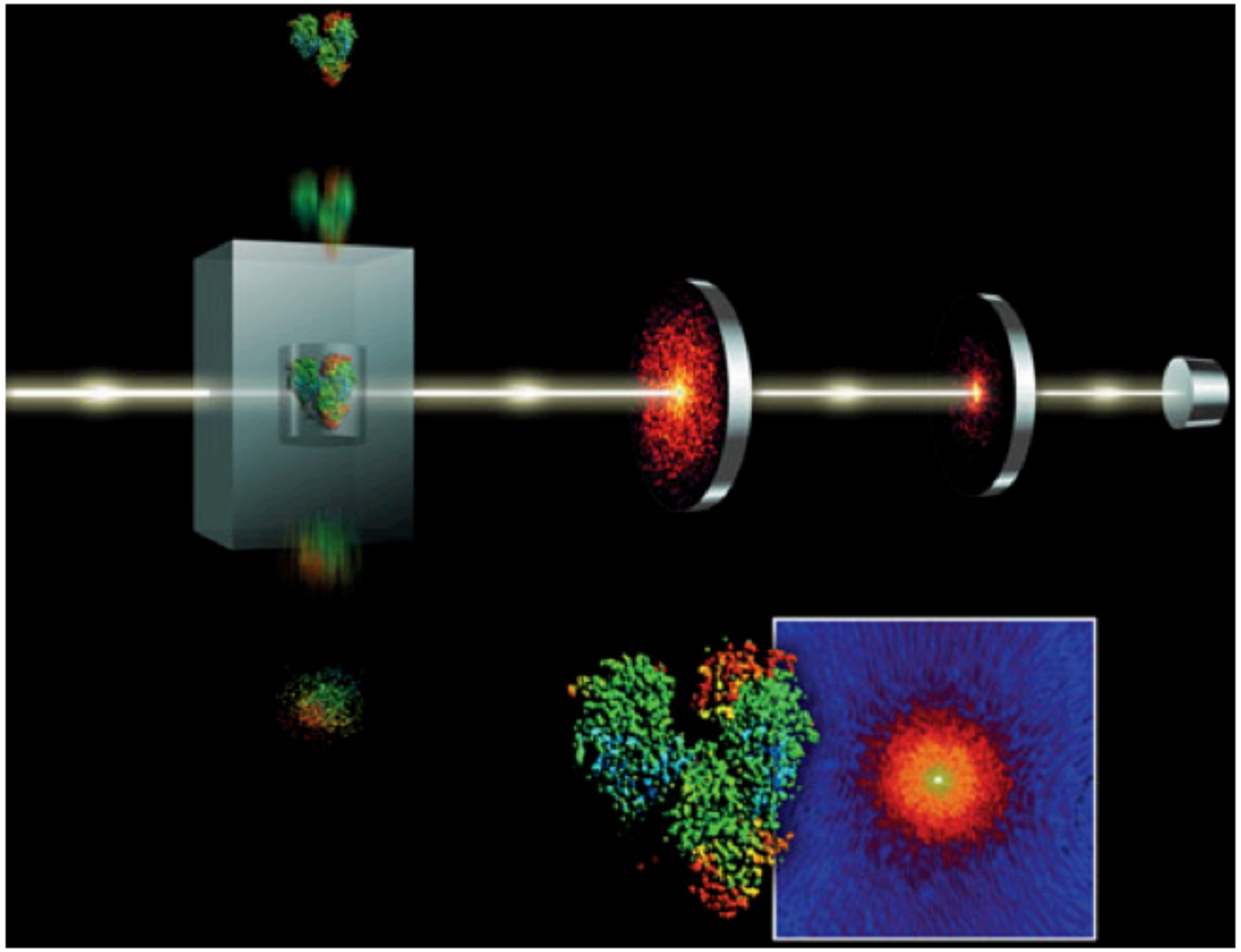
Investigations of controlled large molecules mitigating the requirement of *prior knowledge*

- Ultrafast time-domain experiments (i.e., in the ground state)



Time-resolved (ns) conformer interconversion (population transfer) experiments:
Dian, Clarkson, Zwier, Science, 303, 1169 (2004)

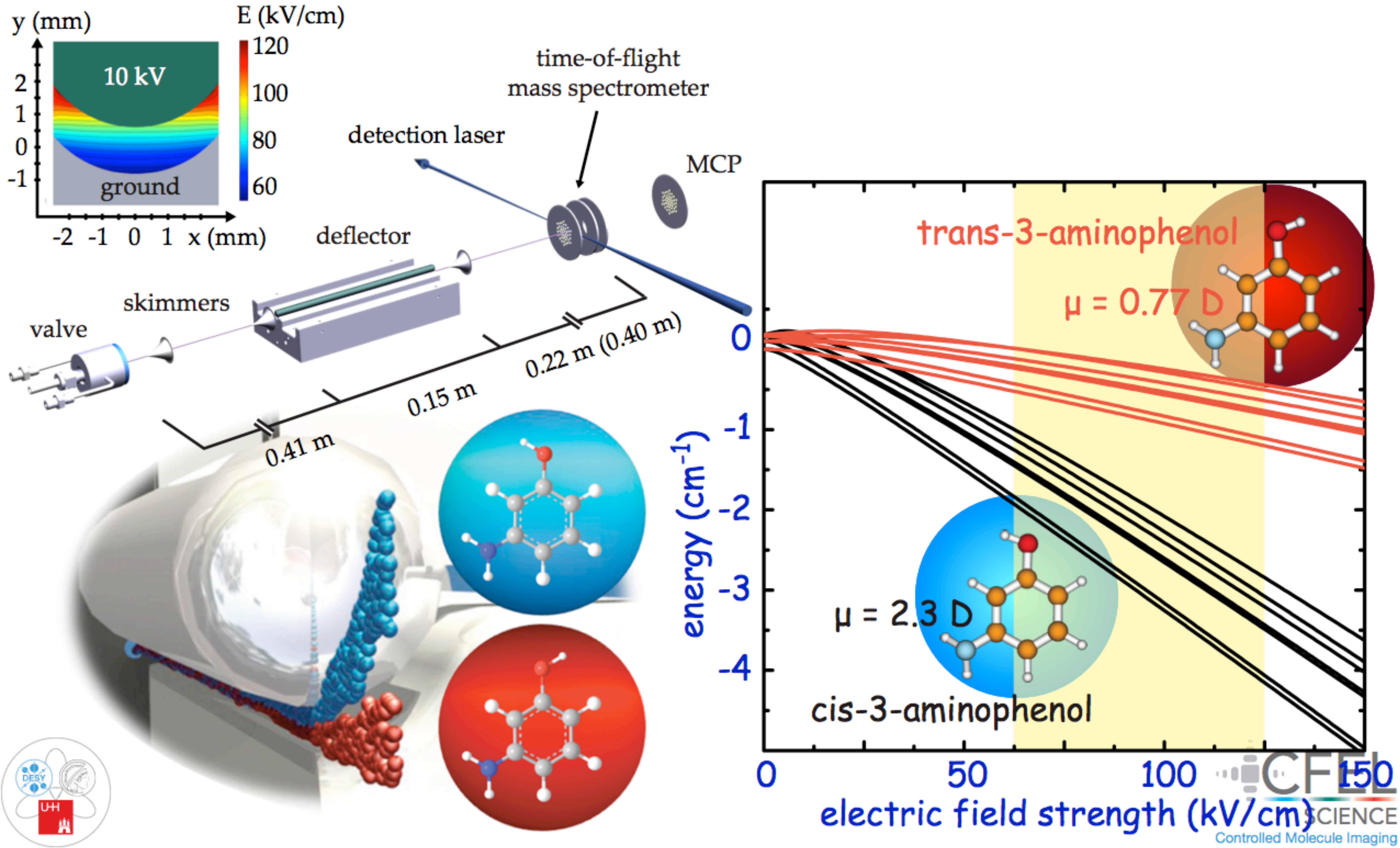
- reaction (stereo-) dynamics
- Imaging
  (Ultrafast X-ray or electron diffraction, "diffraction from within", …)



Controlled Molecule Imaging

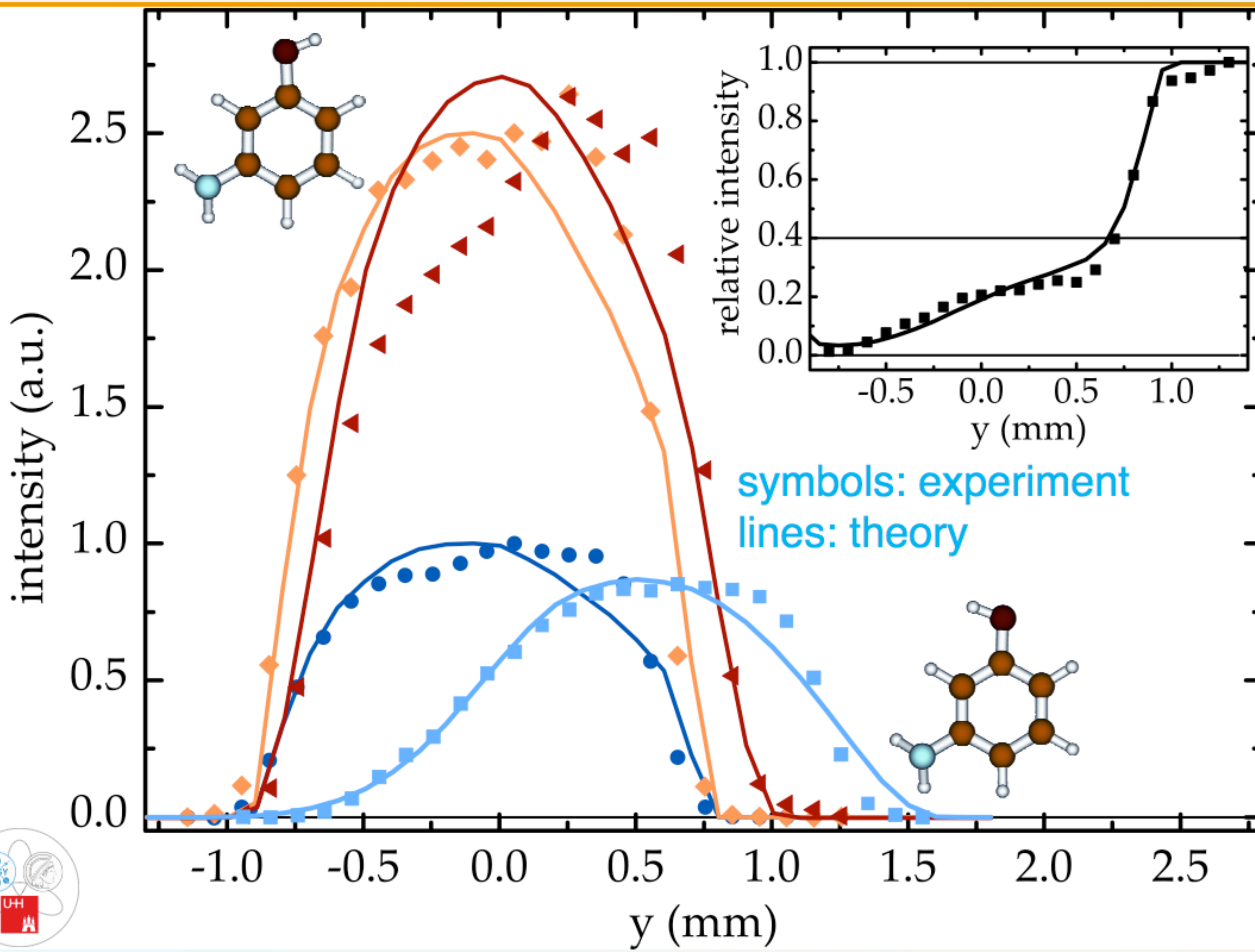
Filsinger, Meijer, Stapelfeldt, Chapman, JK, *Phys. Chem. Chem. Phys.* **13**, 2076 (2011)

m/μ deflector – conformer selection



Filsinger, JK, Meijer, Hansen, Maurer, Nielsen, Holmegaard, Stapelfeldt, *Angew. Chem. Int. Ed.* **48**, 6900 (2009)

m/μ deflector – conformer selection



Filsinger, JK, Meijer, Hansen, Maurer, Nielsen, Holmegaard, Stapelfeldt, *Angew. Chem. Int. Ed.* **48**, 6900 (2009)

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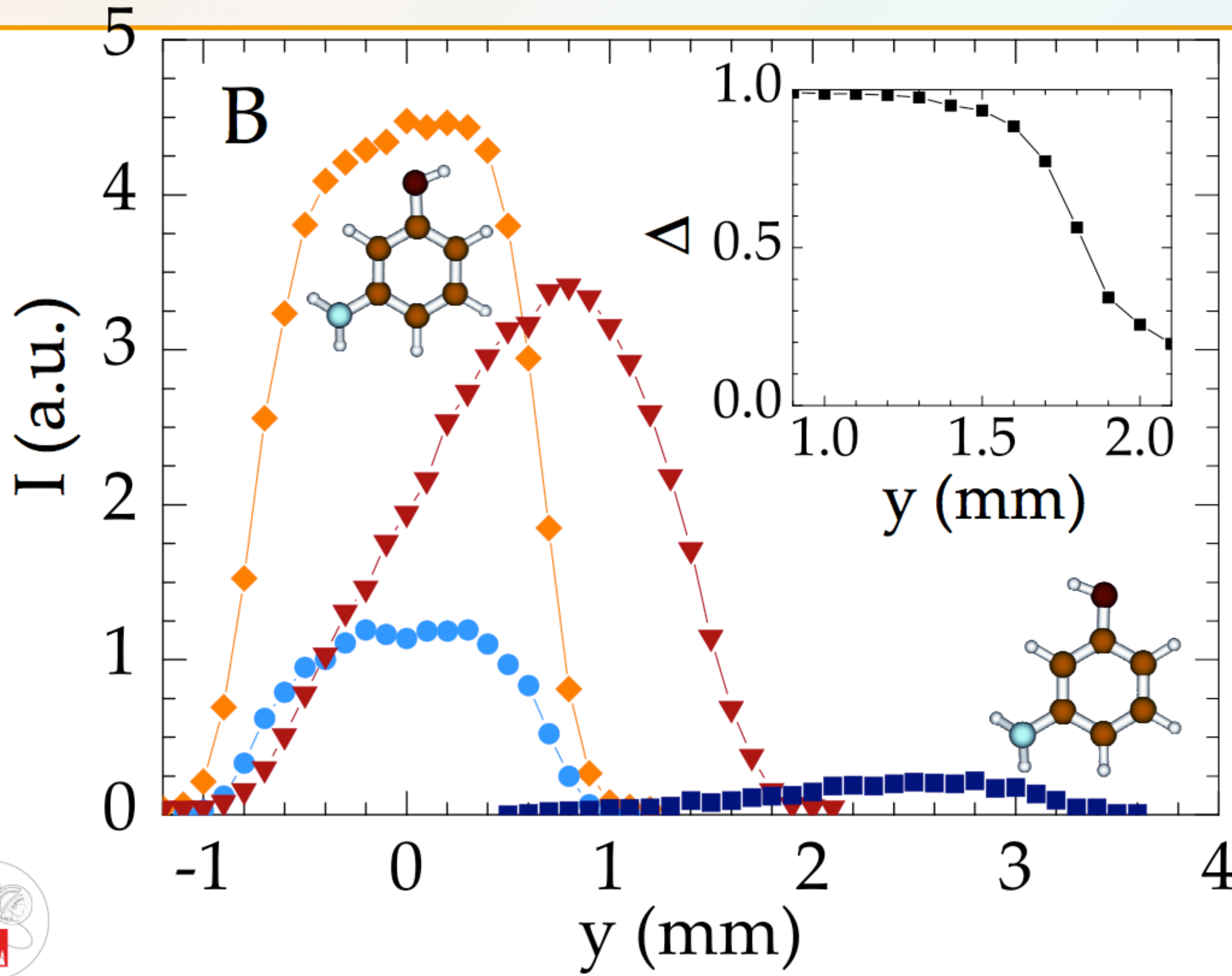
Getting to the other conformers



DESY
UHH
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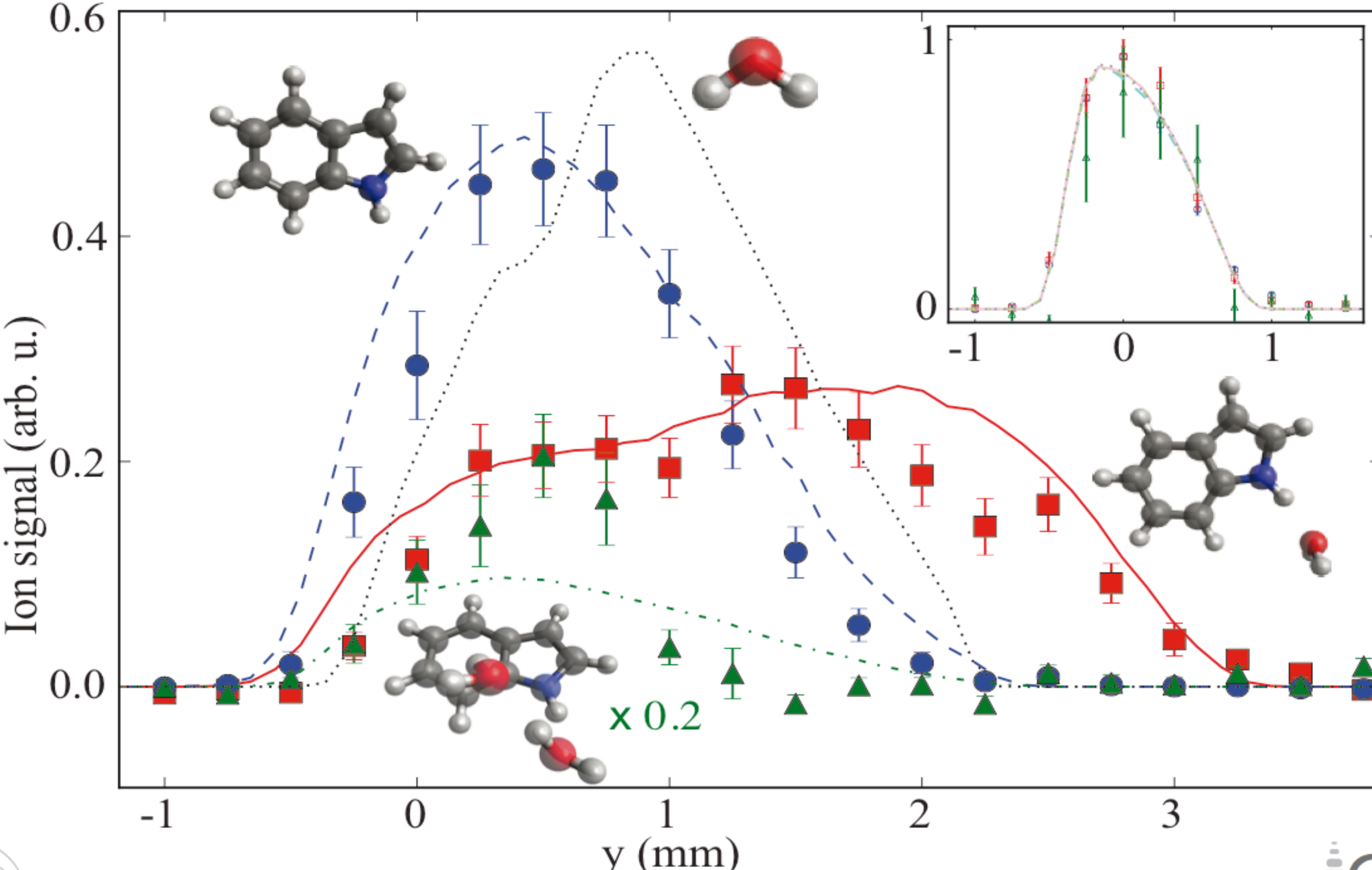
Spatial separation of neutral clusters using the m/μ deflector pure samples of indole-water (indole-$(H_2O)_1$)



Trippel, Chang, Stern, Mullins, Holmegaard, JK, *Phys. Rev.* A **86**, 033202 (2012)

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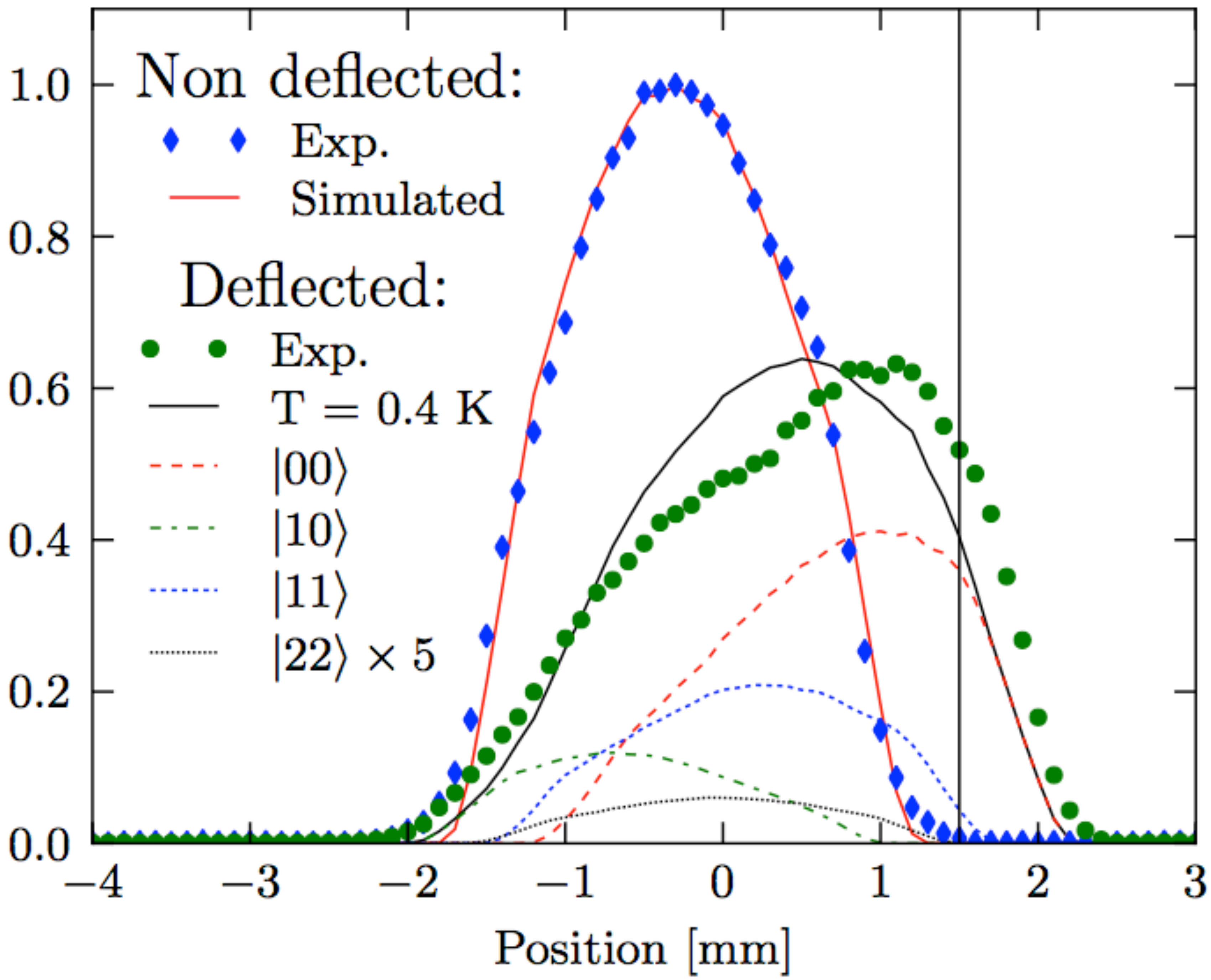
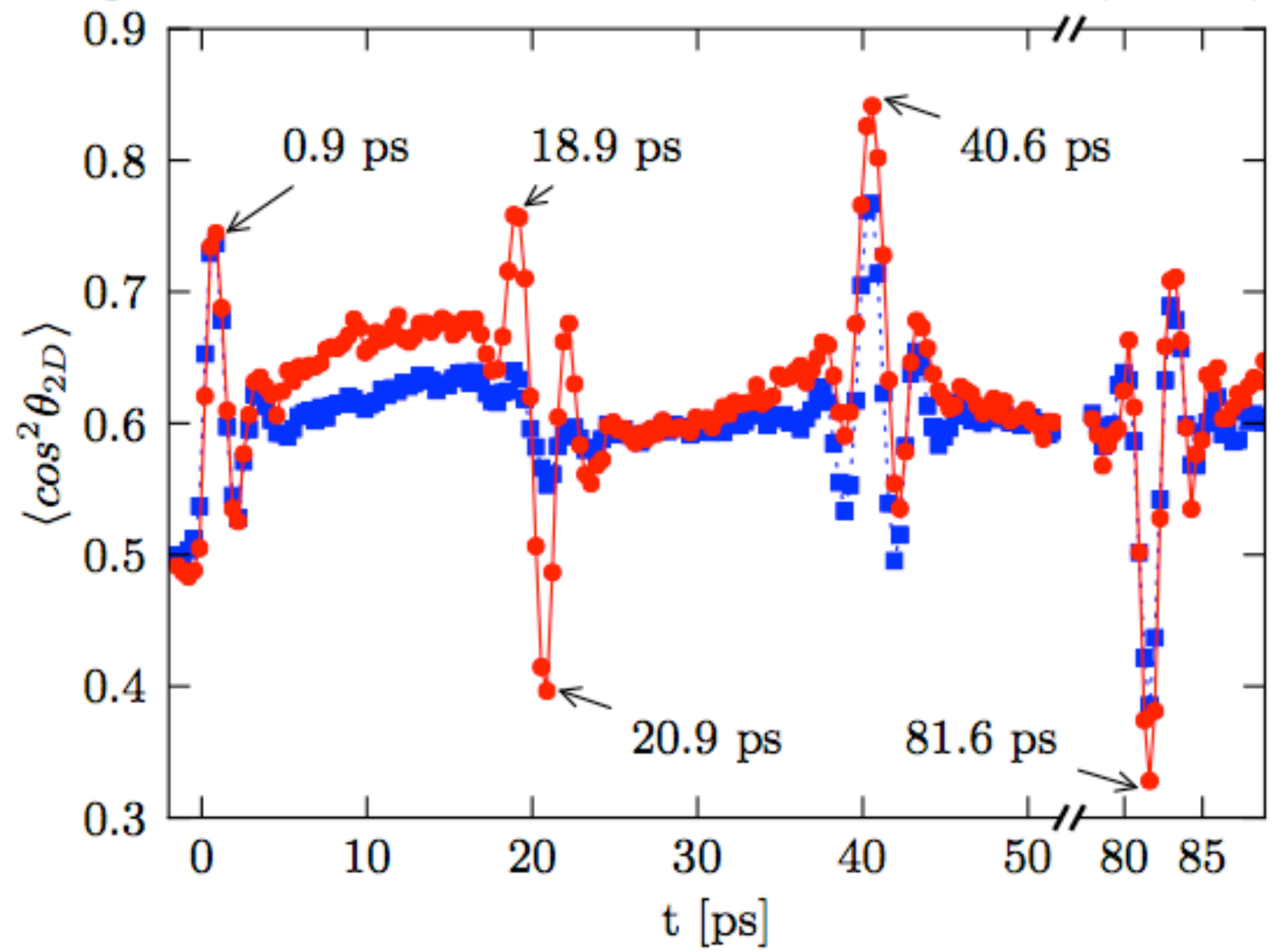
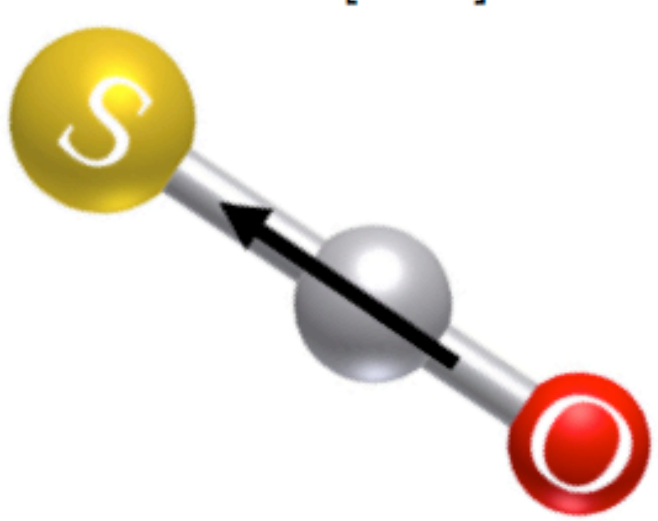
Single-quantum-state molecular ensembles probed by impulsive alignment



quarter revival of
non-inversion-symmetric molecules
only even-J states contribute (J=0)





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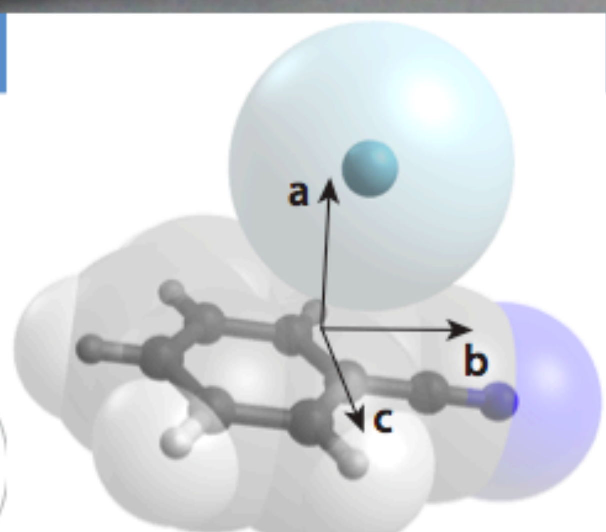
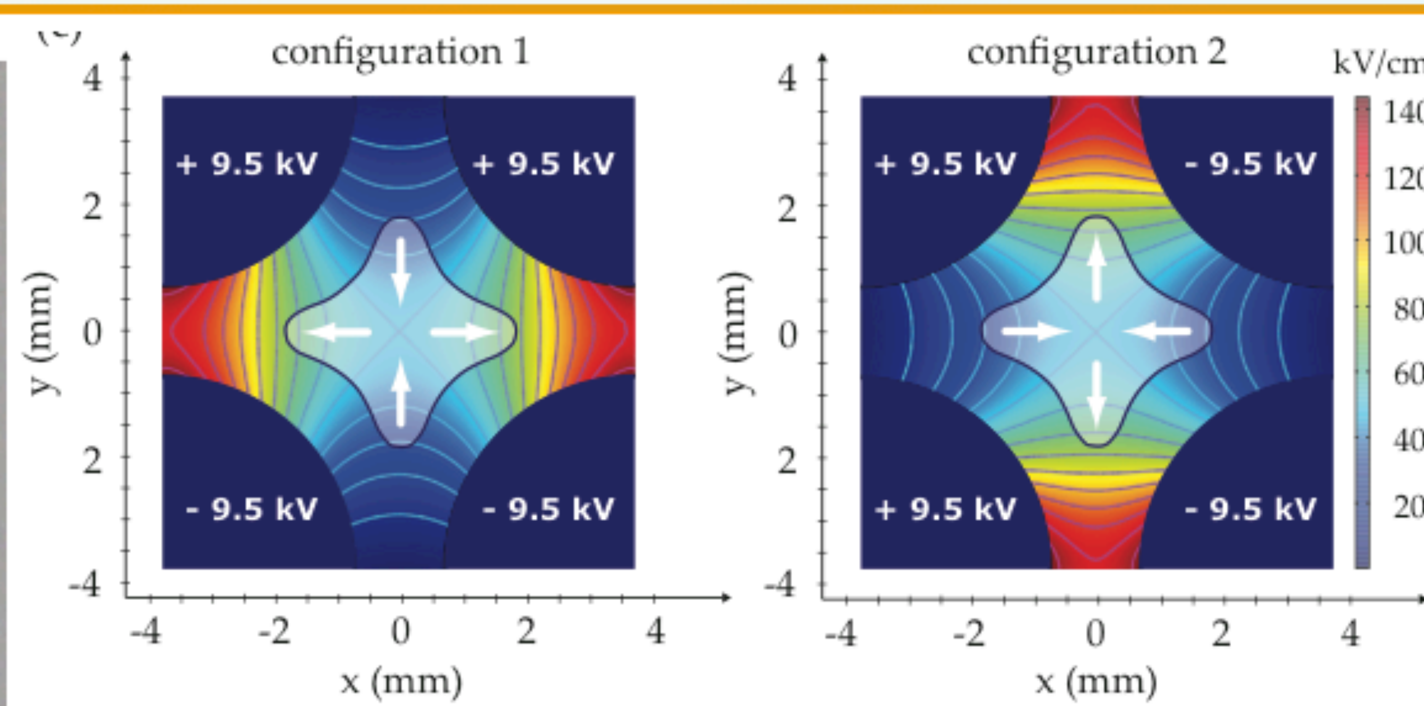
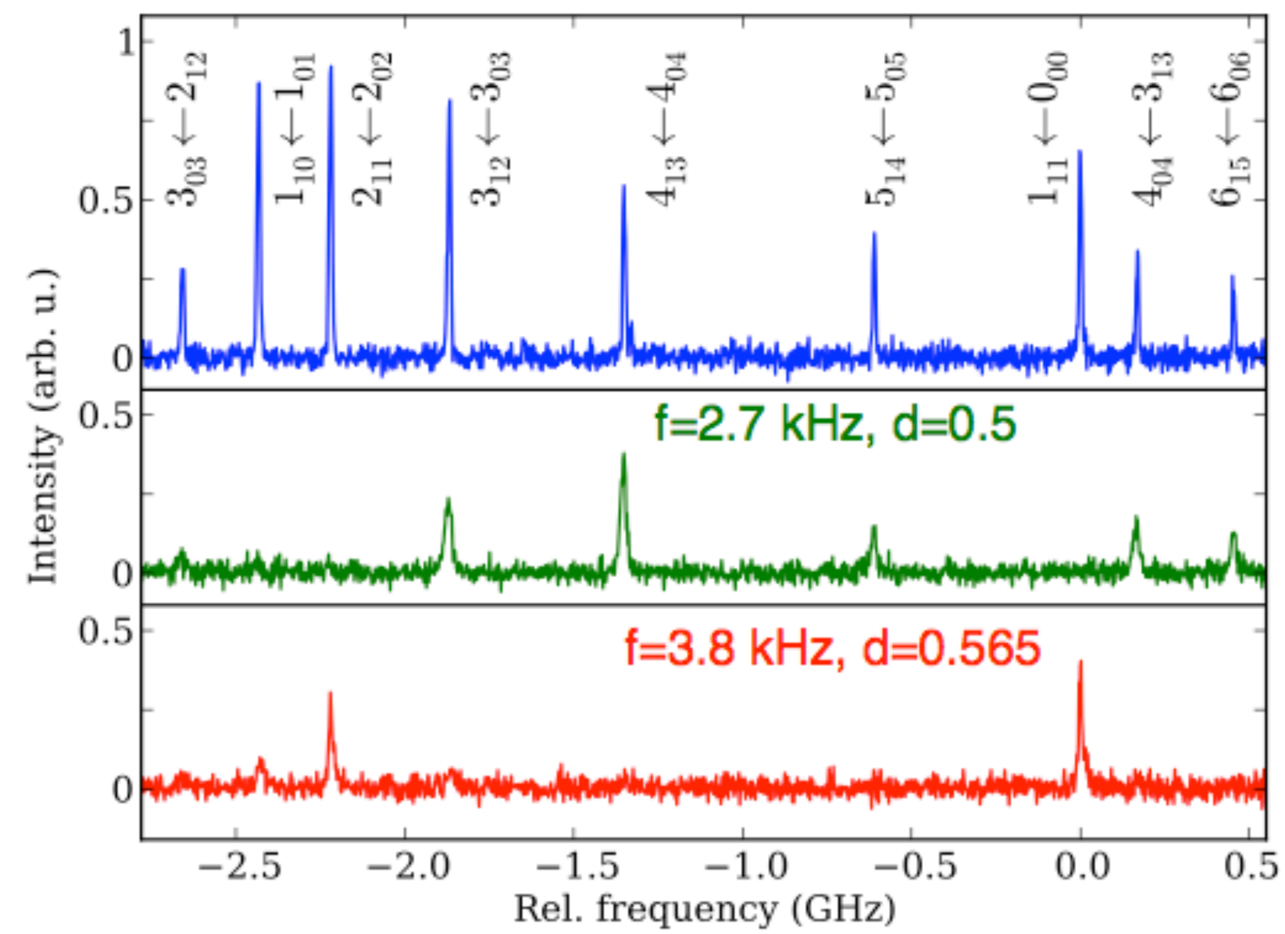
Nielsen, Simesen, Bisgaard, Stapelfeldt, Filsinger, Friedrich, Meijer, JK, *Phys. Chem. Chem. Phys.* 13, 18971–18975 (2011)

State- and size-selective alternating-gradient focusing of benzonitrile and the benzonitrile-argon cluster







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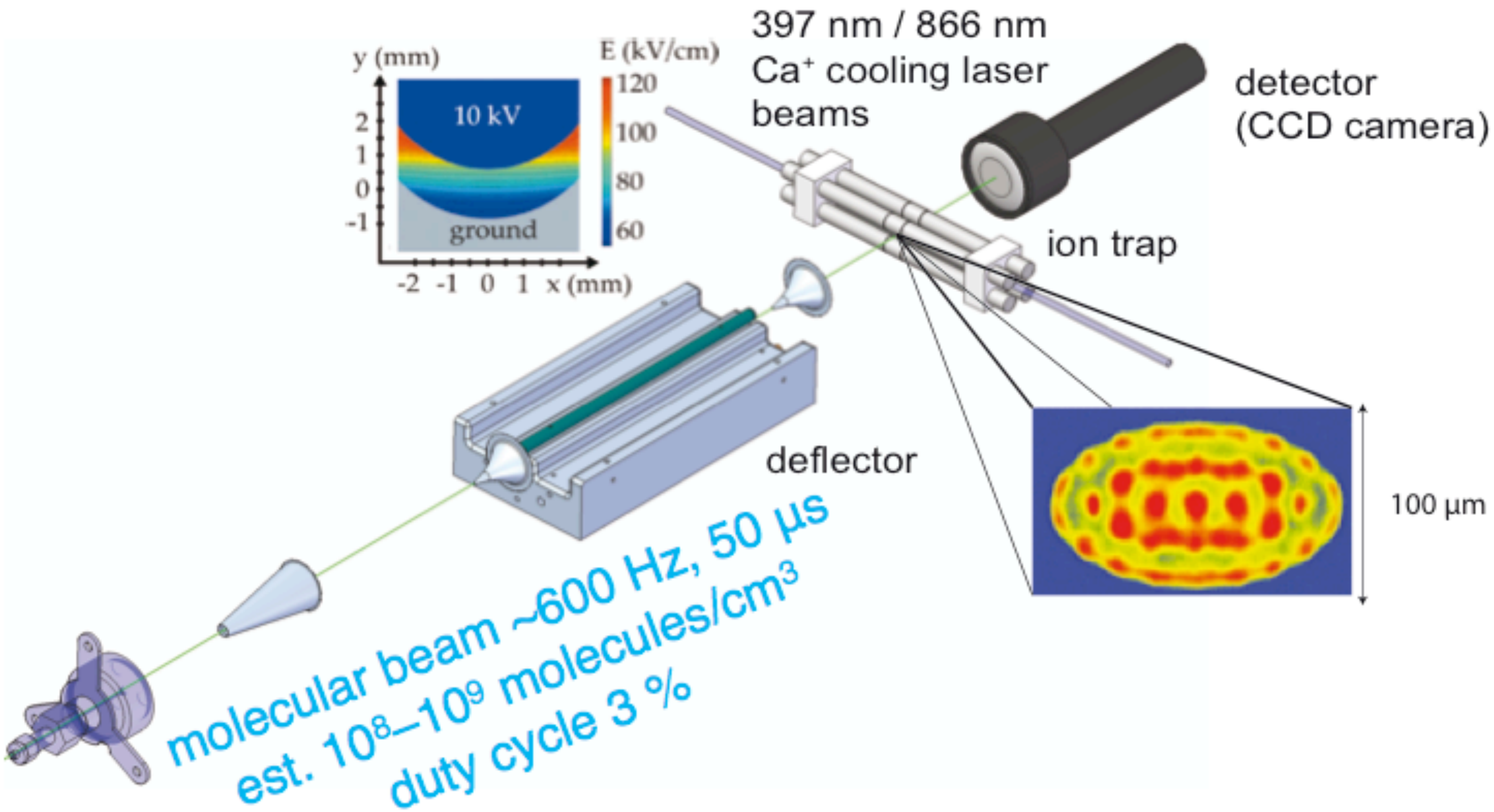
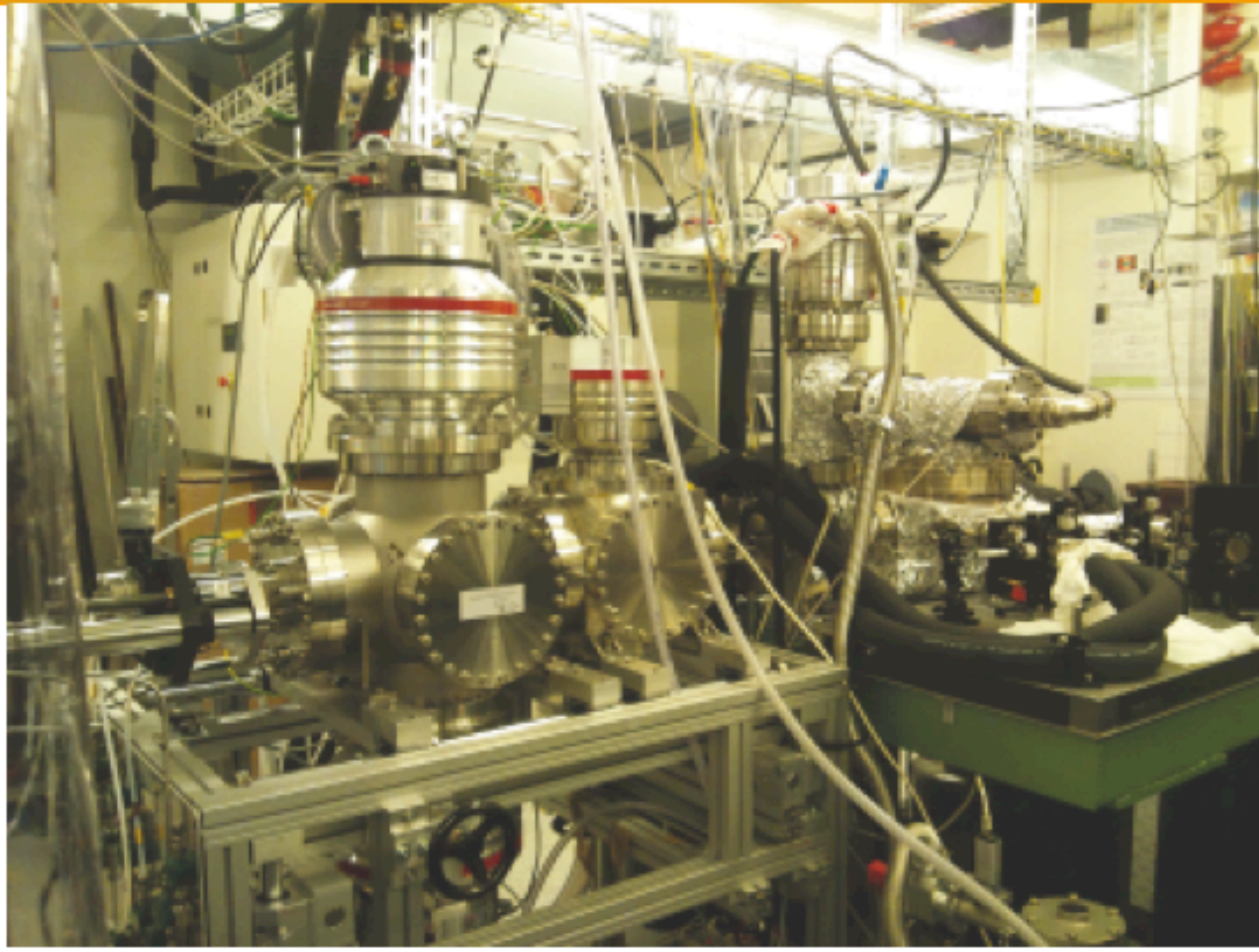
Filsinger, Erlekam, von Helden, JK, Meijer, *Phys. Rev. Lett*. **100**,133003 (2008)

Putzke, Filsinger, Haak, JK, Meijer, *Phys. Chem. Chem. Phys.* **13**, 18962 (2011) & *J. Chem. Phys.* **137**, 104310 (2012)

Conformer-specific rates of chemical reactions





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see our poster for details

CFEL SCIENCE Controlled Molecule Imaging

Rösch, Willitsch, Chang, Długołęcki, JK, *in preparation*

Manipulating the motion of “large molecules”
Optically controlled particles – polystyrene beads at atmosphere

our first experiment
in the new CFEL lab!
December 2013

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***Photophoresis*:**

P. Lebedew, “Untersuchungen über die Druckkräfte des Lichtes” *Ann. Phys.* **311**, 433 (1901)

F. Ehrenhaft, “Die Photophorese” *Ann. Phys.* **361**, 81 (1918)

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Controlled Molecule Imaging

Shvedov, Desyatnikov, Rode, Krolikowski, Kivshar, *Opt. Exp.* **17**, 5743 (2009)

Manipulating the motion of “large molecules”
Beams of proteins, viruses, or cells (or 2 µm polystyrene beads)

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Manipulating the motion of “large molecules”
Optically controlled particles – polystyrene beads in “vacuum”

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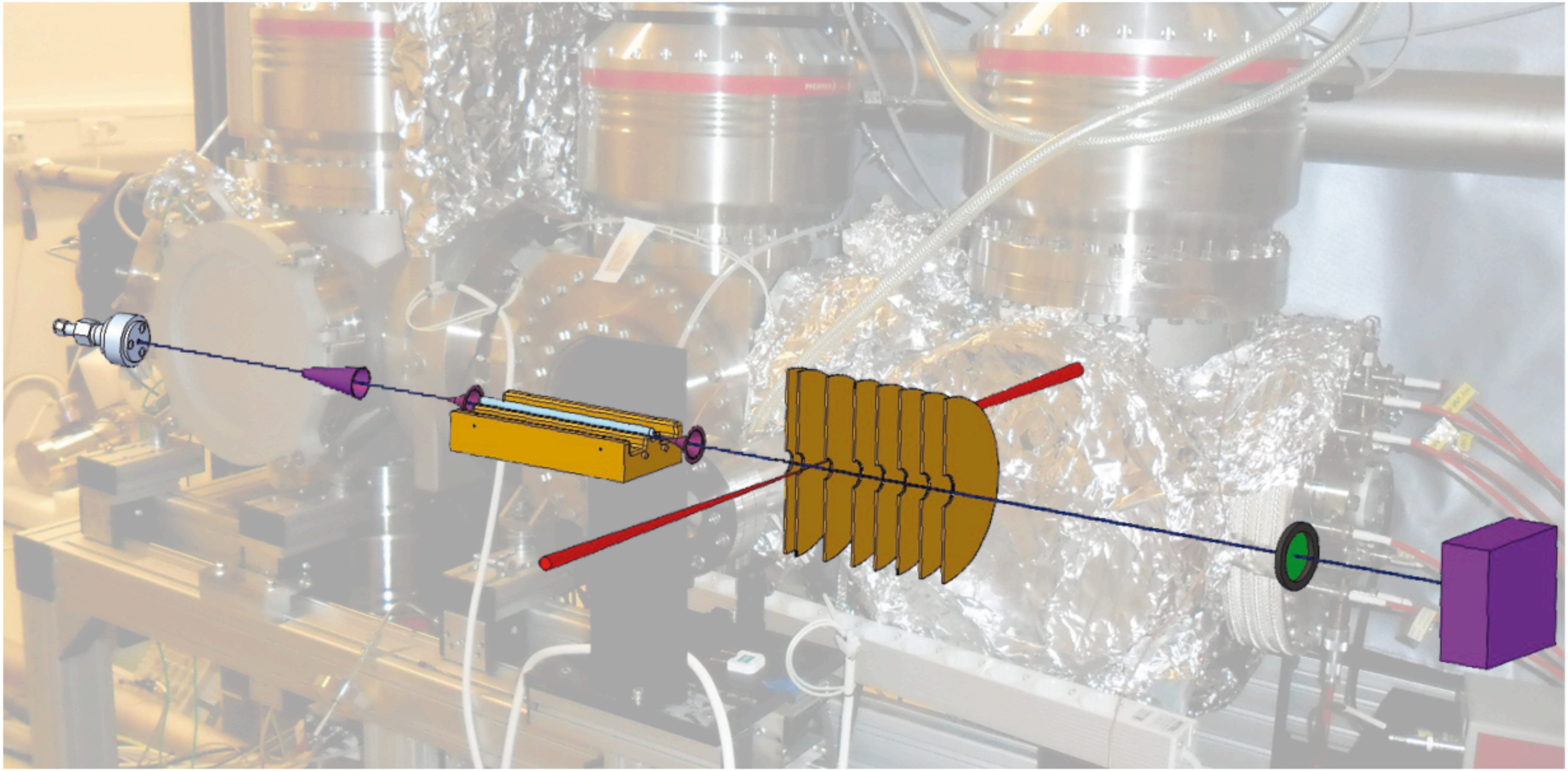
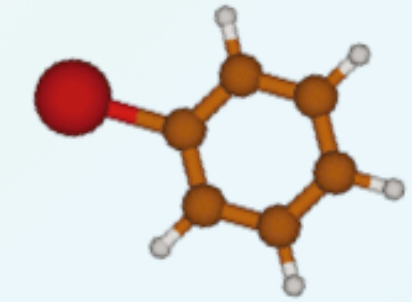
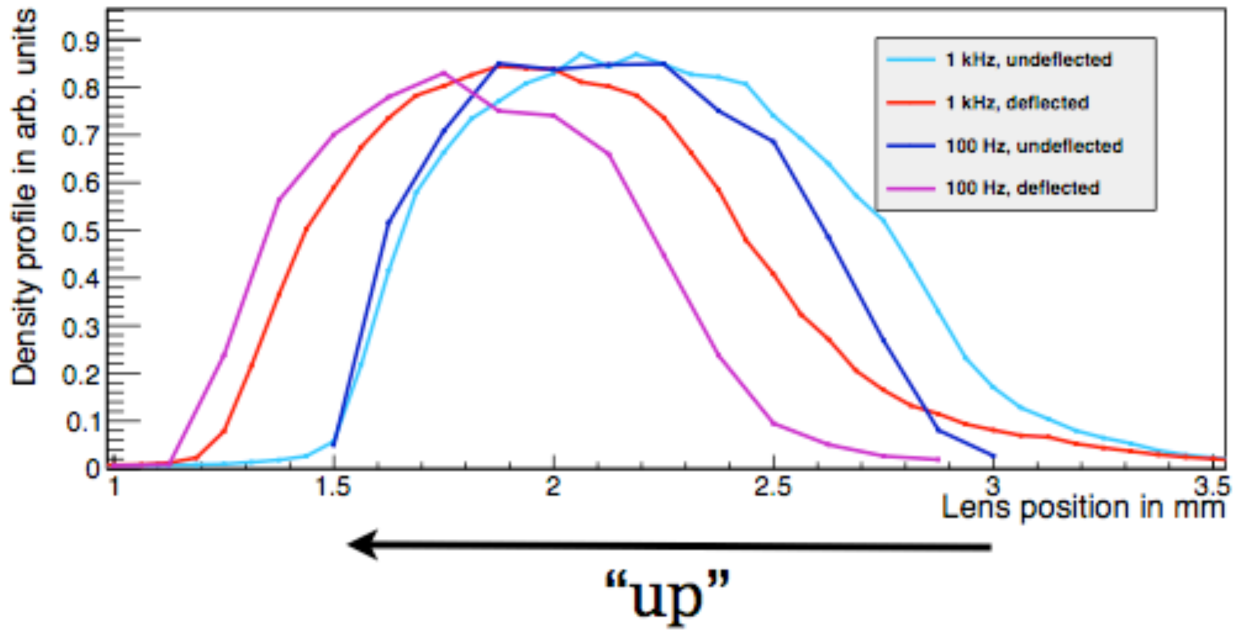
Toward the investigating of chemical dynamics kHz-rate manipulation experiments

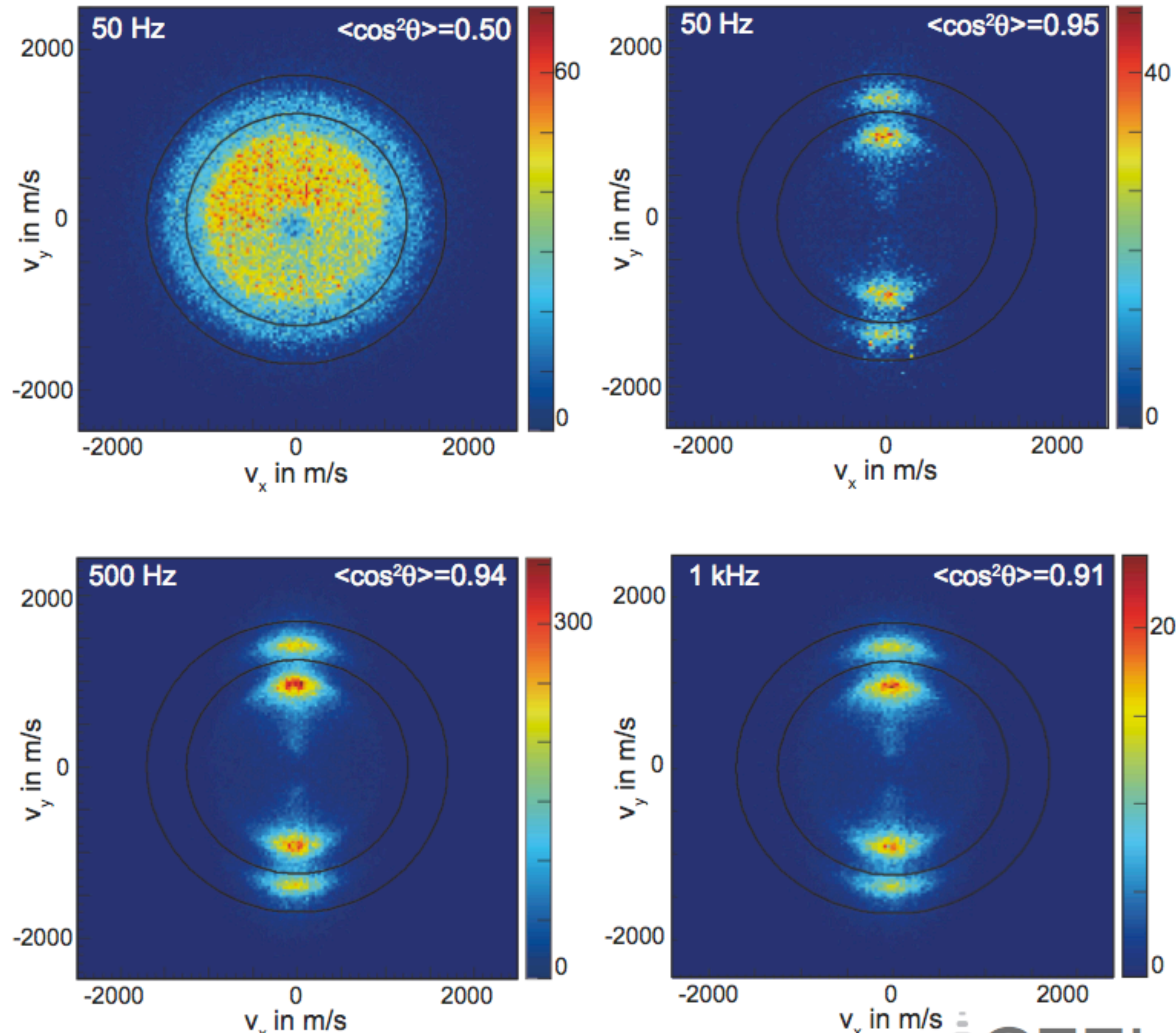


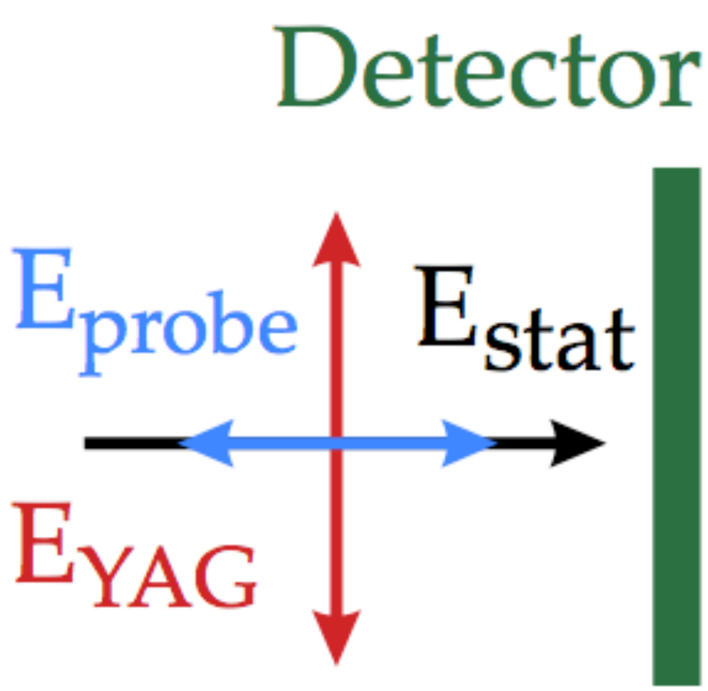
Trippel, Mullins, Müller, Kienitz, Długołęcki, JK, *Mol. Phys.,* in press (2013) DOI: 10.1080/00268976.2013.780334

Toward the investigating of chemical dynamics kHz-rate manipulation experiments – *alignment*







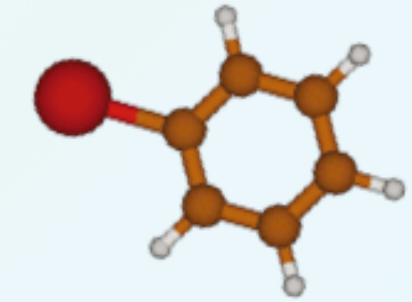


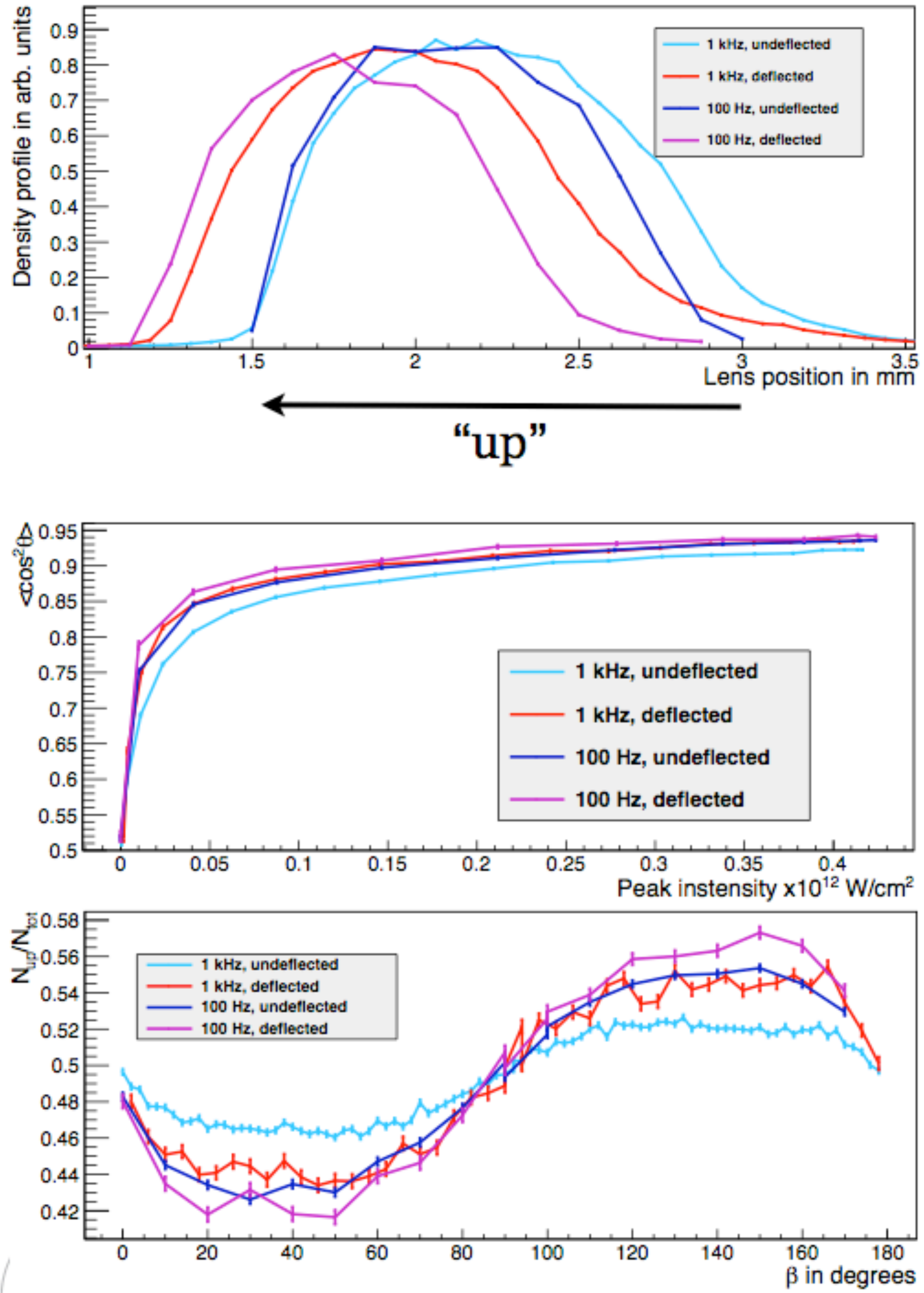
cf. "world record" of <$\cos^2\theta$>=0.97 @ 20 Hz
Holmegaard, Nielsen, Nevo, Stapelfeldt, Filsinger, JK, Meijer, *Phys. Rev. Lett.* **102**, 023001, (2009)

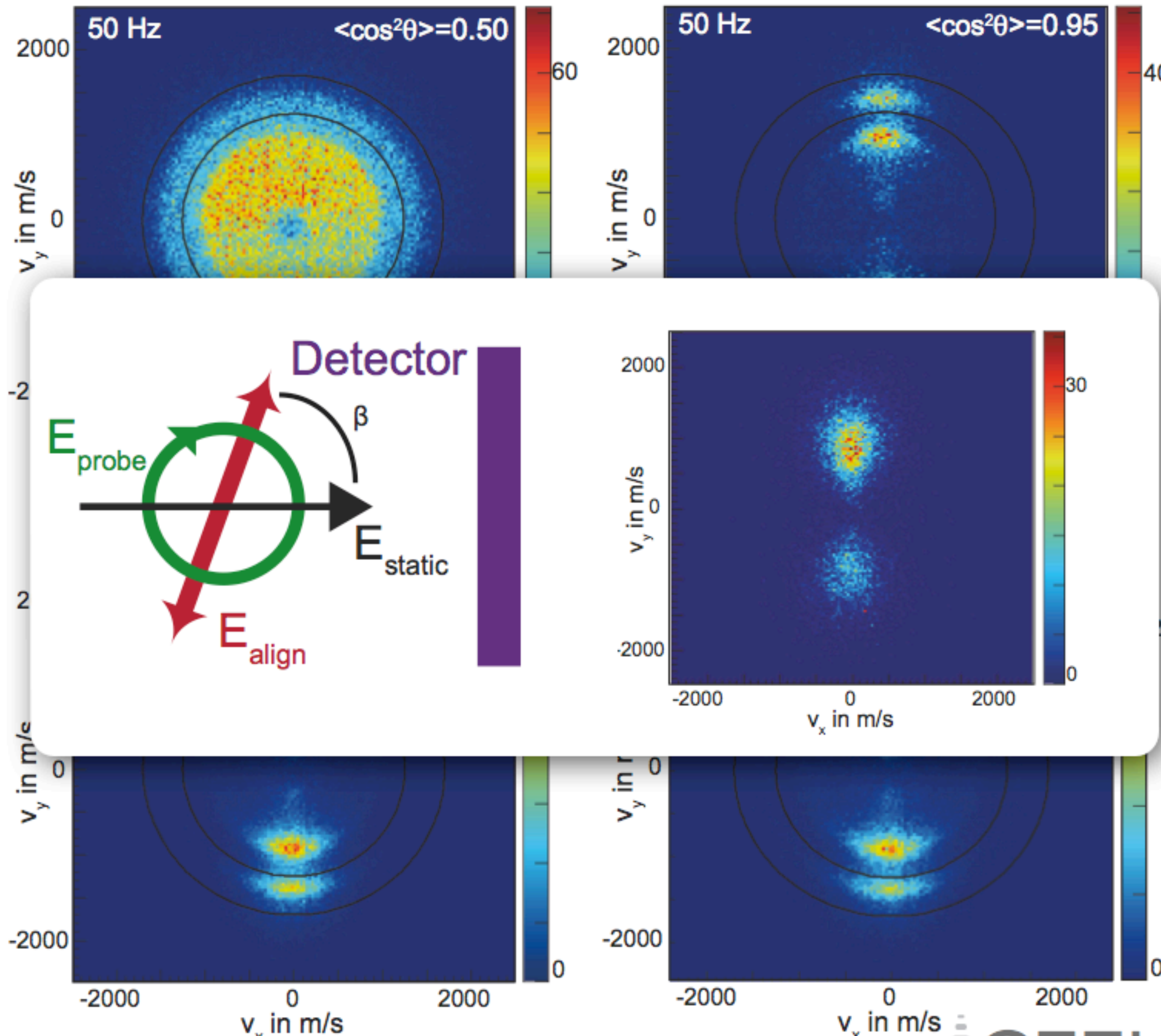
CFEL SCIENCE
Controlled Molecule Imaging

Trippel, Mullins, Müller, Kienitz, Długołęcki, JK, *Mol. Phys.*, in press (2013) DOI: 10.1080/00268976.2013.780334

Toward the investigating of chemical dynamics kHz-rate manipulation experiments – *alignment*







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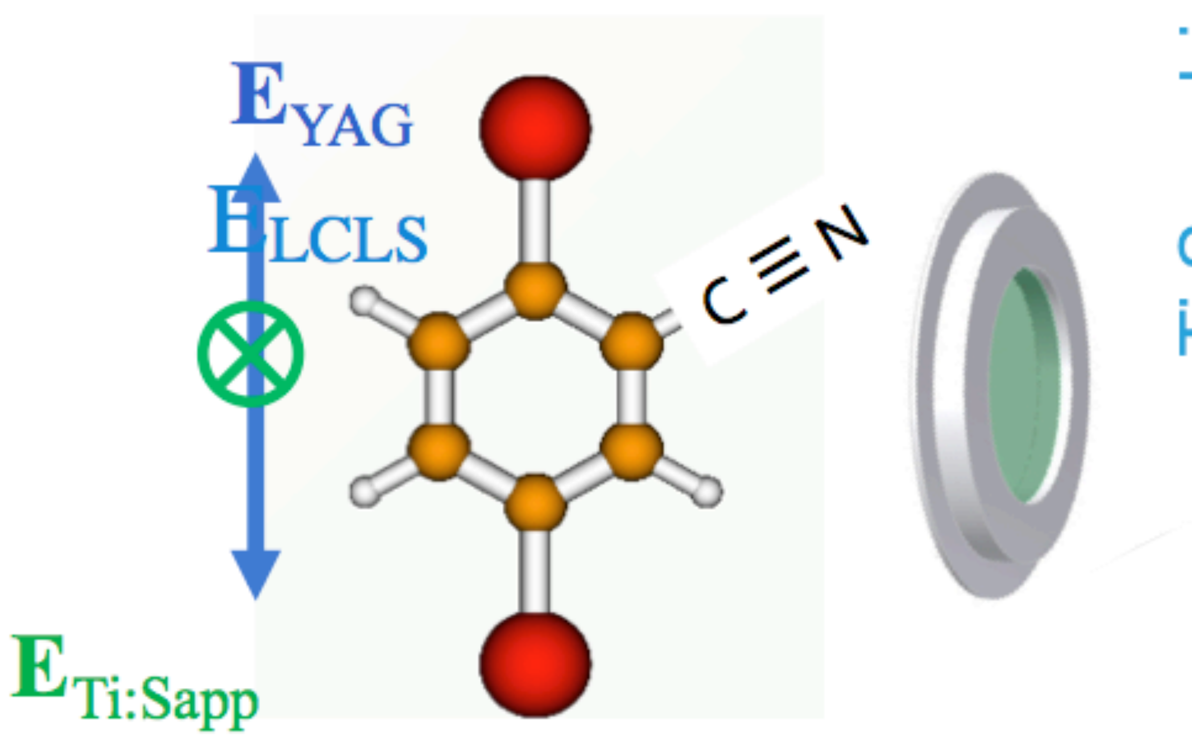
Holmegaard, Nielsen, Nevo, Stapelfeldt, Filsinger, JK, Meijer, *Phys. Rev. Lett.* **102**, 023001, (2009)

CFEL SCIENCE Controlled Molecule Imaging

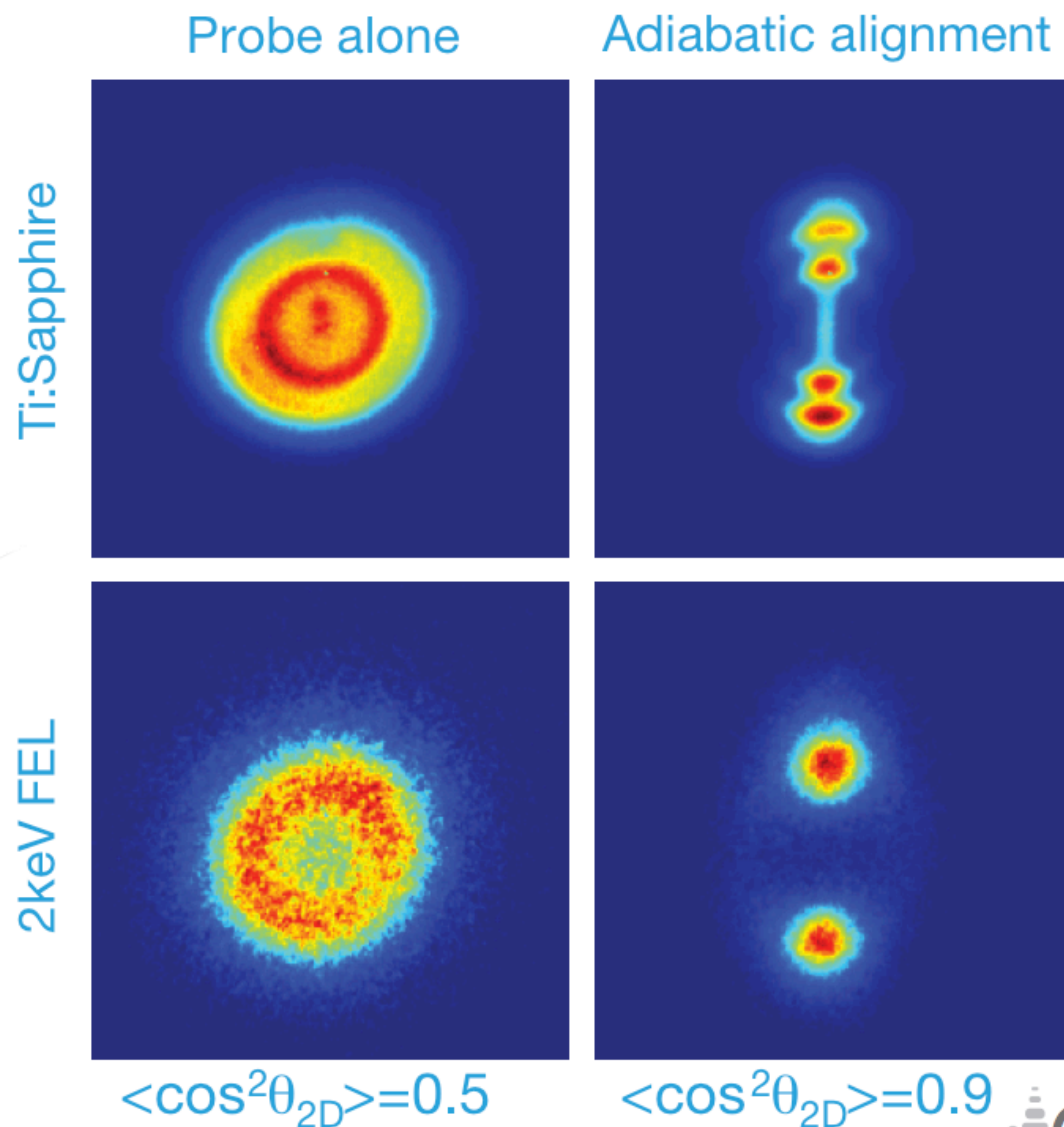
Trippel, Mullins, Müller, Kienitz, Długołęcki, JK, *Mol. Phys.*, in press (2013) DOI: 10.1080/00268976.2013.780334

Strongly aligned complex molecules at the LCLS (X-FEL) *"laser-lab on the highway"* setting

velocity-map imaging of I^+ ions



far-off-resonant x-rays are an "ideal probe" of angular confinement



radial distributions provide information on "*radiation damage*"

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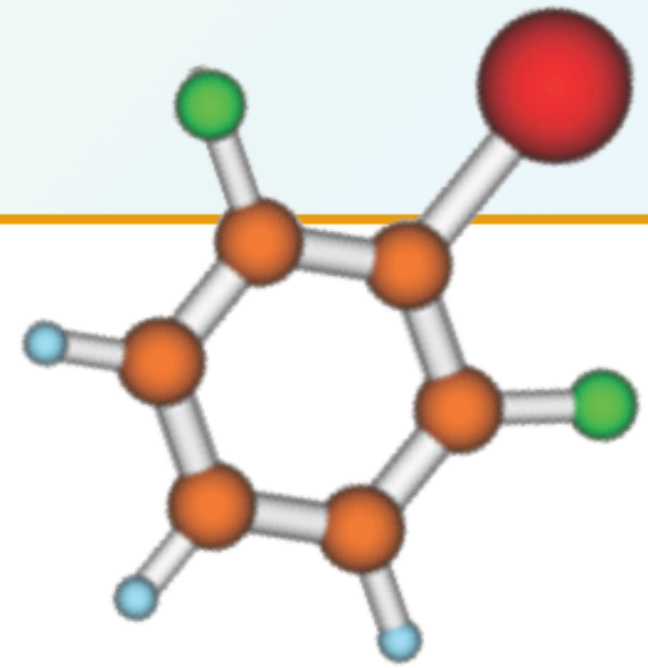
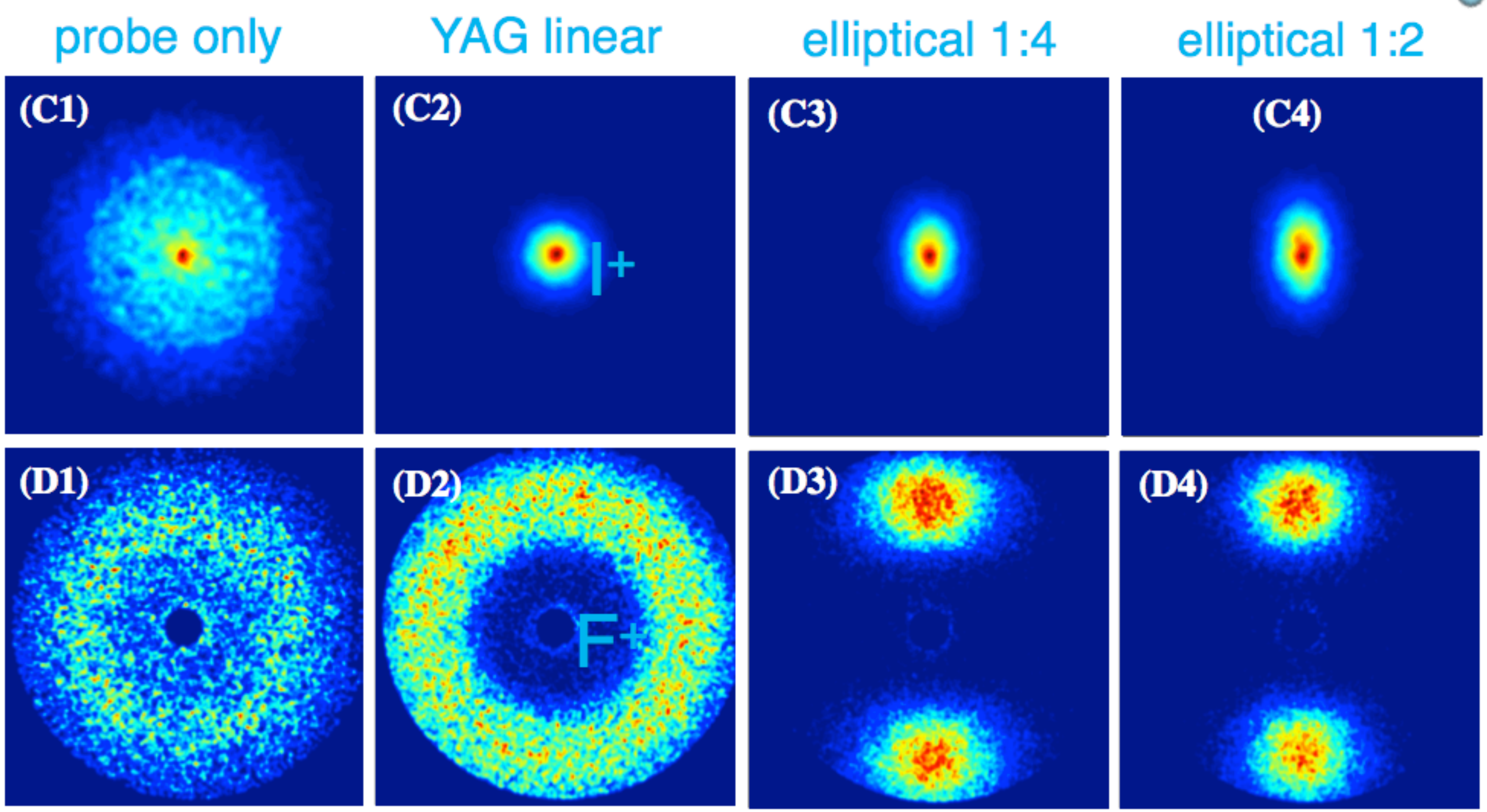
Controlled Molecule Imaging

JK, Stern, Holmegaard *et al* (52 authors), *under internal review*

3D alignment and orientation of 2,6-difluoro-iodobenzene





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Nevo, Holmegaard, Nielsen, Hansen, Stapelfeldt, Filsinger, Meijer, JK, *Phys. Chem. Chem. Phys.* **11**, 9912 (2009)

Imaging experiments at (X)FELs and at *home*
... *seeing* the dynamics of complex molecules

- Fragment ion spectra and momentum distributions
- Photoelectron spectra
- Molecular frame photoelectron angular distributions
  - “diffraction from within”, “photoelectron holography”
- “Coherent” X-ray and electron diffractive imaging
- of *state-selected* and *strongly oriented* molecular ensembles
- of *individual cluster sizes*
- of *individual structural isomers*

with as/fs/ps time resolution

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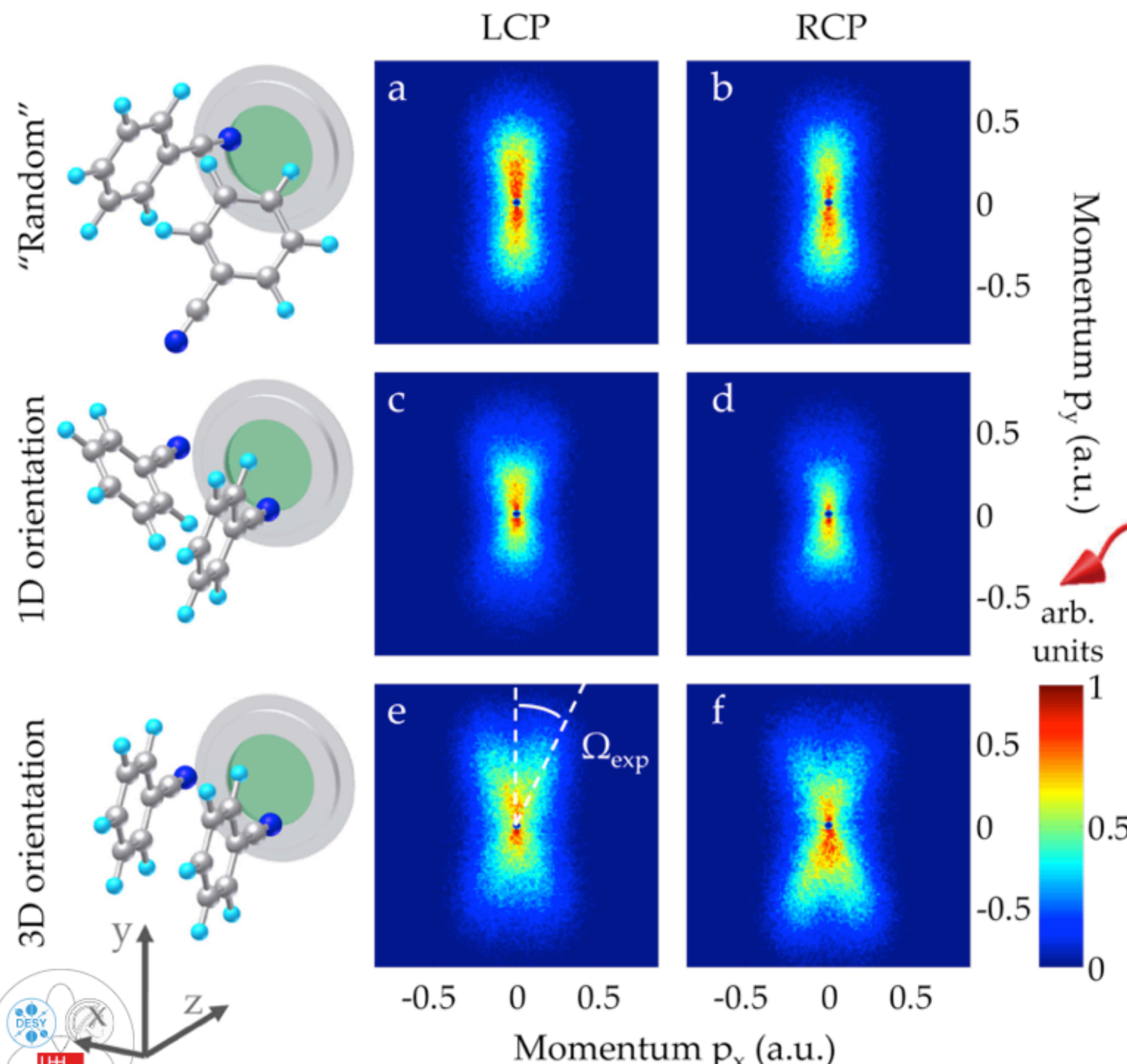
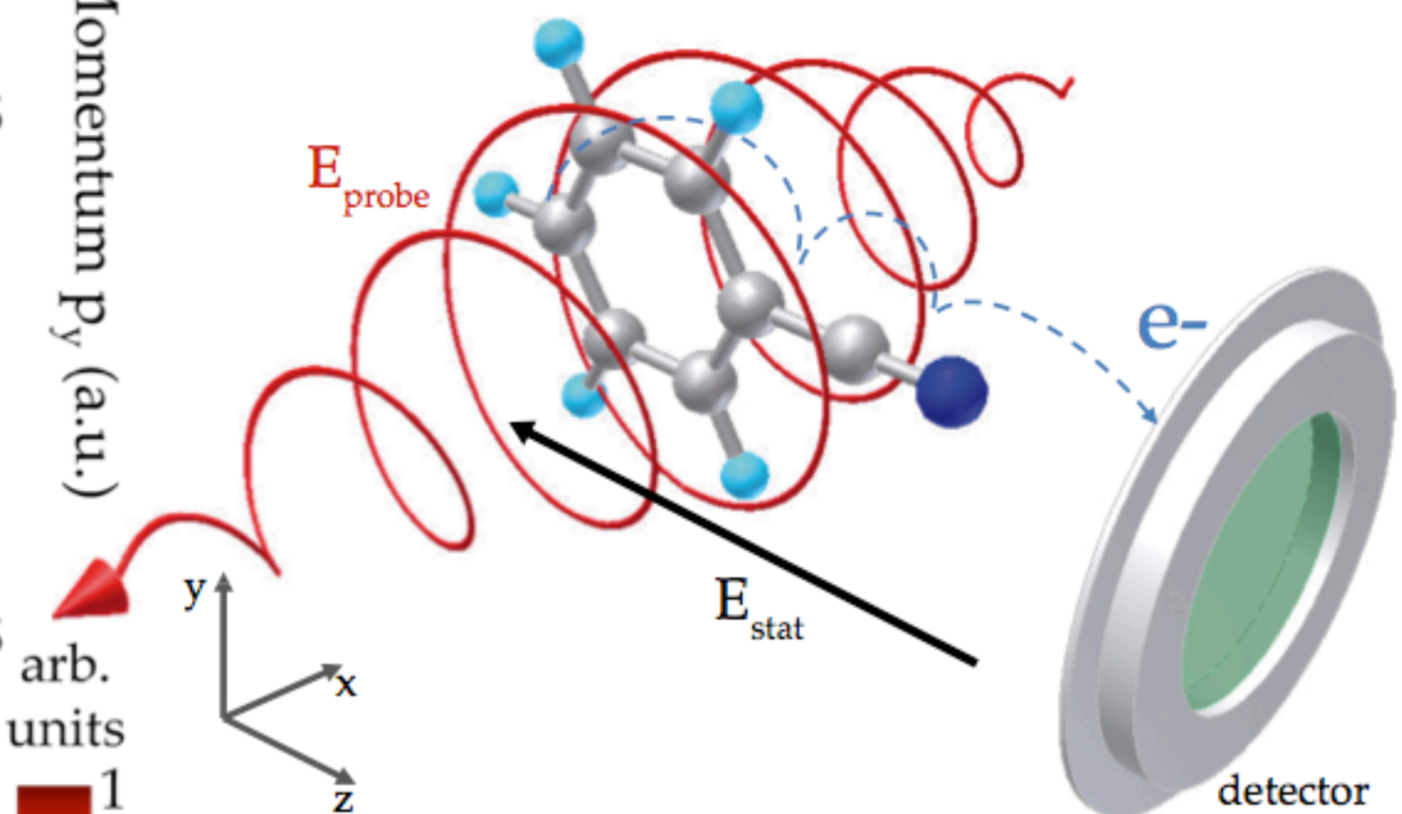
Controlled Molecule Imaging

Molecular frame photoelectron angular distributions of 3D-oriented large molecules: benzonitrile





E_{stat} = 467 V/cm

E_{align} = 7 × 10^{11} W/cm²

E_{probe} = 1.2 × 10^{14} W/cm²

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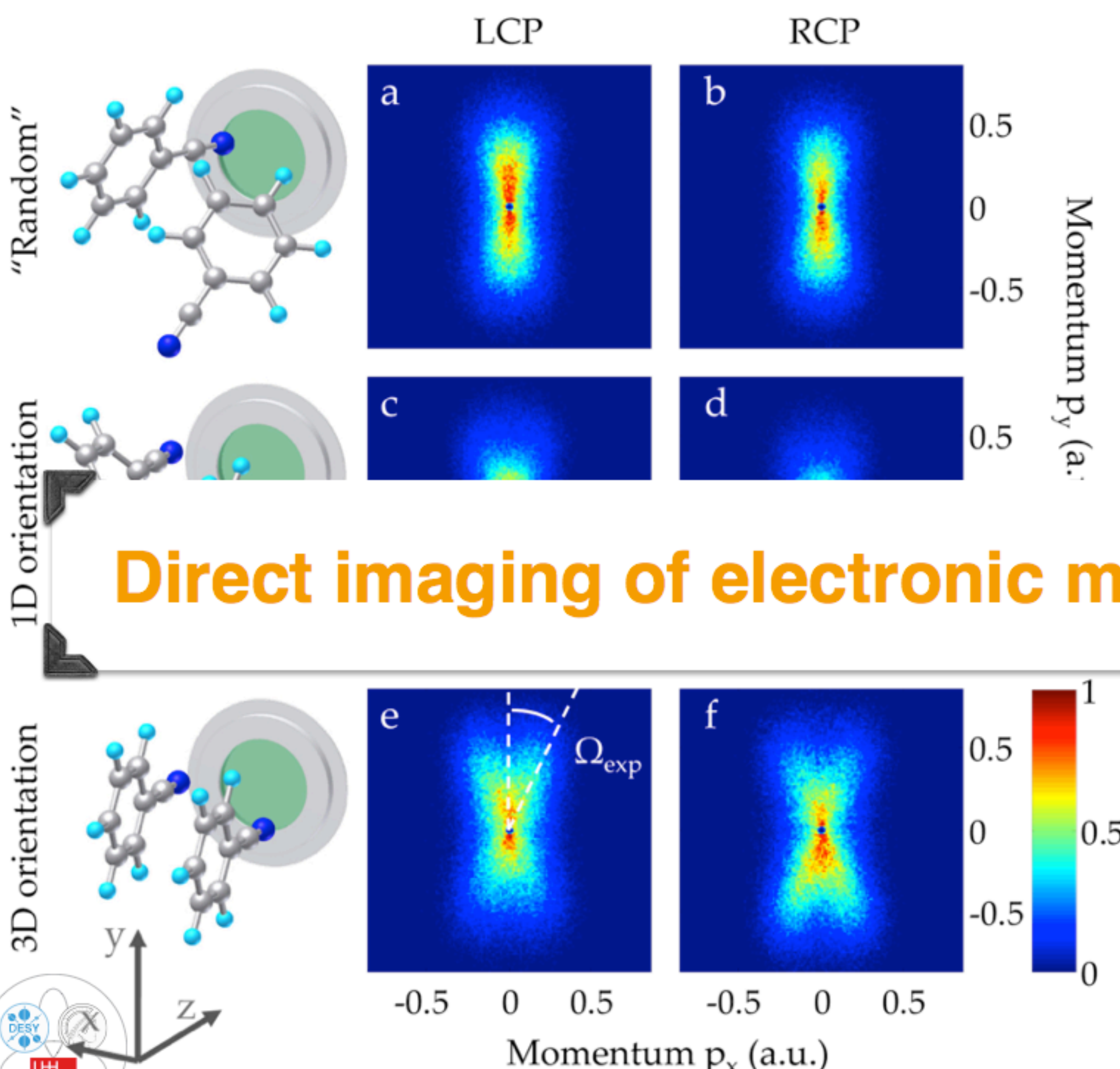
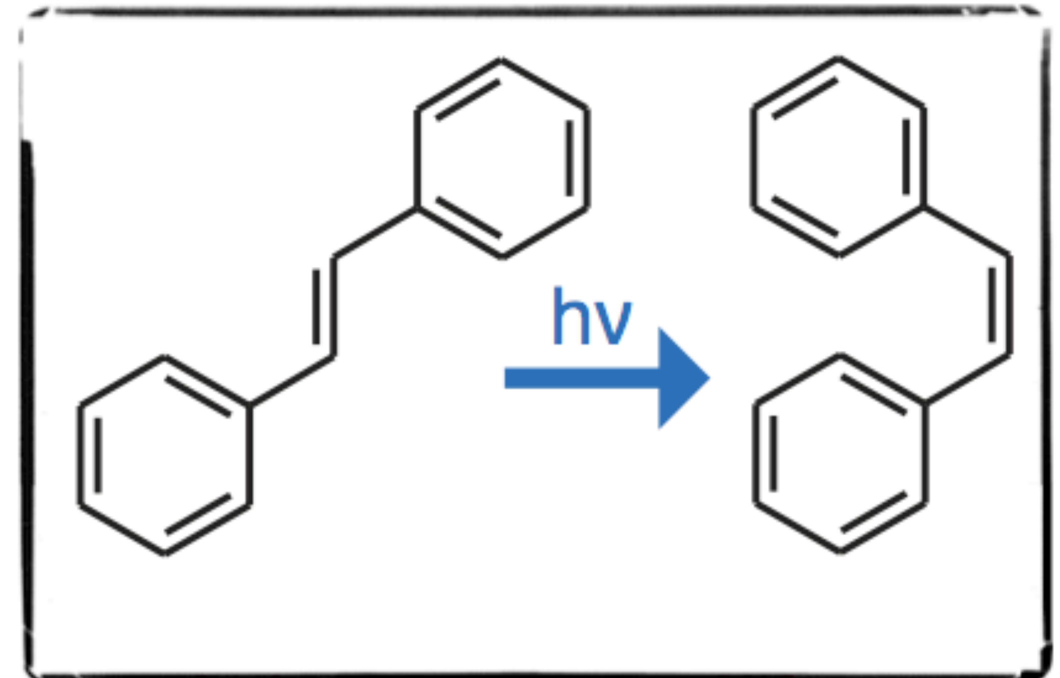
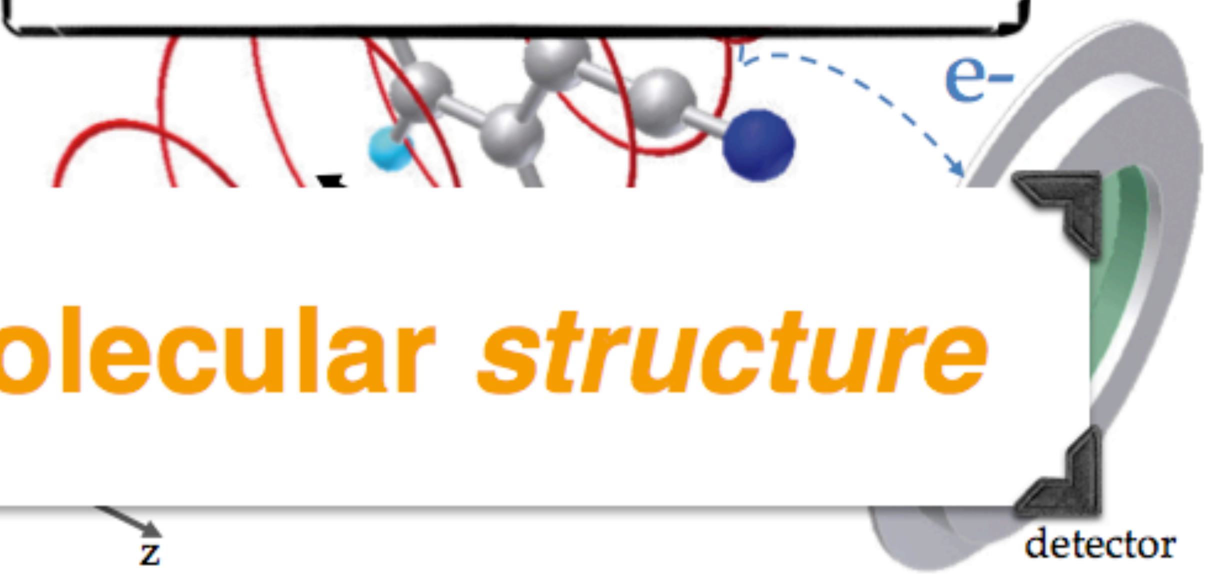
Controlled Molecule Imaging

Holmegaard, Hansen, Kalhøj, Kragh, Stapelfeldt, Filsinger, JK, Meijer, Dimitrovski, Abu-samha, Martiny, Madsen, *Nature Phys.* **6**, 428 (2010)

Molecular frame photoelectron angular distributions of 3D-oriented large molecules: benzonitrile







Direct imaging of electronic molecular *structure*

E_{stat} = 467 V/cm

E_{align} = 7 × 10^{11} W/cm²

E_{probe} = 1.2 × 10^{14} W/cm²

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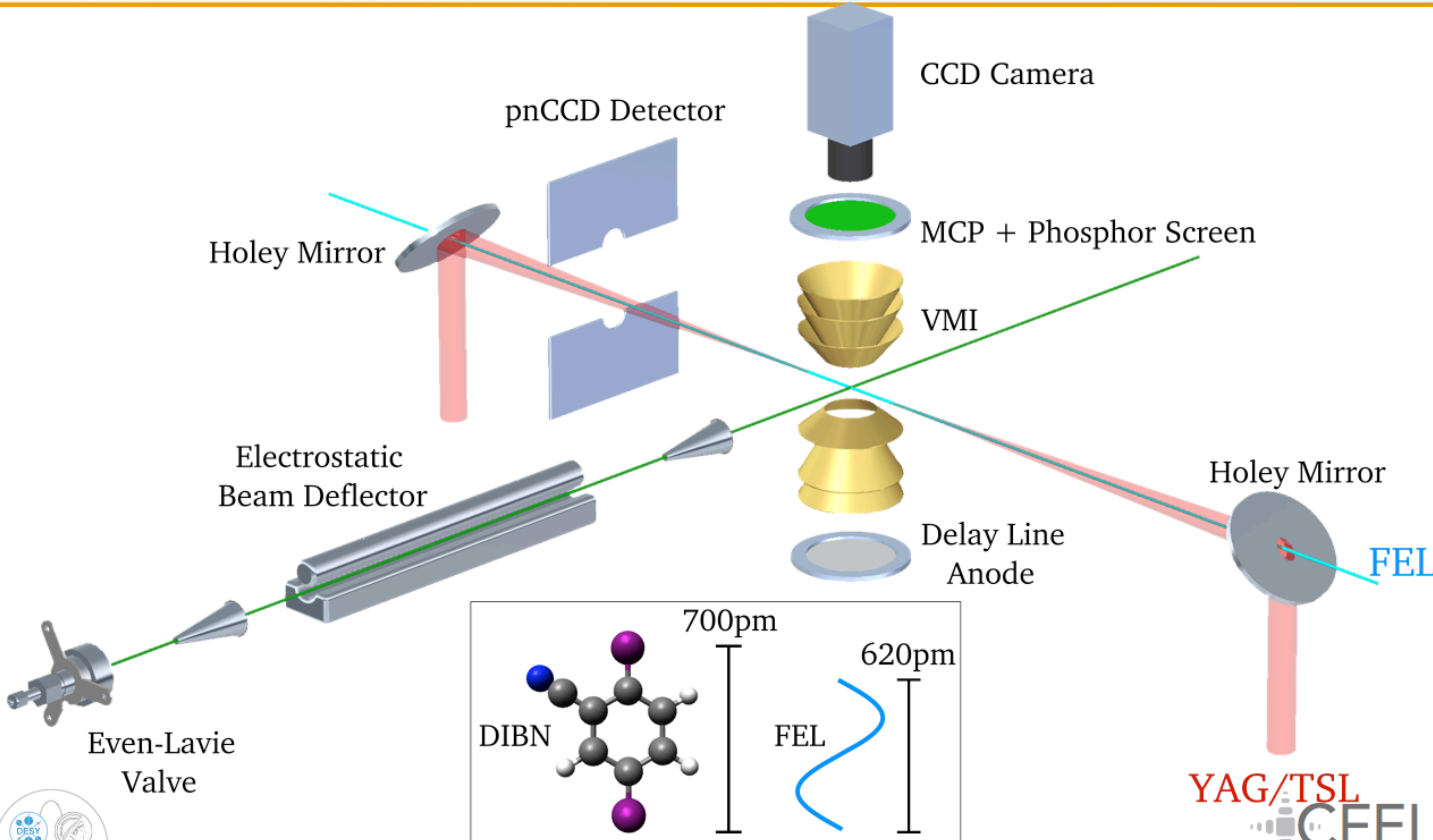
Controlled Molecule Imaging

Holmegaard, Hansen, Kalhøj, Kragh, Stapelfeldt, Filsinger, JK, Meijer, Dimitrovski, Abu-samha, Martiny, Madsen, *Nature Phys.* **6**, 428 (2010)

CFEL ASG Multi-Purpose Chamber (CAMP)
A *moving* FEL endstation



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JK, Stern, Holmegaard *et al* (52 authors), in preparation

Photoelectron diffraction of aligned molecules
F(1s) ionization of 1-ethynyl-4-fluorobenzene using the LCLS

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Controlled Molecule Imaging

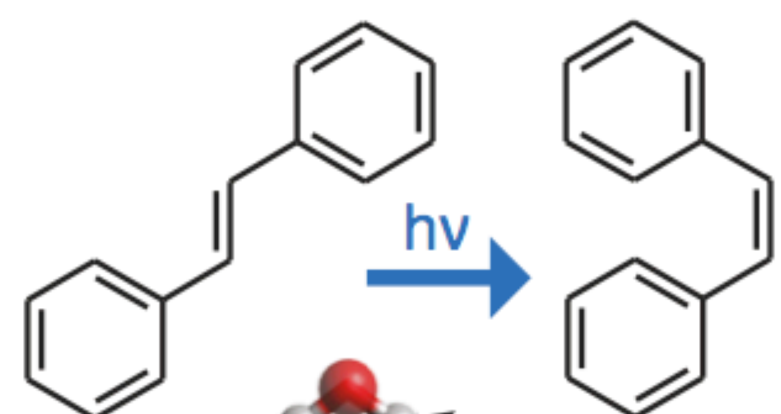
ongoing analysis: Rolles, Boll, Christensen, *et al*

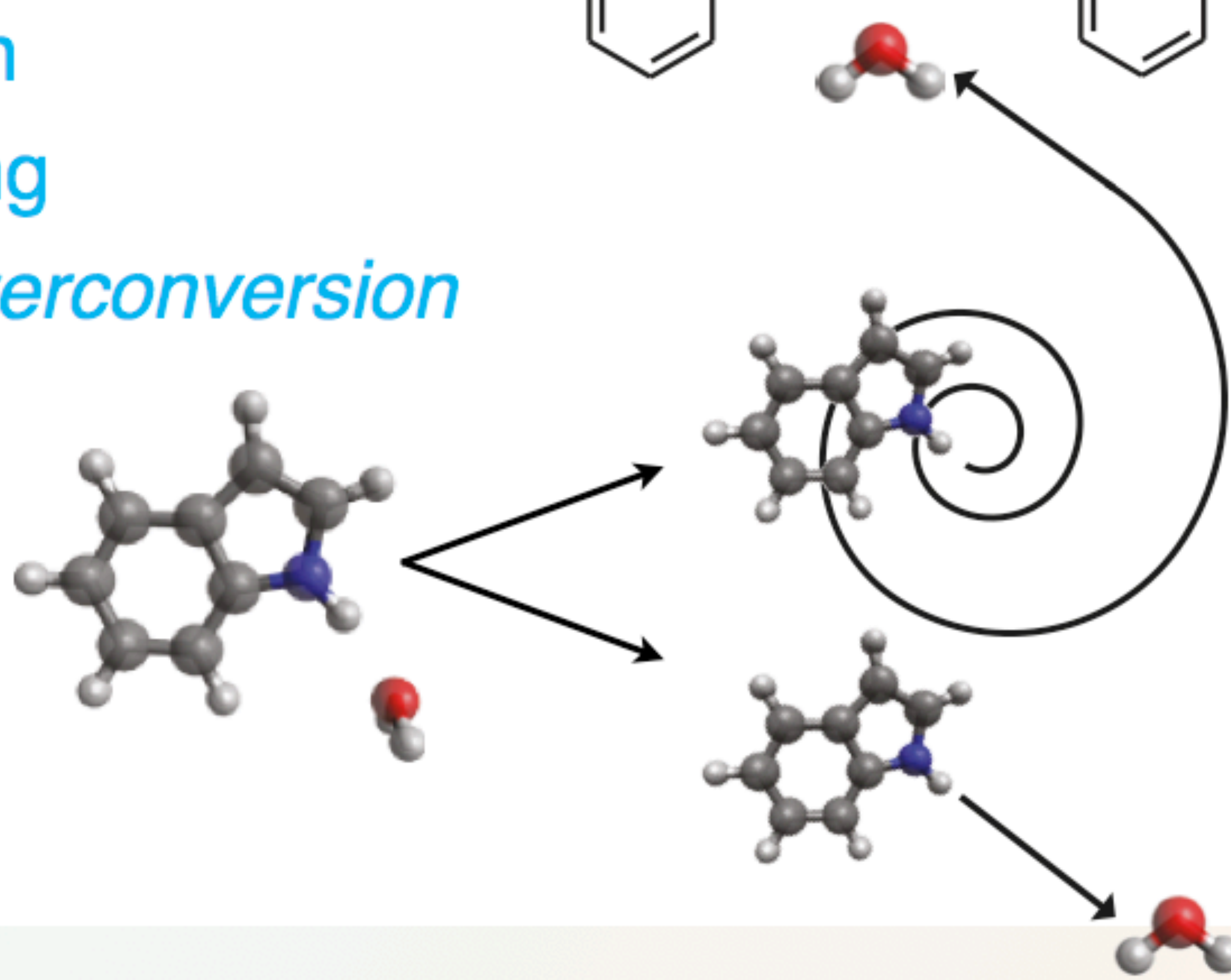
Summary

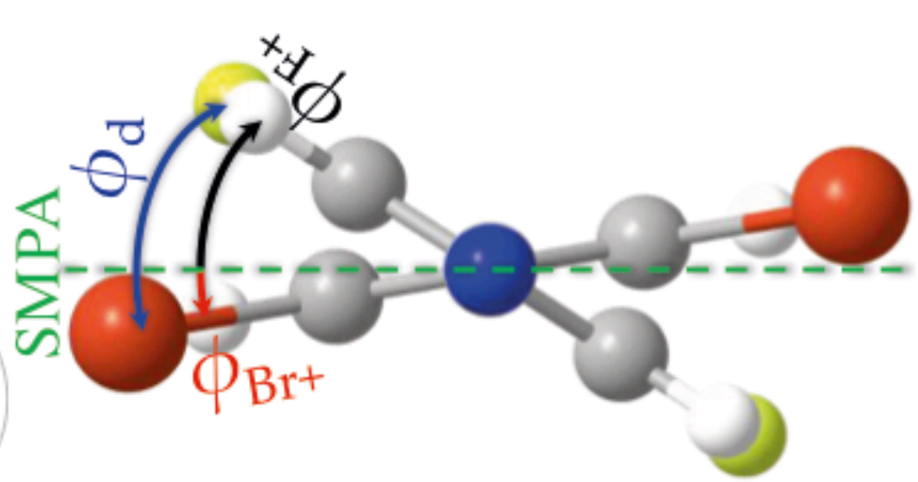
- Motion of large neutral molecules can be controlled with inhomogeneous electric fields
- Quantum-state specific interaction:
  - *Samples of molecules in a small set of (in a single) quantum states*
  - *Separation of neutral clusters according to size*
  - *Separation of structural isomers*
  - *Separation of molecules from seed gas*
  - *Extremely polar samples*
- Unprecedented degrees of alignment and mixed-field 1D and 3D orientation of large molecules
- Molecular-frame photoelectron angular distributions of 3D-oriented molecules from strong-field ionization
- first benchmark "diffraction" experiments using LCLS and FLASH
  - *strong alignment demonstrated in experiments at FELs*
  - *X-ray diffraction and molecular-frame-photoelectron angular distributions experiments on static samples of complex molecules*
  - *time-resolved molecular-frame-photoelectron angular distributions of OCS*

Outlook

- Improve achievable control
- Extend experiments to considerably larger molecules (viruses, cells)
- Exploit high repetition rates (EuXFEL, sliced synchrotrons, table-top-laser HHG) to investigate weak processes, i.e., ultrafast time-resolved investigations
  - isolated *molecular switches*
  - (ground state) *chemical dynamics*
    - conformer interconversion
    - bond-forming and breaking
  - chirality and *enantiomer interconversion*







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Short summary and outlook

- we can manipulate and control complex molecules (from OCS to µm-sized objects)
- we can exploit controlled samples in novel experiments in physics (including chemistry and biology)
  - molecular structure
  - structural dynamics (ms to as timescales)
  - chemical reactions
- Improved control experiments are necessary to state, size, and species select complex molecules at *arbitrary* repetition rates

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Lotte Holmegaard

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Lars Bojer Madsen, et al

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Australian National University
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and the whole x-ray diffraction team (*vide supra*)

as dynamics: Giuseppe Sansone, Marc Vrakking, Arnaud Rouzee, Per Johnsson, and others

Bundesministerium für Bildung und Forschung

DESY DFG cui

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We are looking for motivated colleagues!