

## MO 1: Coulomb-explosion Imaging (joint session MO/A)

Time: Monday 11:00–13:00

Location: HS 3044

## Invited Talk

MO 1.1 Mon 11:00 HS 3044

**Imaging ultrafast molecular dissociation dynamics; from conventional to surprising paths** — ●HEIDE IBRAHIM — Advanced Laser Light Source (ALLS) @ Institut National de la Recherche Scientifique (INRS-EMT), Varennes, QC, Canada

Coulomb explosion imaging (CEI) is a powerful tool to track a broad variety of molecular dynamics; even if they occur in a non-concerted manner and require single-molecule detection sensitivity. Upon photoexcitation of a molecule it will break apart. We can see fragments following direct, conventional dissociation paths, as well as fragments deviating from this minimum energy path. The latter are called roaming fragments and explore the potential energy landscape in a statistical manner. At the user facility ALLS we use CEI in combination with high repetition rate laser systems. Dissociating and roaming fragments in formaldehyde are directly captured using CEI, a hard-to-grasp statistically occurring signal. Individual pathways are distinguished based on state-of-the-art theory analysis.

MO 1.2 Mon 11:30 HS 3044

**Dynamics of H<sub>2</sub>-roaming processes, H<sub>3</sub><sup>+</sup> formation in ethanol and aminoethanol initiated by two-photon double-ionization**

— ●AARON NGAI<sup>1</sup>, SEBASTIAN HARTWEG<sup>1</sup>, JAKOB ASMUSSEN<sup>2</sup>, BJÖRN BASTIAN<sup>3</sup>, LTAIEF BEN LTAIEF<sup>2</sup>, MATTEO BONANOMI<sup>4,5</sup>, CARLO CALLEGARI<sup>6</sup>, MICHELE DI FRAIA<sup>6</sup>, KATRIN DULITZ<sup>7</sup>, RAIMUND FEIFEL<sup>8</sup>, SARANG GANESHAMANDIRAM<sup>1</sup>, SIVARAMA KRISHNAN<sup>9</sup>, AARON LAFORGE<sup>10</sup>, LANDMESSER FRIEDEMANN<sup>1</sup>, MICHELBACH MORITZ<sup>1</sup>, PAL NITISH<sup>6</sup>, PLEKAN OKSANA<sup>6</sup>, RENDLER NICNICOLAS<sup>1</sup>, RICHTER FABIAN<sup>1</sup>, SCOGNAMIGLIO AUDREY<sup>1</sup>, SIXT TOBIAS<sup>1</sup>, SQUIBB RICHARD<sup>8</sup>, SUNDARALINGAM AKGASH<sup>2</sup>, STIENKEMEIER FRANK<sup>1</sup>, and MUDRICH MARCEL<sup>2</sup> — <sup>1</sup>Physikalisches Institut, Albert-Ludwigs-Universität Freiburg, Freiburg, Germany — <sup>2</sup>Department of Physics and Astronomy, Aarhus University, Aarhus, Denmark — <sup>3</sup>Wilhelm Ostwald Institute for Physical and Theoretical Chemistry, University of Leipzig, Leipzig, Germany — <sup>4</sup>Dipartimento di Fisica Politecnico, Milano, Italy — <sup>5</sup>Istituto di Fotonica e Nanotecnologie (CNR-IFN) Milano, Italy — <sup>6</sup>Elettra - Sincrotrone Trieste S.C.p.A., Basovizza, Trieste, Italy — <sup>7</sup>Institut für Ionenphysik und Angewandte Physik, Universität Innsbruck, Innsbruck, Austria — <sup>8</sup>Department of Physics, University of Gothenburg, Göteborg, Sweden — <sup>9</sup>Department of Physics, Indian Institute of Technology Madras, Chennai, India — <sup>10</sup>Department of Physics, University of Connecticut, Storrs, Connecticut, US

The trihydrogen cation (H<sub>3</sub><sup>+</sup>) is the simplest and one of the most abundant triatomic cations in the universe. It plays a crucial role in interstellar gas-phase chemistry as it facilitates molecule-forming chemical reactions. Dynamics in simple alcohols that lead to H<sub>3</sub><sup>+</sup> formation typically involve the unusual so-called "roaming"-mechanism of a neutral H<sub>2</sub> moiety. In comparison to previous experiments using strong-field ionization by infrared (IR) pulses [1], we produce dicationic ethanol and 2-aminoethanol molecules using two-photon double-ionization with extreme ultraviolet (XUV) light, and probe the dynamics of H<sub>3</sub><sup>+</sup> formation with a visible (VIS) pulse in a time-resolved pump-probe scheme. We compare results between measurements with XUV photons either below or above the double-ionization threshold, including the lifetimes of intermediate states.

[1] Ekanayake, N. *et al. Nat. Commun.* **9**, 5186 (2018)

MO 1.3 Mon 11:45 HS 3044

**New endstation for controlled molecule experiments and ultrafast dynamics of OCS** — ●WUWEI JIN<sup>1,2</sup>, IVO VINKLÁREK<sup>1</sup>, SEBASTIAN TRIPPEL<sup>1,3</sup>, HUBERTUS BROMBERGER<sup>1</sup>, SERGEY RYABCHUK<sup>1</sup>, ERIK MÄNSSON<sup>1</sup>, ANDREA TRABATTONI<sup>1</sup>, VINCENT WANIE<sup>1</sup>, FRANCESCA CALEGARI<sup>1</sup>, and JOCHEN KÜPPER<sup>1,2,3</sup> — <sup>1</sup>Center for Free-Electron Laser Science, Deutsches Elektronen-Synchrotron DESY, Hamburg — <sup>2</sup>Department of Physics, Universität Hamburg — <sup>3</sup>Center for Ultrafast Imaging, Universität Hamburg

Imaging ultrafast photochemical reactions with atomic-spatial and femtosecond-temporal resolution is one of the ultimate goals of physical chemistry and the molecular sciences [1]. We present details on our newly established transportable endstation for controlled molecules (eCOMO) and discuss our ultrafast (sub 10 fs) time-resolved study of the photodissociation dynamics of carbonyl sulfide (OCS) after UV-

photoexcitation at  $\lambda = 267$  nm. OCS was purified and separated from the helium seed gas using the electrostatic deflector [2]. The UV-induced dynamics was probed through strong field ionization using a velocity map imaging spectrometer in combination with a Timepix3 camera [3].

[1] J Onvlee, S Trippel, and J Küpper, *Nat. Commun.* **7462**, 13 (2022)

[2] YP Chang, D Horke, S Trippel, and J Küpper, *Int. Rev. Phys. Chem.* **557**, 34 (2015)

[3] H Bromberger, *et int.* (9 authors), S Trippel, B Erk, and J Küpper, *J. Phys. B.* **144001**, 55 (2022)

MO 1.4 Mon 12:00 HS 3044

**Complete imaging of the reaction pathways of ionized water dimer** — ●LUIA BLUM<sup>1,2</sup>, IVO S. VINKLÁREK<sup>1</sup>, HUBERTUS BROMBERGER<sup>1</sup>, SEBASTIAN TRIPPEL<sup>1</sup>, and JOCHEN KÜPPER<sup>1,2,3</sup>

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We applied a pure ensemble (92 %) of water dimer (H<sub>2</sub>O)<sub>2</sub>, spatially separated by electrostatic deflection, and subsequently ionized by strong-field ionization, to investigate the ion-radical chemistry of water clusters [1]. The direct observation of fragmentation channels of (H<sub>2</sub>O)<sub>2</sub><sup>+</sup> and (H<sub>2</sub>O)<sub>2</sub><sup>+2</sup> by multi-mass imaging reveals several yet unknown ion-radical pathways and provides a comprehensive picture of (H<sub>2</sub>O)<sub>2</sub><sup>+/+2</sup>, including experimental branching ratios. Furthermore, the ion yields for the Coulomb explosion channels of (H<sub>2</sub>O)<sub>2</sub><sup>+2</sup> indicate electron-recoil-impact ionization as the dominant process, opening the discussion about avenues to control electron recoil and multiple-ionization processes in supramolecular complexes. The study of the (H<sub>2</sub>O)<sub>2</sub><sup>+/+2</sup> ionization fragmentation process is highly relevant to ion-radical heterogeneous chemistry occurring on ice mantles in the Earth's atmosphere and in interstellar space [2].

[1] Vinklársek, I. S., Bromberger, H., Vadassery N., Jin W., Küpper, J., Trippel, S., *submitted*; arXiv:2308.08006 [physics].

[2] Vogt, E., Kjaergaard, H. G., *Annu. Rev. Phys. Chem.*, **73**, 209-231 (2022).

MO 1.5 Mon 12:15 HS 3044

**Understanding fragmentation dynamics of difluorodiiodomethane** — ●NIDIN VADASSERY<sup>1,3</sup>, IVO VINKLÁREK<sup>1</sup>, SEBASTIAN TRIPPEL<sup>1,2</sup>, and JOCHEN KÜPPER<sup>1,2,3</sup>

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Unimolecular photo-fragmentation is prevalent in the many chemical reactions that affect the environment, like ozone depletion, synthesis of oxidative hydrocarbons, formation of aerosol particles, *etc.* [1]. The photo-dissociation of man-made and naturally occurring polyhalohydrocarbons is among the major causes which contribute to such climate-impacting reactions. Difluorodiiodomethane (CF<sub>2</sub>I<sub>2</sub>) one such example of polyhalohydrocarbon shows unconventional dynamics near dissociative energies [2]. Here, we present our experimental result of exploring the dissociation dynamics of CF<sub>2</sub>I<sub>2</sub> using near-infrared laser pulses. A pure sample of CF<sub>2</sub>I<sub>2</sub> was produced using the deflector in the eCOMO endstation [3]. We show capability of the end-station to reveal metastable states and unravel the complex quantum-state-specific dynamics during photo-fragmentation.

[1] J. C. G. Martin, *et al., J. Am. Chem. Soc.* **144**, 9240 (2022).

[2] P. Z. El-Khoury, *et al., J. Chem. Phys.* **132**, 124501 (2010).

[3] I. S. Vinklársek, *et int.* (3 authors), J. Küpper, S. Trippel, arXiv:2308.08006 [physics] (2023).

MO 1.6 Mon 12:30 HS 3044

**Ultrafast photofragmentation studies of CF<sub>3</sub>I-I<sup>-</sup> using mass-selected ion-molecule cluster beam apparatus** — ●XIAOJUN WANG<sup>1,4</sup>, MAHMUDUL HASAN<sup>1</sup>, LIN FAN<sup>1</sup>, YIBO WANG<sup>1</sup>, HUI LI<sup>2</sup>, DANIEL SLAUGHTER<sup>3</sup>, and MARTIN CENTURION<sup>1</sup>

— <sup>1</sup>Department of Physics and Astronomy, University of Nebraska-Lincoln, Lincoln, Nebraska 68588, USA — <sup>2</sup>Department of Chemistry, Nebraska Center for Materials and Nanoscience, and Center for Integrated Biomolecular Communication, University of Nebraska-Lincoln, Lincoln, Nebraska

68588, USA — <sup>3</sup>Chemical Sciences Division, Lawrence Berkeley National Laboratory, 1 Cyclotron Rd., Berkeley, California 94720, USA — <sup>4</sup>Deutsches Elektronen-Synchrotron DESY, Notkestr. 85, 22607 Hamburg, Germany.

We describe an apparatus for investigating the excited-state dissociation dynamics of mass-selected ion-molecule clusters by mass-resolving and detecting photofragment-ions and neutrals, in coincidence, using an ultrafast laser operating at high repetition rates. The apparatus performance is tested by measuring the photofragments:  $I^-$ ,  $CF_3I^-$  and neutrals from photoexcitation of the ion-molecule cluster  $CF_3I-I^-$  using femtosecond UV laser pulses with a wavelength of 266 nm. The experimental results are compared with our ground state and excited state electronic structure calculations as well as the existing results and calculations, with particular attention to the generation mechanism of the anion fragments and dissociation channels of the ion-molecule cluster  $CF_3I-I^-$  in the charge-transfer excited state.

Reference: Rev. Sci. Instrum. 94, 095111 (2023)

MO 1.7 Mon 12:45 HS 3044

**Coulomb explosion imaging of ultrafast photochemistry in molecular photoswitches** — KIERAN CHEUNG<sup>1</sup>, CLAUS PETER

SCHULZ<sup>2</sup>, ARNAUD ROUZÉÉ<sup>2</sup>, TILL JAHNKE<sup>3</sup>, DANIEL ROLLES<sup>4</sup>, GIUSEPPE SANSONE<sup>5</sup>, MICHAEL MEYER<sup>3</sup>, MARK BROUARD<sup>1</sup>, TERRY MULLINS<sup>1</sup>, and •KASRA AMINI<sup>2</sup> — <sup>1</sup>Chemistry Research Laboratory, Department of Chemistry, University of Oxford, Oxford OX1 3TA, UK — <sup>2</sup>Max-Born-Institut, Max-Born-Str. 2A, 12489 Berlin, Germany — <sup>3</sup>European XFEL, Schenefeld, Germany — <sup>4</sup>J.R. Macdonald Laboratory, Department of Physics, Kansas State University, Manhattan, KS, USA — <sup>5</sup>Physikalisches Institut, Universität Freiburg, D-79106 Freiburg, Germany

Here, we present an X-ray Coulomb explosion imaging (CEI) study into the photofragmentation and photochemistry of trans-4,4-difluoroazobenzene (DFAB) measured with the COLTRIMS Reaction Microscope at the SQS station of European XFEL. We first provide a systematic study of X-ray fragmentation in DFAB with covariance analysis. We then present pump-probe X-ray CEI measurements of DFAB excited to its first excited state under different visible pump excitation conditions. We discuss the limited ability of trans-DFAB to undergo trans-to-cis isomerization after initial population of its  $S_1$  state, and reveal the onset of a dissociative ionization photodissociation process.