

A 4: Coulomb-explosion Imaging (joint session MO/A)

Time: Monday 11:00–13:00

Location: HS 3044

Invited Talk

A 4.1 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.

A 4.2 Mon 11:30 HS 3044

Dynamics of H₂-roaming processes, H₃⁺ formation in ethanol and aminoethanol initiated by two-photon double-ionization

— ●AARON NGAI¹, SEBASTIAN HARTWEG¹, JAKOB ASMUSSEN², BJÖRN BASTIAN³, LTAIEF BEN LTAIEF², MATTEO BONANOMI^{4,5}, CARLO CALLEGARI⁶, MICHELE DI FRAIA⁶, KATRIN DULITZ⁷, RAIMUND FEIFEL⁸, SARANG GANESHAMANDIRAM¹, SIVARAMA KRISHNAN⁹, AARON LAForge¹⁰, LANDMESSER FRIEDEMANN¹, MICHELBACH MORITZ¹, PAL NITISH⁶, PLEKAN OKSANA⁶, RENDLER NICNICOLAS¹, RICHTER FABIAN¹, SCOGNAMIGLIO AUDREY¹, SIXT TOBIAS¹, SQUIBB RICHARD⁸, SUNDARALINGAM AKGASH², STIENKEMEIER FRANK¹, and MUDRICH MARCEL² — ¹Physikalisches Institut, Albert-Ludwigs-Universität Freiburg, Freiburg, Germany — ²Department of Physics and Astronomy, Aarhus University, Aarhus, Denmark — ³Wilhelm Ostwald Institute for Physical and Theoretical Chemistry, University of Leipzig, Leipzig, Germany — ⁴Dipartimento di Fisica Politecnico, Milano, Italy — ⁵Istituto di Fotonica e Nanotecnologie (CNR-IFN) Milano, Italy — ⁶Elettra - Sincrotrone Trieste S.C.p.A., Basovizza, Trieste, Italy — ⁷Institut für Ionenphysik und Angewandte Physik, Universität Innsbruck, Innsbruck, Austria — ⁸Department of Physics, University of Gothenburg, Göteborg, Sweden — ⁹Department of Physics, Indian Institute of Technology Madras, Chennai, India — ¹⁰Department of Physics, University of Connecticut, Storrs, Connecticut, US

The trihydrogen cation (H₃⁺) 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 H₃⁺ formation typically involve the unusual so-called "roaming"-mechanism of a neutral H₂ 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 H₃⁺ 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)

A 4.3 Mon 11:45 HS 3044

New endstation for controlled molecule experiments and ultrafast dynamics of OCS — ●WUWEI JIN^{1,2}, IVO VINKLÁREK¹, SEBASTIAN TRIPPEL^{1,3}, HUBERTUS BROMBERGER¹, SERGEY RYABCHUK¹, ERIK MÄNSSON¹, ANDREA TRABATTONI¹, VINCENT WANIE¹, FRANCESCA CALEGARI¹, and JOCHEN KÜPPER^{1,2,3} — ¹Center for Free-Electron Laser Science, Deutsches Elektronen-Synchrotron DESY, Hamburg — ²Department of Physics, Universität Hamburg — ³Center 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)

A 4.4 Mon 12:00 HS 3044

Complete imaging of the reaction pathways of ionized water dimer — ●LUIA BLUM^{1,2}, IVO S. VINKLÁREK¹, HUBERTUS BROMBERGER¹, SEBASTIAN TRIPPEL¹, and JOCHEN KÜPPER^{1,2,3}

— ¹Center for Free-Electron Laser Science CFEL, Deutsches Elektronen-Synchrotron DESY, Hamburg — ²Department of Physics, Universität Hamburg — ³Center for Ultrafast Imaging, Universität Hamburg

We applied a pure ensemble (92 %) of water dimer (H₂O)₂, 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 (H₂O)₂⁺ and (H₂O)₂⁺² by multi-mass imaging reveals several yet unknown ion-radical pathways and provides a comprehensive picture of (H₂O)₂^{+/+2}, including experimental branching ratios. Furthermore, the ion yields for the Coulomb explosion channels of (H₂O)₂⁺² 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 (H₂O)₂^{+/+2} 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).

A 4.5 Mon 12:15 HS 3044

Understanding fragmentation dynamics of difluorodiodomethane — ●NIDIN VADASSERY^{1,3}, IVO VINKLÁREK¹, SEBASTIAN TRIPPEL^{1,2}, and JOCHEN KÜPPER^{1,2,3}

— ¹Center for Free-Electron Laser Science, Deutsches Elektronen-Synchrotron DESY, Hamburg, Germany — ²Department of Physics, Universität Hamburg — ³Department of Chemistry, Universität Hamburg

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. Difluorodiodomethane (CF₂I₂) one such example of polyhalohydrocarbon shows unconventional dynamics near dissociative energies [2]. Here, we present our experimental result of exploring the dissociation dynamics of CF₂I₂ using near-infrared laser pulses. A pure sample of CF₂I₂ 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).

A 4.6 Mon 12:30 HS 3044

Ultrafast photofragmentation studies of CF₃I-I⁻ using mass-selected ion-molecule cluster beam apparatus — ●XIAOJUN WANG^{1,4}, MAHMUDUL HASAN¹, LIN FAN¹, YIBO WANG¹, HUI LI², DANIEL SLAUGHTER³, and MARTIN CENTURION¹

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

68588, USA — ³Chemical Sciences Division, Lawrence Berkeley National Laboratory, 1 Cyclotron Rd., Berkeley, California 94720, USA — ⁴Deutsches 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)

A 4.7 Mon 12:45 HS 3044

Coulomb explosion imaging of ultrafast photochemistry in molecular photoswitches — KIERAN CHEUNG¹, CLAUD PETER

SCHULZ², ARNAUD ROUZÉÉ², TILL JAHNKE³, DANIEL ROLLES⁴, GIUSEPPE SANSONE⁵, MICHAEL MEYER³, MARK BROUARD¹, TERRY MULLINS¹, and •KASRA AMINI² — ¹Chemistry Research Laboratory, Department of Chemistry, University of Oxford, Oxford OX1 3TA, UK — ²Max-Born-Institut, Max-Born-Str. 2A, 12489 Berlin, Germany — ³European XFEL, Schenefeld, Germany — ⁴J.R. Macdonald Laboratory, Department of Physics, Kansas State University, Manhattan, KS, USA — ⁵Physikalisches 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.