

MO 8: Ultrafast Spectroscopy with XUV (joint session MO/A)

Time: Monday 16:15–17:45

Location: PA 2.150

Invited Talk

MO 8.1 Mon 16:15 PA 2.150

Measurement of femtosecond dynamics in HCL molecules with THz streaking — KATHARINA WENIG¹, MAREK WIELAND¹, SOPHIE WALTHER¹, ARNE BAUMANN¹, ANASTASIOS DIMITRIOU¹, MARK PRANDOLINI¹, OLIVER SCHEPP¹, IVETTE BERMUNDEZ MACHIAS², MALTE SUMFLETH¹, NIKOLA STOJANOVIC², STEFAN DÜSTERER², JULIANE RÖNTSCH-SCHULENBURG², MARKUS DRESCHER¹, and •ULRIKE FRÜHLING¹ — ¹Institut für Experimentalphysik, Universität Hamburg, Deutschland — ²Deutsches Elektronen-Synchrotron DESY, Hamburg, Deutschland

We have studied the ultrafast electronic decay of core excited HCL molecules directly in the time domain. In the experiment we used femtosecond XUV pulses from the free-electron laser in Hamburg (FLASH) to resonantly excite $2p_{3/2}$ core electrons to the antibonding σ^* orbital. After the excitation the molecules dissociate and the electronic excitation relaxes via the emission of an Auger-electron. In order to follow the evolution of this coupled electronic and nuclear dynamics we have collinearly superimposed the exciting XUV pulses with intense THz pulses from the FLASH THz undulator. Electrons emitted from the molecules are accelerated (streaked) by the THz electric field whereby the resulting momentum change depends on the THz phase at the ionization time. Thus the ionization dynamics can be studied by measuring the streaked electron spectra. In the talk the experimental technique and first experimental results will be presented.

MO 8.2 Mon 16:45 PA 2.150

A chemical understanding of the lack of site-specificity in molecular inner-shell photofragmentation — •LUDGER INHETER^{1,2}, BART OOSTENRIJK³, SANG-KIL SON^{1,2}, ROBIN SANTRA^{1,2,4}, LINDA YOUNG⁵, and STACEY L. SORENSEN³ — ¹Center for Free-Electron Laser Science, DESY — ²The Hamburg Centre for Ultrafast Imaging — ³Department of Physics, Lund University — ⁴Department of Physics, University of Hamburg — ⁵Argonne National Laboratory

In many cases fragmentation of molecules upon inner-shell ionization is very unspecific with respect to the initially localized ionization site. Often this finding is interpreted in terms of an equilibration of internal energy into vibrational degrees of freedom after Auger decay. Here we investigate the x-ray photofragmentation of ethyl trifluoroacetate upon core electron ionization at environmentally distinct carbon sites using photoelectron-photoion-photoion coincidence measurements and ab-initio electronic structure calculations. For all the 4 carbon ionization sites, the Auger decay weakens the same bonds and transfers the two charges to opposite ends of the molecule, which leads to a rapid dissociation into 3 fragments followed by further fragmentation steps. The lack of site-specificity is attributed to the character of the dicationic electronic states after Auger decay, instead of a fast equilibration of internal energy.

MO 8.3 Mon 17:00 PA 2.150

Electron-ion coincidence spectroscopy on small quantum systems — •LENA WORBS¹, ANDREAS PRZYSTAWIK¹, DAVID SCHWICKERT¹, SERGEY USENKO^{1,2}, and TIM LAARMANN^{1,2} — ¹Deutsches Elektronen-Synchrotron DESY, Notkestrasse 85, Hamburg 22607, Germany — ²The Hamburg Centre for Ultrafast Imaging CUI, Luruper Chaussee 149, Hamburg 22761, Germany

Relative phase control with sub-cycle precision of extreme-ultraviolet (XUV) pulses from a free-electron laser has been demonstrated by using a Michelson-type all reflective interferometric autocorrelator to trace the light-wave oscillation with a period of 129 as [1]. Novel applications using these phase controlled pulses in an electron-ion co-

incidence experiment will be discussed in the present contribution.

For this purpose a double-sided electron-ion coincidence spectrometer with delay-line detectors has been developed to detect electrons and ions generated in the same ionization event. The spectrometer detects ions and electrons with position-sensitivity providing angular-resolved information. The use of delay-line detectors enhances the information content of the electron spectra due to additional time-of-flight information on the ion side and thus the selection of wanted coincidence events, i.e. the selection of particularly charged ions as counterparts. First results on Xe atoms and C_{60} molecules will be discussed.

[1] Usenko, S. et al. Attosecond interferometry with self-amplified spontaneous emission of a free electron laser. Nat. Commun. 8, 15626 doi: 10.1038/ncomms15626 (2017)

MO 8.4 Mon 17:15 PA 2.150

Molecular Light-Induced Couplings Revealed by Attosecond Transient Absorption Spectroscopy — •LORENZ DRESCHER, GEERT REITSMA, TOBIAS WITTING, SERGUEI PATCHKOVSKII, MARC VRACKING, and JOCHEN MIKOSCH — Max-Born-Institut, Berlin, Deutschland

Attosecond transient absorption spectroscopy (ATAS) is the study of the modulation of absorption of short attosecond XUV pulses in matter by a phase-locked strong IR pulse. In recent years, ATAS has been used to study the light-induced coupling of nearby states in atoms [1] and homonuclear molecules.

Here we present results from our experimental and theoretical study of ATAS of a polyatomic molecule (CH_3I) in the core-to-valence and core-to-Rydberg transition region. Core-excited states are an interesting target for transient absorption because element specific transitions give a local view into the molecular valence [2]. For ATAS they are additionally intriguing because the short coherence time (due to fast Auger decay of the core-hole) enhances the sub-cycle dependence on the probing IR field.

Our results show the sub-cycle dependent light-induced coupling of states due to the AC Stark effect. It shows that the coupling of the core-to-Rydberg states is much stronger than the coupling of the core-to-valence states.

[1] C. Ott, et al., Nature 516, 374-378 (2014).

[2] L. Drescher et al., JCP 145, 011101 (2016).

MO 8.5 Mon 17:30 PA 2.150

Electron spectra of a water molecule irradiated by an x-ray free-electron laser pulse — •JULIA M. SCHÄFER^{1,2}, LUDGER INHETER^{1,3}, SANG-KIL SON¹, REINHOLD F. FINK², and ROBIN SANTRA^{1,3,4} — ¹Center for Free-Electron Laser Science, DESY, Hamburg — ²Institut für Physikalische und Theoretische Chemie, Universität Tübingen — ³The Hamburg Center for Ultrafast Imaging, Hamburg — ⁴Fachbereich Physik, Universität Hamburg

With the high intensity X-ray light that is generated by X-ray free-electron lasers (XFELs) molecular samples can be ionized many times. We investigate the spectrum of electrons emitted from molecules exposed to these intense X-ray pulses. Calculated photoelectron and Auger electron spectra are presented for a single water molecule that reaches through repeated ionization steps many electronic hole configurations. The rich details in the spectra depend in a non-intuitive way on X-ray pulse parameters. We discuss how the observed trends can be explained by the competition of microscopic electronic processes. A detailed comparison between spectra calculated for independent atoms and spectra calculated for molecules is made. Our results demonstrate how multiple X-ray ionization related effects like Charge-Rearrangement-Enhanced X-ray Ionization in Molecules (CREXIM) and frustrated absorption manifest themselves in the electron spectra.