

MO 16: Molecules in Intense Laser Fields (joint session MO/A)

Time: Wednesday 14:00–15:45

Location: PA 2.150

Invited Talk

MO 16.1 Wed 14:00 PA 2.150

Strong-field ionization of laser-aligned molecules — ●JOCHEN KÜPPER — Center for Free-Electron Laser Science, DESY, Hamburg — Center for Ultrafast Imaging, Universität Hamburg — Department of Physics and Department of Chemistry, Universität Hamburg

The interaction of strong laser fields with matter intrinsically enables the imaging of transient dynamics with extremely high spatiotemporal resolution. This paradigm of photophysics has grown into new emerging research areas, ranging from attosecond science to laser-induced electron diffraction, providing new insight into atoms, molecules and, more recently, condensed matter. Also, the earliest moments of strong-field interactions have attracted attention for capturing the intrinsic nature of strong-field physics. While pioneering attosecond science experiments and molecular-frame measurements revealed non-trivial spatiotemporal features in electron tunneling, these initial conditions are generally considered a weak perturbation. We investigated strong-field ionization in the molecular frame. Carbonyl sulfide (OCS) molecules were quantum-state selected, strongly laser aligned, and ionized using short near-infrared laser pulses. We analyzed the dynamics of the electron and discuss the obtained molecular-frame photoelectron-angular distributions. Our findings have strong impact in the interpretation of laser induced electron diffraction, where the photoelectron momentum distribution is used to retrieve molecular structures. Furthermore, the encoding of the time-energy relation in the photoelectron momenta provides new ways of probing electron tunneling and the molecular potential with sub-femtosecond resolution.

MO 16.2 Wed 14:30 PA 2.150

Intermolecular vibration in (NO₂)₂ molecules studied with few-cycle laser pulses — KATRIN REININGER, JINGMING LONG, FEDERICO FURCH, MARC J.J. VRAKING, ●CLAUS P. SCHULZ, and JOCHEN MIKOSCH — Max-Born-Institut, Max-Born-Str. 2a, 12489 Berlin, Germany

The intermolecular vibration of the NO₂ dimer molecule is an interesting object of study for high-harmonic generation and strong-field ionization probes of molecular dynamics [1]. The vibration can be conveniently excited by impulsive stimulated Raman scattering (ISRS).

Here we measure the amplitude of the intramolecular motion using photofragment kinetic energy spectroscopy. We employ a newly developed high repetition rate Optical Parametric Chirped-Pulse Amplification (OPCPA) laser system delivering sub-7 fs duration pulses [2] in combination with a novel velocity map imaging (VMI) spectrometer based on an in-vacuum pixel detector [3].

Following ISRS, the molecule is strong-field ionized to the (NO₂)₂⁺ A_g state and undergoes dissociation. We observe an oscillation in the kinetic energy release of NO₂⁺ with pump-probe delay on the 130fs time-scale of the O₂N–NO₂ vibration. From the kinetic energy spectrum and the calculated potential surfaces, we determine the amplitude of the vibrational motion in the (NO₂)₂ ground state.

[1] W. Li, *et al.*, *Science* 322, 1207 (2008).

[2] F. J. Furch, *et al.*, *Opt. Express* 24, 19293 (2016).

[3] J. M. Long, *et al.*, *J. Chem. Phys.* 147, 10, 013919 (2017).

MO 16.3 Wed 14:45 PA 2.150

Intensity dependence of Photon Energy Sharing in H₂ Multi-photon Ionization — ●PATRICK FROSS, DENHI MARTINEZ, NICOLAS CAMUS, THOMAS PFEIFER, and ROBERT MOSHAMMER — Max-Planck-Institut für Kernphysik, Heidelberg, Germany

When ionizing molecules with strong laser fields, the dynamics of photofragmentation determines how the photon energy is shared between the resulting fragments. Understanding this photon energy sharing helps to understand and eventually control the fragmentation process in intense laser fields. Recently Joint-Energy-Spectra (JES) have been observed for dissociative and double ionization of H₂ in order to investigate photon-number-resolved ionization dynamics at laser intensities of $5 \cdot 10^{13}$ – $1 \cdot 10^{14}$ W/cm². We present measurements of full 3D photon-number-resolved momentum distributions in H₂ dissociative and double ionization for different intensities using 35fs laser pulses with a central wavelength of 395nm and recording the reaction fragments with a Reaction Microscope. The JES clearly shows that

for high laser intensities events involving different numbers of photons cannot be distinguished anymore.

MO 16.4 Wed 15:00 PA 2.150

Fragmentation dynamics of HeH⁺ in intense ultrashort laser pulses — ●PHILIPP WUSTELT^{1,2}, FLORIAN OPPERMAN³, LUN YUE⁴, MAX MÖLLER^{1,2}, A. MAX SAYLER^{1,2}, MANFRED LEIN³, STEFANIE GRÄFE⁴, and GERHARD G. PAULUS^{1,2} — ¹Institute of Optics and Quantum Electronics, Friedrich-Schiller-University Jena, D-07743 Jena, Germany — ²Helmholtz Institute Jena, D-07743 Jena, Germany — ³Institut für Theoretische Physik, Leibniz Universität Hannover, Appelstraße 2, 30167 Hannover — ⁴Institute of Physical Chemistry, Friedrich-Schiller University-Jena, Helmholtzweg 4, D-07743 Jena, Germany

The helium hydride molecular ion, HeH⁺, is the simplest heteronuclear polar molecule and serves as a benchmark system for the investigation of multi-electron molecules and molecules with a permanent dipole. We specifically address the question: How does the permanent dipole of HeH⁺ affect the fragmentation dynamics in intense ultrashort laser pulses? We study the laser induced laser-induced fragmentation; including non-ionizing dissociation, single ionization and double ionization; of an ion beam of helium hydride and an isotopologue at various wavelengths and intensities. These results are interpreted using reduced dimensionality solutions to the time-dependent Schrödinger equation and with simulations based on Dressed surface hopping.

MO 16.5 Wed 15:15 PA 2.150

Electron-Nuclear Coupling through Autoionizing Rydberg States after Strong-field Excitation of Molecules — ●YONGHAO MI, NICOLAS CAMU, LUTZ FECHNER, MARTIN LAUX, ROBERT MOSHAMMER, and THOMAS PFEIFER — Max-Planck-Institute for Nuclear Physics

Channel-selective electron emission from strong-field photo-ionization of H₂ molecules is experimentally investigated by using ultrashort laser pulses and a Reaction Microscope. The electron momenta and energy spectra in coincidence with bound and dissociative ionization channels are compared. Surprisingly, we observed an enhancement of the photoelectron yield in the low-energy region for the bound (H₂⁺) ionization channel. By further investigation of asymmetrical electron emission using two-color laser pulses, this enhancement is understood as the population of the autoionizing states of neutral H₂ molecules in which vibrational energy is transferred to electronic energy. This general mechanism, provides access to the excited-state population of molecular ions produced in a strong-field interaction.

MO 16.6 Wed 15:30 PA 2.150

Probing electronic structure via molecular-frame photoelectron imaging — ●JOSS WIESE¹, SEBASTIAN TRIPPEL¹, and JOCHEN KÜPPER^{1,2,3} — ¹Center for Free-Electron Laser Science, DESY, Hamburg — ²The Hamburg Center for Ultrafast Imaging, Hamburg — ³Department of Physics, University of Hamburg

Chemical function arises from the interplay of valence electrons and a view at the evolution of the highest occupied molecular orbitals (HOMOs) during a chemical reaction promises direct insight into the fundamentals of chemistry. For this purpose we employ tomographic photoelectron imaging [1] of spatially confined ensembles of indole strong-field ionized by intense near-infrared laser pulses. Reconstructed static three-dimensional photoelectron distributions in molecular-frame momentum space (MF-ARPES) will be presented. These exhibit distinct manifestations of tunneling as well as multiphoton ionization, which allow for the mapping of the molecule's electronic structure. The technique yields experimental access to properties of the HOMO potential energy surface that are commonly only approachable through quantum chemistry. Based on the measured photoelectron distributions the importance of resonant electronic states for the tunneling ionization pathway will be enquired and the validity of molecular strong-field ionization models will be tested.

[1] Maurer, Dimitrovski, Christensen, Madsen, Stapelfeldt, *Phys. Rev. Lett.* 109, 123001 (2012)