

MO 9: Molecules in Intense Laser Fields

Time: Monday 16:15–17:45

Location: S HS 002 Biologie

MO 9.1 Mon 16:15 S HS 002 Biologie

Imaging experiments in the molecular frame — ●EVANGELOS T. KARAMATSKOS^{1,2,3}, SEBASTIAN RAABE⁴, TERRY MULLINS¹, ANDREA TRABATTONI¹, PHILIPP STAMMER⁴, GILDAS GOLDSZTEJN⁴, RASMUS R. JOHANSEN⁵, KAROL DLUGOLECKI¹, HENRIK STAPELFELDT⁵, MARC J.J. VRAKKING⁴, SEBASTIAN TRIPPEL^{1,3}, ARNAUD ROUZÉE⁴, and JOCHEN KÜPPER^{1,2,3} — ¹CFEL, DESY, Hamburg, Germany — ²Departments of Physics and Chemistry, Universität Hamburg, Germany — ³CUI, Universität Hamburg, Germany — ⁴Max-Born Institute, Berlin, Germany — ⁵Department of Chemistry, Aarhus University, Denmark

Imaging the ultrafast dynamics of molecules requires experimental methods, that offer atomic spatial and (sub-)femtosecond temporal resolution. The possibility to prepare cold, controlled molecular samples in the gas phase, combined with elaborate methods to fix the molecules in space, are important prerequisites to image molecular dynamics directly in the molecule-fixed frame. We present results on strong field-free alignment, achieved for the linear carbonyl sulfide (OCS) molecule and the complex, asymmetric top rotor indole. Furthermore, employing the laser-induced electron diffraction (LIED) method, the molecular structure of OCS could be retrieved with atomic resolution. Molecular-frame angularly-resolved photoelectron spectra show clear signatures of strong field photoelectron holography, which differ, depending on the orientation of the molecules with respect to the laser polarization of the ionizing field. A discussion of the observed interference patterns will be presented. References: Karamatskos et al., arXiv:1807.01034

MO 9.2 Mon 16:30 S HS 002 Biologie

A comparative study on ionization-induced dissociation of Methane, irradiated by 800 nm and 400 nm laser fields — LAZAROS VARVAREZOS¹, ALEXANDER ACHNER², ●RENÉ WAGNER², JOHN COSTELLO¹, MICHAEL MEYER², and PATRIK GRYSHTOL² — ¹School of Physical Sciences and NCPST, Dublin City University, Dublin 9, Ireland — ²Small Quantum Systems Group, European XFEL GmbH, 22869 Schenefeld, Germany

Irradiation of methane by intense laser fields leads to the production of several molecular ions CH_n^{m+} ($m = 1, 2$ with $n = 0 - 4$) and atomic fragments, such as H_n^{m+} ($m = 0, 1$ with $n = 1, 2$) as a result of dissociation [1]. Previous studies on methane ionization irradiated by pulses of nanosecond duration showed an absence of the parent fragment CH_4^+ . This was explained as a result of the dissociation into the CH_3^+ and a hydrogen atom. On the other hand, when the pulse duration was decreased down to a few femtoseconds all the fragment ions disappeared except for the CH_4^+ and CH_3^+ molecular ions indicating that methane did not have enough time to dissociate after the ionization step. All the aforementioned studies were confined to the fundamental wavelength of 800 nm. In the present work, we extend the study to include the second harmonic field at 400 nm.

[1] Mathur and Rajgara, Journal of Chemical Physics **124** (2006).

MO 9.3 Mon 16:45 S HS 002 Biologie

Static coherent states method: one and two electron laser-induced systems with classical nuclear dynamics — ●MOHAMMAD REZA EIDI¹, MOHSEN VAFAEE², and ALEXANDRA LANDSMAN^{1,3} — ¹Max Planck Institute for the Physics of Complex Systems, Dresden, Germany — ²Department of Chemistry, Tarbiat Modares University, Tehran, Iran — ³Max Planck Korea, Department of Physics, Postech, Pohang, Gyeongbuk 37673, Republic of Korea

In the presented work, the static coherent states method (SCS) is introduced to investigate quantum electron dynamics in a one or two-electron laser-induced system. The SCS method solves the time-dependent Schrödinger equation (TDSE) both in imaginary and real times on the base of a static grid of coherent states. Moreover, classical dynamics is considered for the nuclei by solving their Newtonian equation of motion. Implementing classical nuclear dynamics, the electronic state potential energy curves of H_2^+ in the absence and presence

of an ultra-short intense laser field is computed. We use this method to investigate charge migration in H_2^+ . In particular, we find that the charge migration time increases exponentially with internuclear distance. We also observe substantial charge localization for sufficiently long molecular bonds.

MO 9.4 Mon 17:00 S HS 002 Biologie

Probing electronic structure via molecular-frame photoelectron imaging — ●JOSS WIESE^{1,2}, SEBASTIAN TRIPPEL^{1,3}, and JOCHEN KÜPPER^{1,3,4} — ¹Center for Free-Electron Laser Science, Deutsches Elektronen-Synchrotron DESY, Hamburg — ²Department of Chemistry, Universität Hamburg — ³The Hamburg Center for Ultrafast Imaging, Universität Hamburg — ⁴Department of Physics, Universität Hamburg

Chemical function arises from the interplay among valence electrons. Thus, 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 molecular-frame photoelectron imaging of spatially confined ensembles of indole strong-field ionised by intense near-infrared laser pulses. Reconstructed static three-dimensional photoelectron distributions in molecular-frame momentum space (MF-ARPES) will be presented. The evaluation of the intensity differentials of these MF-ARPES allows for the mapping of the molecule's valence electronic structure and enables the investigation of strong-field ionisation phenomena in the intermediate regime between multi-photon and tunnel ionisation.

MO 9.5 Mon 17:15 S HS 002 Biologie

On the Role of singly-excited Rydberg states in Sequential H2 Multiphoton Ionization — ●PATRICK FROSS, DENHI MARTINEZ, NICOLAS CAMUS, THOMAS PFEIFER, and ROBERT MOSHAMMER — Max-Planck-Institut für Kernphysik, Heidelberg, Germany

Nowadays Sequential Multiphoton Ionization of H₂ is mainly understood in terms of a stepwise mechanism. First the ground state molecule is ionized by removing one electron. Then eventually dissociation takes place and finally there is a chance of removing the second and last electron. We present COLTRIMS results that show that this mechanism is not correct for 35fs laser pulses with a central wavelength of 400nm ($I=1 \cdot 10^{14}$ W/cm²). Our observations demonstrate that singly-excited Rydberg states play a major role in the dynamics of photoionization. In addition the underlying ionization pathway is explained.

MO 9.6 Mon 17:30 S HS 002 Biologie

An alternative dissociation pathway of N_2^{2+} — LAZAROS VARVAREZOS¹, ●ALEXANDER ACHNER², RENÉ WAGNER², JOHN COSTELLO¹, MICHAEL MEYER², and PATRIK GRYSHTOL² — ¹School of Physical Sciences and NCPST, Dublin City University, Dublin 9, Ireland — ²Small Quantum Systems Group, European XFEL GmbH, 22869 Schenefeld, Germany

Since the advent of lasers as spectroscopic tools, N_2 has served as a benchmark system to shed light on the phenomena induced by intense laser fields. In our study, we focus on the dissociation mechanism of the molecular dication N_2^{2+} . Our interest in the dication was stimulated by a study that revealed the presence of two different $N^+ + N^+$ states, energetically separated by about 4eV for the first time [1]. Our findings suggest the presence of two ion pairs, separated by approximately 2eV originating from fast and slow decay channels. Most likely, the fast pair of ions stems from the $1^1\Sigma_g^+$ state after the removal of two $3\sigma_g$ electrons, while the slow ion pair results from the $3^1\Pi_u$ state of the molecular ion after the removal of one $3\sigma_g$ electron and one $1\pi_u$ electron. Furthermore, we reach to the very important conclusion that the slow channel is the precursor for the N_2^{2+} ions, due to the existence of a strong intensity dependence, which is not present in the case of the faster ions.

[1] Lai et al., Physical Review A **84**, 043413 (2011).