

MO 5: X-Ray and XUV Spectroscopy (joint session MO/A)

Time: Monday 14:00–15:45

Location: PA 2.150

Invited Talk

MO 5.1 Mon 14:00 PA 2.150

Theoretical soft X-ray spectroscopy of transition metal compounds: A multi-reference wave function approach — ●SERGEY I. BOKAREV — Institut für Physik, Universität Rostock, Albert-Einstein-Str. 23-24, 18059, Rostock

To date, X-ray spectroscopy has become a routine tool that can reveal highly local and element-specific information on the electronic structure of atoms in complex environments. Here, we aim at the development of an efficient and versatile theoretical methodology for the treatment of soft X-ray spectra of transition metal compounds mainly based on the multi-configurational self-consistent field electronic structure theory combined with a perturbative LS-coupling scheme for spin-orbit coupling. A special focus is put on the L-edge photon-in/photon-out and photon-in/electron-out processes, i.e. X-ray absorption, resonant inelastic scattering, partial fluorescence yield, photoelectron and Auger spectroscopy treated on the same theoretical footing. We address the application of the X-ray metal L-edge and ligand K-edge as well as XUV spectroscopy to unraveling electronic structure and nature of chemical bonds, oxidation and spin-states, the interplay of radiative and non-radiative decay channels, fingerprints of nuclear dynamics and non-adiabatic transitions as well as ultrafast electron dynamics triggered by X-ray light. The investigated systems range from the small prototypical coordination compounds and catalysts to the aggregates of biomolecules.

MO 5.2 Mon 14:30 PA 2.150

Solving the Graphene Oxide Puzzle - a TDDFT-XAS Study — ●FABIAN WEBER, JIAN REN, TRISTAN PETIT, and ANNIKA BANDE — Helmholtz-Zentrum Berlin

Graphene Oxide (GO) quantum dots and its derivatives have proven to be a resource efficient material for photocatalytic water splitting. Due to the amorphous nature of GO derived materials, it has however not been possible yet to fully understand what structural features are allowing the catalytic reaction.

Since X-Ray spectroscopy is a standard technique to identify neighbouring functionalities in a very selective way, we developed a time-dependent density functional theory (TDDFT) approach to probe the constitution of single specific atoms in model systems in a meaningful way. Since these localized Carbon K-edge XAS-spectra have shown to be specific up to about 3 chemical bonds, we may use a multitude of model systems of relatively small size to generate a surrounding-specific database of several unique functionalization patterns.

In this talk we show how systematic comparison of this database of several hundreds of different surrounding-specific XAS spectra with experimental data leads to a straightforward method to gain insights on structural features of amorphous materials.

MO 5.3 Mon 14:45 PA 2.150

Hydrogen bond dissociation dynamics of indole-water clusters — ●MELBY JOHNY¹, THOMAS KIERSPEL^{1,2}, JOSS WIESE^{1,2}, JOLIJN ONVLEE¹, HELEN BIEKER^{1,2}, TERRY MULLINS^{1,2}, ANDREA TRABATTONI¹, RUTH LIVINGSTONE^{1,2}, SEBASTIAN TRIPPEL^{1,2}, JOCHEN KÜPPER^{1,2,3}, and AND OTHERS^{1,2,3} — ¹Center for Free Electron Laser Science (CFEL), Deutsches Elektronen Synchrotron (DESY), Hamburg, Germany — ²The Hamburg Center for Ultrafast Imaging, University of Hamburg, Germany — ³Department of Physics, University of Hamburg, Germany

The properties of atoms and molecules are strongly dependent on their local environment and hydrogen bonds are of universal importance in chemistry and biochemistry. Therefore, it is highly desirable to bridge the gap between single, isolated molecules and molecules in solvation. Photophysics of microsolvated indole is significantly relevant being strongest UV chromophore of tryptohan as well as the structure of singly hydrogen bonded indole-water cluster is known [1].

Here, we show our recent experiment performed at LCLS at SLAC for the investigation of hydrogen bond breaking dynamics of indole-water clusters via UV-pump (266 nm) and x-ray probe (2.5 nm) photoelectron-photoion-photoion-coincidence (PEPIPICO) imaging. Our preliminary analysis shows time-dependent dissociation channels with distinguishable velocities for hydronium ions detected in coincidence with the photo-fragments of indole.

[1] Korter, Pratt, Küpper, J. Phys. Chem. A 1998, 102, 7211-7216

MO 5.4 Mon 15:00 PA 2.150

Time-Resolved Pump-Probe Spectroscopy of XUV-induced Dynamics in Water, Ammonia and Methanol Clusters — ●RUPERT MICHIELS¹, AARON LAForge¹, MATTHIAS BOHLEN¹, CARLO CALLEGARI², ANDREW CLARK³, MARCEL DRABELLS³, KEVIN C. PRINCE², STEFANO STRANGES⁴, MARCELLO CORENO⁵, OKSANA PLEKAN², VERONICA OLIVER³, AARON VON CONTA⁶, MARTIN HUPPERT⁶, HANS-JAKOB WÖRNER⁶, and FRANK STIENKEMEIER¹ — ¹Universität Freiburg, Germany — ²Elettra-Sincrotrone Trieste, Italy — ³Ecole Polytechnique Fédérale de Lausanne, Switzerland — ⁴University Sapienza, Italy — ⁵ISM-CNR, Trieste, Italy — ⁶ETH Zürich, Switzerland

Results for ammonia, methanol and water clusters investigating the dynamics upon XUV ionization with the FERMI free electron laser are presented. In an XUV-UV pump-probe scheme we obtain a time-resolved picture of the resulting fragments and their electronic states. Recombination and molecular dissociation lead to a great variety of electron energies and ionic fragments that give insight into the underlying dynamics. The vertical detachment energies of the electrons and masses of the ions were measured by means of a VMI velocity map imaging detector combined with a time-of-flight mass spectrometer which also allows a covariance analysis.

Invited Talk

MO 5.5 Mon 15:15 PA 2.150

Tracing the spatial and electronic structure of excited molecules using X-ray FEL and HHG light — ●KIRSTEN SCHNORR — Max-Planck-Institut für Kernphysik — University of California Berkeley

Free-Electron Lasers (FELs) are capable of producing intense and ultrashort X-ray pulses, which enable femtosecond time-resolved diffractive imaging experiments. This allows to initiate chemical reactions in molecules using an optical pump pulse and probing the induced changes in the nuclear structure by X-ray scattering using a delayed FEL pulse. Here, results on the strong-field induced dynamics of C₆₀ molecules probed by soft and hard X-ray scattering will be presented.

A soft X-ray source based on high harmonic generation with photon energies up to 310 eV has been successfully commissioned and first time-resolved experiments carried out. Transient absorption experiments on the UV-induced dynamics of small carbon containing molecules, probed with a broadband soft X-ray pulse, will be shown.