A 29: Interaction with VUV and X-ray light I

Time: Thursday 11:00–13:15 Location: C/HSW

Invited Talk A 29.1 Thu 11:00 C/HSW

Electronic structure in high-intensity x-ray fields — •Robin Santra — Center for Free-Electron Laser Science, DESY, Hamburg, Germany — Department of Physics, University of Hamburg, Hamburg, Germany

Generally, the probability that a given atom in a material absorbs an x-ray photon in a single x-ray pulse is much less than unity for storage-ring-based x-ray sources, even for third-generation synchrotron radiation sources. This situation has changed dramatically with the arrival of x-ray free-electron lasers: In the micro-focus of an x-ray free-electron laser, saturation of x-ray photoabsorption is routinely achieved. The immediate consequence is that the overall behavior of matter under such extreme conditions is characterized by efficient multiphoton absorption via a sequence of single-photon absorption events combined with inner-shell decay cascades and collisional ionization processes. In this way, unusual, highly excited states of matter are formed, whose properties and dynamics are challenging to describe theoretically. In this talk, I will discuss progress that has been made towards developing suitable electronic structure tools. The performance of currently available electronic structure tools will be assessed by direct comparison with experimental data.

A 29.2 Thu 11:30 C/HSW

Theoretical characterization of the collective resonance states underlying the xenon giant dipole resonance — •Yi-Jen Chen[1,2], Stefan Pabst[1], and Robin Santra[1,2] — [1]Center for Free-Electron Laser Science, DESY, Hamburg, Germany — [2]Department of Physics, University of Hamburg, Hamburg, Germany

We present a detailed theoretical characterization of the two fundamental collective resonances underlying the xenon giant dipole resonance (GDR). This is achieved consistently by two complementary methods implemented within the framework of the configuration-interaction singles (CIS) theory. The first method accesses the resonance states by diagonalizing the many-electron Hamiltonian using the smooth exterior complex scaling technique. The second method involves a new application of the Gabor analysis to wave-packet dynamics. We identify one resonance at an excitation energy of 74 eV with a lifetime of 27 as, and the second at 107 eV with a lifetime of 11 as. Our work provides a deeper understanding of the nature of the resonances associated with the GDR: a group of close-lying intrachannel resonances splits into two far-separated resonances through interchannel couplings involving the 4*d* electrons. The CIS approach allows a transparent interpretation of the two resonances as new collective modes. Due to the strong entanglement between the excited electron and the ionic core, the resonance wave functions are not dominated by any single particle-hole state. This gives rise to plasma-like collective oscillations of the 4*d* shell as a whole.

A 29.3 Thu 11:45 C/HSW

Theoretical calculation of above-threshold ionization of xenon involving the giant dipole resonance — •Antonia Karamatskou[1,2], Robin Santra[1,2], Tommaso Mazza[3], and Michael Meyer[3] — [1]Center for Free-Electron Laser Science, DESY, Hamburg, Germany — [2]Universität Hamburg, Hamburg, Germany — [3]European XFEL GmbH, Hamburg, Germany

We present a theoretical study on above-threshold ionization (ATI) of xenon in the photon energy range between 100 and 150 eV. Our numerical method relies on the exact solution of the Schrödinger equation within the time-dependent configuration interaction singles scheme (TDCIS). Within TDCIS it is possible to include and distinguish certain electronic correlation effects that are mediated by Coulomb interaction. In this way, the influence of collectiveness on the process studied can be quantified. Analyzing the 2-photon ATI cross section we find that, in contrast to the 1-photon ionization case, in the nonlinear regime two distinct resonance states underlying the giant dipole resonance can be resolved. The existence of two energy poles was predicted already in the 70's by Wendin [J.Phys. B At. Mol. Opt. Phys. 6, 42 (1973)]. We compare our theoretical results to recent measurements performed at the free-electron laser FLASH in Hamburg, which provided nonlinear electron-spectroscopy data on xenon in the XUV energy range.

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A 29.4 Thu 12:00 C/HSW

Time-resolved relaxation dynamics in resonantly excited He nanodroplets — •Aaron LaForge[1], Alessandra Ciavardini[2], Mike Ziemkiewicz[3], Yevheniy Ovcharenko[4], Oksana Plekan[5], Paola Finetti[5], Robert Richter[5], Kevin Prince[5], Paolo Piseri[6], Michele Di Friaia[7], Arik Mika[8], Marcel Drabbels[8], Carlo Callegari[5], Thomas Moeller[4], Frank Stienkemeier[1], Patrick O'Keeffe[2], and Marcel Mudrich[1] — [1]Universität Freiburg, 79104 Freiburg, Germany — [2]CNR_IMP, 00016 Monterotondo Scalo, Italy — [3]University of California, Berkeley, California 94720, USA — [4]TU Berlin, 10623 Berlin, Germany — [5]Elettra, Basovizza, 34149 Trieste, Italy — [6]Università degli Studi di Milano, 20133 Milano, Italy — [7]University of Trieste, 34128 Trieste, Italy — [8]EPFL, CH-1015 Lausanne, Switzerland

The relaxation dynamics of resonantly excited helium droplets by XUV radiation has been investigated. Using an XUV-UV pump-probe setup, we initially resonantly excite the droplet and observe the various relaxation pathways prior to UV ionization. From the results we can clearly see an initial droplet band relaxation followed by the electron relaxing to an atomic state. Here, we will present the time resolved results for various resonant excitations and droplet sizes.

A 29.5 Thu 12:15 C/HSW

Two-color X-ray pump X-ray probe study of core-hole decay dynamics — Carl Stefan Lehmann[1], Antonio Picón[1], Steve Southworth[1], Gilles Doumy[1], Anne Marie March[1], Dooshaye Moonshiram[1], Bertold Krässig[1], Elliot Kanter[1], Linda Young[1], Benjamin Erk[2], Cedric Bomme[2], Daniel Rolles[2,3], Artem Rudenko[3], •Steve Pratt[4], Dipanwita Ray[5], Timur Osipov[6], Nora Berrah[6], and Christoph Bostedt[7] — [1]X-ray Science Division, Argonne National Laboratory — [2]Max-Planck-Advanced Study Group at CFEL, DESY — [3]Department of Physics, Kansas State University — [4]Chemical Sciences & Engineering Division, Argonne National Laboratory — [5]Chemical Science Division, Lawrence Berkeley National Laboratory — [6]Department of Physics, UCONN — [7]LCLS, SLAC National Accelerator Laboratory

To resolve the femtosecond inner-shell dynamics and the subsequent induced electron transfer in a molecule, the core-hole decay dynamics in XeF2 have been directly studied using femtosecond time-resolved x-ray pump x-ray probe coincidence imaging.

The recently developed capability at LCLS was used to generate two-color x-ray pulses with variable delay. A time and position sensitive detector has been used to record the ion fragments in coincidence.

XeF2 is a very interesting molecule, as it allows us to compare the molecular core-hole decay with the atomic case, Xe atom. Hence, we used a specific photon energy for the pump pulse to induce a core-hole in the Xe site, and with the probe pulse we ionize again for different time delays and photon energies, both for Xe atom and XeF2 molecule.

A 29.6 Thu 12:30 C/HSW

Testing calculated potential energy curves of molecular oxygen by measuring vibrational wave-packets with an XUV–IR experiment — •Philipp Cörlin, Andreas Fischer, Michael Schönwald, Tomoya Mizuno, Thomas Pfeifer, and Robert Moshammer — Max-Planck-Institut für Kernphysik, Heidelberg

In our experiment XUV photons (18 eV to 40 eV) created from 10 fs IR laser pulses by high-harmonic generation are used to ionize molecular oxygen. The ionic state is probed by a delayed IR pulse with an effective intensity of 3e12 W/cm^2. Charged fragments are detected with a reaction microscope and a pronounced oscillation in the count rate of very low energetic O^+ ions is measured. The low KER and the oscillation period of 40.0 fs point towards a wave-packet oscillation within the binding ionic $a^4\Pi_u$ potential which is probed via the weakly repulsive $f^4\Pi_g$ state by resonant absorption of an IR photon [1, 2]. By comparison of experimental spectra with the results of a coupled channel simulation, we are able to test different sets of calculated potential curves. While the agreement between experiment and theory is on a level not observed for O_2^+ wave-packets before, some properties of the experimental spectra cannot be reproduced in a satisfying manner by our simulation, if calculated binding potentials are used. In order

to demonstrate the sensitivity on the shape of the binding potential curve, we perform a simulation with a Morse potential fitted to the experimental data which reproduces the experimental spectra very well.
[1] S. De *et al.*, Phys. Rev. A 84, 043410 (2011)
[2] M. Magrakvelidze *et al.*, Phys. Rev. A 86, 023402 (2012).

A 29.7 Thu 12:45 C/HSW

Single-shot autocorrelation in the vacuum ultraviolet spectral range — •Dimitrios Rompotis[1,2,3], Thomas Gebert[1,2,3], Marek Wieland[1,2,3], Theophilos Maltezopoulos[1,2,3], and Markus Drescher[1,2,3] — [1]Institut fuer Experimentalphysik Universitaet Hamburg, Hamburg, Germany — [2]The Hamburg Centre for Ultrafast Imaging (CUI), Hamburg, Germany — [3]Centre for Free-Electron-Laser Science, Hamburg, Germany

We introduce a method for single-shot time metrology in the VUV based on a non-linear autocorrelation scheme. In an all-reflective design, the technique is applicable to a wide spectral range, from the infrared to the extreme ultraviolet.

Single-shot autocorrelation is demonstrated for the first time, to our knowledge, in the VUV spectral range. Non-resonant two-photon ionization of Krypton is used to obtain a second-order autocorrelation measurement of an intense, Ti:Sa fifth-harmonic pulse at 161.8 nm.

In addition to ultrashort pulse characterization, the experimental scheme is being utilized in a single-shot, VUV pump-VUV probe configuration, for the investigation of ultrafast dynamics. We present results, demonstrating the feasibility of non-linear, time-resolved molecular spectroscopy in the vacuum ultraviolet.

A 29.8 Thu 13:00 C/HSW

Time-Dependent Multiphoton Ionization of Xenon in the Soft-X-Ray Regime — Nils Gerken[1], Stephan Klumpp[1], Andrey Sorokin[2,3], Kai Tiedtke[2], •Mathias Richter[4], Vera Bürk[1], Karolin Mertens[1], Pavle Juranić[2], and Michael Martins[1] — [1]Universität Hamburg, Institut für Experimentalphysik, Luruper Chaussee 149, 22761 Hamburg, Germany — [2]Deutsches Elektronen-Synchrotron, DESY, Notkestraße 85, 22603 Hamburg, Germany — [3]Ioffe Physico-Technical Institute, Polytekhnicheskaya 26, 194021 St. Petersburg, Russia — [4]Physikalisch-Technische Bundesanstalt, PTB, Abbestraße 2-12, 10587 Berlin, Germany

The time-dependent multiphoton ionization of xenon atoms was studied with femtosecond pulses in the excitation range of the 4d giant resonance at the photon energy of 93 eV [1]. Benefiting from a special operation mode of the free electron laser FLASH, the measurements were performed with varying pulse durations. A strong dependence of the ion charge distribution on the pulse duration allowed the different multiphoton mechanisms behind the multiple photoionization of xenon to be disentangled up to a charge state of 10+. The results up to 8+ are well explained by sequences of single photon, multiphoton, and Auger processes, but higher charge state generation suggests the need for collective electron multiphoton excitations.

[1] N. Gerken, S. Klumpp, A. A. Sorokin, K. Tiedtke, M. Richter, V. Bürk, K. Mertens, P. Juranić, M. Martins, Phys. Rev. Lett. 112, 213002 (2014)