

A 3: XUV/X-ray spectroscopy I

Time: Monday 14:30–16:30

Location: N 2

Invited Talk

A 3.1 Mon 14:30 N 2

Excited state molecular dynamics: new insights from x-ray spectroscopy and scattering — ●MARKUS GÜHR — Institut für Physik und Astronomie, Universität Potsdam

The conversion of light energy into other energetic forms is accomplished via electronically excited states. Visible and ultraviolet light initially deposits energy in the molecular electronic degrees of freedom. The coupling between electronic and nuclear degrees of freedom on an ultrafast timescale determines the efficiency and selectivity of energy conversion into heat, chemical transformation or charge transfer. Understanding of the energy conversion processes therefore requires information on electronic and nuclear molecular degrees of freedom.

Time resolved spectroscopy using femtosecond x-ray pulses provides a new window into the molecular excited states dynamics. Due to the x-ray typical element and site selectivity, the method provides highly local information on nonadiabatic dynamics. I will discuss the progress in this field and present new experiments on the excited state decay from a $\pi\pi^*$ state to an $n\pi^*$ state, probed at the oxygen K-edge [1].

In addition, I will present recent approaches to directly image molecular dynamics using x-rays and relativistic electron pulses. For the latter I will show how electrons can be used to directly image vibrational wavepackets in an isolated molecule [2].

[1] T. Wolf, R. Heilemann Myhre et al., arXiv:1610.08498 (2016)

[2] Yang, J., Guehr et al., Phys. Rev. Lett. 117, 173002 (2016)

A 3.2 Mon 15:00 N 2

Optical control of core hole relaxation dynamics in open shell atoms — ●ALEXANDER ACHNER¹, TOMMASO MAZZA¹, CARLO CALLEGARI², STEFANO STRANGES³, and MICHAEL MEYER¹ — ¹European XFEL GmbH, 22869 Schenefeld, Germany — ²Electra-Sincrotrone Trieste, 34149 Basovizza, Trieste, Italy — ³Università degli Studi di Roma La Sapienza, 00185 Rome, Italy

To explore the relaxation dynamics of resonantly excited core-hole states of atomic iodine, we used XUV pulses from the FERMI FEL in combination with a synchronized near-infrared (NIR) laser. The ionization process was characterized by ion time of flight spectrometry. After resonant 4d excitation by the XUV radiation the NIR laser field is used to probe the excited states as well as the final ionization products. The I^*4d^96p and the I^*4d^95p resonances (at 55.95 eV and 46.2 eV) were excited by the XUV photons and the dynamics of the electronic decay was probed by varying the relative delay between XUV and NIR. Using only XUV radiation, the resonant excitation is followed by an ultrafast resonant Auger decay leading mainly to singly charged states. Introducing the NIR laser pulses the ionization of the $4d^96p$ resonant state is possible, causing the production of higher charge states via normal Auger process. For the $4d^95p$ resonance the energy difference to the 4d ionization threshold is too high for further ionization with NIR intensities used in the experiment, and therefore only ionization of the excited singly charge states produced via resonant Auger decay was possible.

A 3.3 Mon 15:15 N 2

Site-selective x-ray photofragmentation of molecules — ●LUDGER INHESTER¹, SANG-KIL SON¹, and ROBIN SANTRA² — ¹Center for Free-Electron Laser Science, DESY, Hamburg — ²Department of Physics, University Hamburg

For understanding the impact of radiation damage in biological processes it is important to know how a molecule fragments after x-ray absorption. After x-ray ionization of a core electron and subsequent Auger decay the molecular electronic structure is left in a two-valence hole configuration. This two-valence hole configuration typically initiates molecular dissociation. In contrast to the core electron, the valence holes are often delocalized over large parts of the molecule. Thus, ionization on a specific atomic site may lead to disruption of the molecule at remote parts, which makes it difficult to predict into which fragments the molecule breaks apart. Because of the large number of accessible two-hole configurations the quantitative theoretical prediction of molecular fragments is a challenging task.

We address this issue using our newly developed XMOLECULE toolkit (Y. Hao et al., Struct. Dyn. 2 (2015) 041707, L. Inhester et al., Phys. Rev. A, 94 (2016) 023422). Based on calculated Auger transition rates and Mayer's bond order analysis, we present a way

to efficiently calculate the molecular fragments after x-ray absorption. Results for ethyl trifluoroacetate ($\text{CF}_3\text{-CO-O-CH}_2\text{-CH}_3$, a.k.a. the Siegbahn or the ESCA molecule) are compared with recent experiments. With these results we demonstrate that the abundance of certain molecular fragments is specific for x-ray ionization on a particular atomic site.

A 3.4 Mon 15:30 N 2

Calibration of the fluence of an x-ray free-electron laser using XATOM — ●KOUICHI TOYOTA, ZOLTAN JUREK, SANG-KIL SON, and ROBIN SANTRA — CFEL, DESY, Hamburg

We report on a numerical procedure to calibrate the spatial intensity profile of x-ray free-electron laser pulses (XFELs). Extracting the reliable pulse parameters is indispensable to quantitatively analyze the experimental results and eventually to understand the non-linear physics in matter induced by high intensity XFELs. So far, we have conducted calibrations based on charge state distributions (CSDs) of atomic ions [1] calculated by XATOM toolkit [2]. Such a method has the advantage that the calibration is based on observables directly reflecting the experimental conditions within the interaction volume. By convolving theoretical CSDs with a spatial fluence distribution of a double Gaussian shape profile, which is defined by a few parameters, the experimental CSD was successfully recovered. However, our previous analysis was lacking a comprehensive framework and was manually implemented.

Our new procedure is now built with a well established optimization techniques in numerical analysis and fully automatized via computer codes. Therefore it offers a possibility of deeper investigations based on CSDs over a wide range of a parameter space. We discuss the procedure of optimization algorithms and test different pulse profile ansatz. We demonstrate the workflow and its practical importance on real experimental data. [1] B.F. Murphy et al., Nat. Commun. 5, 4281 (2014); [2] S.-K. Son and R. Santra, XATOM - an integrated toolkit for x-ray and atomic physics, CFEL, DESY, Hamburg, Germany, 2011

A 3.5 Mon 15:45 N 2

Auger cascades in resonantly excited neon — ●SEBASTIAN STOCK^{1,2}, RANDOLF BEERWERTH^{1,2}, and STEPHAN FRITZSCHE^{1,2} — ¹Helmholtz-Institut Jena, 07743 Jena, Germany — ²Friedrich-Schiller-Universität Jena, 07743 Jena, Germany

Auger electron spectroscopy is an important tool for probing the inner-shell structure of atoms and ions. Over the past few decades, multi-electron coincidence spectroscopy has led to substantial progress in our understanding of multiple ionization via Auger processes.

We here present a theoretical study [1] of the Auger decay paths following resonant $1s \rightarrow np$ ($n = 3, 4$) excitation of neutral neon. In contrast to the usual approach that often considers just a few transitions, we model the complete cascade decay paths from neutral to doubly ionized neon. By performing extensive MCDF calculations, we include the effects of electron correlation in the calculation of the transition rates of many possible decays. In addition, we also account for shake processes, which play a significant role in some of these transitions. In order to improve our results, especially for low-energy transitions, we employ experimental values for the energy levels of neutral, singly and doubly ionized neon.

By modeling the complete cascade, we are able to predict electron spectra, shake probabilities, ion yields, and the population of intermediate and final states. Our results are in good agreement with experimental findings, e.g. from Ref. [2].

[1] S. Stock, R. Beerwerth, and S. Fritzsche, to be published.

[2] Y. Tamenori and I. H. Suzuki, *J. Phys. B* **47**, 145001 (2014).

A 3.6 Mon 16:00 N 2

Relativistic calculations of the non-resonant two-photon K-shell ionization of neutral atoms — ●JIRI HOFBRUCKER^{1,2}, ANDREY VOLOTKA^{1,3}, and STEPHAN FRITZSCHE^{1,2} — ¹Helmholtz-Institut Jena, Germany — ²Friedrich-Schiller-Universität, Jena, Germany — ³St. Petersburg State University, Russia

The non-resonant two-photon one-electron K-shell ionization of neutral atoms is studied within the framework of relativistic second-order perturbation theory and independent particle approximation. The importance of relativistic and screening effects in the total as well as dif-

ferential cross sections is investigated. Our results show that, at near two-photon ionization threshold energies, the account for the screening effects of the remaining electrons leads to occurrence of an unexpected minimum in the total two-photon ionization cross section [1] and to elliptical dichroism in the photoelectron angular distribution. For ionization of heavy atoms, relativistic effects result in a significant decrease of the total cross section [2], and in distortion of the angular distribution into forward direction.

[1]J. Hofbrucker, A. V. Volotka, S. Fritzsche, accepted in Phys. Rev. A.

[2]J. Hofbrucker, A. V. Volotka, S. Fritzsche, submitted to Nucl. Instr. Meth. Phys. Res.

A 3.7 Mon 16:15 N 2

State-of-the-art MMC detector arrays for X-ray spectroscopy

— •D. HENGSTLER¹, F. MÜCKE¹, J. GEIST¹, M. KELLER¹, M. KRANTZ¹, C. SCHÖTZ¹, R. PONS¹, S. KEMPF¹, L. GASTALDO¹, A. FLEISCHMANN¹, C. ENSS¹, T. GASSNER^{2,3}, K.H. BLUMENHAGEN^{2,3}, R. MÄRTIN^{2,3}, G. WEBER^{2,3}, and TH. STÖHLKER^{2,3,4} — ¹KIP, Hei-

delberg University — ²Helmholtz-Institute Jena — ³GSI Darmstadt — ⁴IOQ, Jena University

Metallic magnetic calorimeters (MMCs) are energy dispersive X-ray detectors which provide a very good energy resolution over a large dynamic range combined with an excellent linearity. MMCs are operated at millikelvin temperatures and convert the energy of an incoming X-ray into a temperature rise which is monitored by a paramagnetic temperature sensor. The resulting change of sensor magnetization is read out by a SQUID and serves as a measure for the energy input. We developed several micro-fabricated one- and two-dimensional detector arrays optimized for energies up to 20, 30 and 200 keV providing energy resolutions as good as 1.6 eV, 7 eV and 26 eV, respectively. The detectors are operated in a mobile dilution refrigerator. We discuss the performance of our detectors and show results of our most recent 8x8 pixel detector array providing an active detection area of 4 mm x 4 mm. We present promising results of first beamline experiments at ESR@GSI showing that MMCs are an ideal tool for future precision experiments at FAIR as well as new detector designs optimized for upcoming beamtimes at Cryring@ESR.