

MO 1: Clusters I (joint session MO/A)

Time: Monday 11:00–13:15

Location: f102

Invited Talk

MO 1.1 Mon 11:00 f102

Infrared Spectroscopy of HHe_n^+ and DHe_n^+ Complexes — ●OSKAR ASVANY and STEPHAN SCHLEMMER — I. Physikalisches Institut, Universität zu Köln, Zùlpicher Str. 77, Köln

The combination of a cryogenic ion-trap machine, operated at 4.7 K, with the free-electron-laser FELIX allowed the first experimental characterization of the antisymmetric stretch (ν_3) and bending (ν_2) fundamentals of the linear He-X⁺-He (X=H, D) cores of the in situ prepared HHe_n^+ and DHe_n^+ complexes for $n = 3 - 6$. The found band origins, at around 1290 cm^{-1} for ν_3 and around 850 cm^{-1} for ν_2 (for HHe_n^+), are fully supported by first-principles quantum-chemical computations. These results are consistent with structures for the species with $n = 3$ and 6 being of T-shaped C_{2v} and of D_{4h} symmetry, respectively, while the species with $n = 4$ are suggested to exhibit interesting dynamical phenomena related to large-amplitude motions. Future high-resolution measurements will give detailed insight into the structure of these fundamental complexes.

MO 1.2 Mon 11:30 f102

Photon-energy dependent ionisation of isolated Xenon atoms

— THOMAS BAUMANN¹, REBECCA BOLL¹, MARKUS ILCHEN¹, JOAKIM LAKSMAN¹, TOMMASO MAZZA¹, MICHAEL MEYER¹, VALERIA MUSICI¹, DANIEL RIVAS¹, ●ALJOSCHA RÖRIG¹, SVITOZAR SERKEZ¹, PHILIPP SCHMIDT¹, SERGEY USENKO¹, JULIA SCHÄFER², ROBIN SANTRA², SANG-KIL SON², SHASHANK PATHAK³, DANIEL ROLLES³, and BENJAMIN ERK⁴ — ¹European XFEL GmbH, Schenefeld, Germany — ²Center for Free-Electron Laser Science (CFEL), Hamburg, Germany — ³Kansas State University, Manhattan, USA — ⁴Deutsches Elektronen-Synchrotron (DESY), Hamburg, Germany

In October 2019 we investigated the photon-energy-dependence of the ionisation process of isolated Xenon atoms exposed to extremely intense soft X-ray radiation at the European XFEL free-electron laser in Schenefeld, Germany. We were able to tune the photon energy over a wide range (700 eV - 3000 eV) while maintaining constant high pulse energy of several millijoule. As a function of photon energy, we detected distinct variations of the ion yield in certain charge state regions due to non-linear effects, and extreme-charge states up to +42 of Xenon. These variations are the result of transient resonances that increase the photoionisation cross section at certain charge states and photon energies, as the binding energies in the atomic orbitals shift as a result of increasing charge state during a single FEL pulse. The experimentally obtained data is in good accordance with the calculated charge state distributions, thus allowing us a detailed insight into the transient electronic structure of highly excited atoms.

MO 1.3 Mon 11:45 f102

Oscillations in immiscible liquids due to persistent currents

— ●FREDERIC FOLZ, AYOUB ELYAAGOUBI, ALBRECHT OTT, and GIOVANNA MORIGI — Universität des Saarlandes

Absorption spectroscopy exhibits a noisy absorption pattern in samples of colloidal gold at marginal stability [1]. This might be the signature of an oscillatory behavior. Here, we discuss a theoretical model that traces these oscillations back to agglomeration and sedimentation processes, given a persistent influx of molecules. We analyse the results in relation to the experimental findings.

[1] Experiments carried out by Ayoub Elyaagoubi in the group of Prof. Ott at Saarland University, unpublished

MO 1.4 Mon 12:00 f102

Geometry and Stability of Small Cationic Silicon Carbide Clusters

— ●ROBERT RADLOFF, LARS DAHLLÖF, KAI POLLW, KARIM AHMED SAROUKH, MARKO FÖRSTEL, and OTTO DOPFER — Technische Universität Berlin, Institut für Optik und Atomare Physik, Germany

Ejecta of carbon-rich stars contribute significantly to interstellar dust. The emitted particles are generally hot and consist of single atoms or ions. With increasing distance from the star, this matter cools down and starts to condense, eventually forming first molecules and creating a rich chemistry. Observations suggest not only small molecules like SiC [1], Si₂C [2], SiC₂ [3], etc. to be present in the circumstellar environment of carbon-rich stars but also solid silicon carbide grains [4]. Up to now, it has remained unclear how these dust grains are created.

It seems likely that they form from small SiC molecules via condensation but no intermediates have been detected so far.

In this contribution we present the energetic, geometric, and optical properties of small cationic silicon carbide clusters obtained via photodissociation spectroscopy, mass spectrometry, and quantum chemical calculations. We show the first optical spectrum of a silicon carbide cation and present hitherto unreported fragmentation channels and ground state geometries of several cationic silicon carbide clusters.

[1] J. Cernicharo et. al., *Astrophys. J. Lett.* **341**, L25 (1989)

[2] J. Cernicharo et. al., *Astrophys. J. Lett.* **806**, L3 (2015)

[3] P. Thaddeus et. al., *Astrophys. J. Lett.* **283**, L45-48 (1984)

[4] R. Treffers and M. Cohen, *Astrophys. J.* **188**, 545-552 (1974)

MO 1.5 Mon 12:15 f102

N₂ activation on size selected Tantalum clusters investigated by cryo kinetics and IR spectroscopy

— ●MATTHIAS P. KLEIN¹, DANIELA V. FRIES¹, ANNIKA STEINER¹, HELMUT SCHWARZ², and GEREON NIEDNER-SCHATTEBURG¹ — ¹Fachbereich Chemie und Forschungszentrum OPTIMAS, TU Kaiserslautern, Erwin-Schrödinger-Straße 52, 67663 Kaiserslautern — ²Institut für Chemie, Technische Universität Berlin, 10623 Berlin

Catalytic N₂ activation by active transition metal surfaces is one of the most important industrial processes. For a thorough understanding of the activation steps, we need suitable model systems. By means of cryo IR-PD spectroscopy and cryo adsorption kinetics supported by DFT modelling, we are able to investigate the vibrational and kinetic behavior of various transition-metal-cluster-N₂/H₂ complexes (Fe, Co, Ni, Rh, Ru) and gain information about reaction precursors and intermediates. Small tantalum clusters have shown to activate N₂ dissociatively and even support the formation of ammonia. We chose to examine the Ta₄⁺ and Ta₅⁺ clusters in more detail. We observe a rearrangement of the adsorbate shell and activated N₂ upon stepwise N₂ adsorption. In the Ta₄⁺ case, the adsorption kinetics suggest a complex dynamics for large adsorption shells. A pseudo-first-order kinetic fit fails in this case. In contrast, the adsorption kinetics of the Ta₅ cluster are reproducible by such a fit. IR bands represent a range from weakly bound N₂ to actual N₂ activation precursors.

MO 1.6 Mon 12:30 f102

Optimizing Aerodynamic Lens Geometries for Gold Nanoparticle Beams

— ●LENA WORBS^{1,2}, JANNIK LÜBKE^{1,2,3}, ARMANDO ESTILLORE¹, AMIT K. SAMANTA¹, LOURDU XAVIER PAULRAJ¹, KARTIK AYYER^{1,4}, and JOCHEN KÜPPER^{1,2,3} — ¹Center for Free-Electron Laser Science, Deutsches Elektronen-Synchrotron DESY, Hamburg — ²Fachbereich Physik, Universität Hamburg — ³Center for Ultrafast Imaging, Hamburg — ⁴Max Planck Institute for the Structure and Dynamics of Matter, Hamburg

Coherent diffractive imaging at free-electron lasers promises to allow the reconstruction of the three-dimensional molecular structures of isolated particles at atomic resolution [1]. However, because of the typically low signal to noise ratio, this requires the collection of a large amount of diffraction patterns. Since every intercepted particle is destroyed by the intense x-ray pulse, a new and preferably identical sample particle has to be delivered into every pulse. One delivery technique is an aerodynamic lens stack (ALS), providing collimated or focused beams of nanoparticles. With the aid of numerical simulations we optimized the geometry of an ALS to produce high-density beams of nanoparticles, e.g., gold nanoparticles. Here, we present the simulation result for an optimized ALS and its experimental characterization using novel laser-based particle detection techniques [2]. We also show an application of ALS for XFEL experiments on gold nanoparticles.

[1] M. M. Seibert, et al., *Nature* **470**, 78 (2011).

[2] L. Worbs, et al., *Opt. Expr.*, arXiv:1909.08922 [physics.optics]

MO 1.7 Mon 12:45 f102

Chemical dynamics in Argon clusters induced by intense x-rays

— ●ZOLTAN JUREK^{1,2}, YOSHIKI KUMAGAI³, HIRONOBU FUKUZAWA^{3,4}, SANG-KIL SON^{1,2}, BEATA ZIAJA^{1,2,5}, ROBIN SANTRA^{1,2,6}, and KIYOSHI UEDA^{3,4} — ¹Center for Free-Electron Laser Science, Deutsches Elektronen-Synchrotron, Notkestrasse 85, 22607 Hamburg, Germany — ²The Hamburg Centre for Ultrafast Imaging, Luruper Chaussee 149, 22671 Hamburg, Germany — ³Institute of Multidisciplinary Research for Advanced Materials, Tohoku University,

Sendai 980-8577, Japan — ⁴RIKEN SPring-8 Center, Sayo, Hyogo 679-5148, Japan — ⁵Institute of Nuclear Physics, PAS, Radzikowskiego 152, 31-342, Krakow, Poland — ⁶Department of Physics, University of Hamburg, Jungiusstrasse 9, 20355 Hamburg, Germany

In this talk, we focus on the dynamics of rare gas clusters exposed to ultrashort X-ray Free Electron Lasers (XFELs) pulses of intermediate x-ray fluence. In this fluence regime the system is only partially ionized. Therefore, chemical properties of the sample may still play an important role in shaping the ionization dynamics beside the emerging Coulomb forces. As a result, complex fragmentation of the cluster can occur. We show signatures of chemical effects in spectroscopy observables, and present our theoretical simulations, unraveling the time evolution of irradiated Ar clusters in detail [1]. This study addresses a regime that is a very common scenario at current XFEL experiments. [1] Y. Kumagai et al, Phys. Rev. Lett. 120, 223201 (2018)

MO 1.8 Mon 13:00 f102

The role of water in the photophysics of indole — •LANHAI HE¹, MELBY JOHNY^{1,2}, THOMAS KIERSPEL^{1,2}, JOLIUN ONVLEE^{1,2}, BERND WINTER⁴, SEBASTIAN TRIPPEL^{1,2}, and JOCHEN KÜPPER^{1,2,3}

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Hydrogen bonds play an important role in biological processes and proteins, e.g., protein folding and molecular recognition. The influence of water in complex systems, such as a protein, can only be explained if the cluster formation and the breaking of the micro-solvated molecular clusters itself is understood. We studied the photo-fragmentation of indole and indole-water₁ upon x-ray absorption, which triggers inner-shell ionization from nitrogen and carbon (1s) orbitals. The generated ions and electrons were measured with an angle-resolved coincidence detection scheme, and the results revealed fragmentation-pathway-dependent electron spectra, linking the structural fragmentation dynamics to different electronic excitations. On the other hand, the photoelectron spectrum of an aqueous indole-water solution has been measured through x-ray ionization of indole in a liquid microjet. Analyzing the chemical shifts for the characteristic photoelectron spectra, the hydration of indole in water solutions is about to be identified.