

## MO 19: Clusters II (with A)

Time: Friday 11:00–13:00

Location: N 3

## Invited Talk

MO 19.1 Fri 11:00 N 3

**The Nanoplasma Oscilloscope** — ●CHRISTIAN PELTZ<sup>1</sup>, A. LAForge<sup>2</sup>, B. LANGBEHN<sup>3</sup>, R. MICHIELS<sup>2</sup>, C. CALLEGARI<sup>4</sup>, M. DI FRAIA<sup>4</sup>, P. FINETTI<sup>4</sup>, R. SQUIBB<sup>5</sup>, C. SVETINA<sup>4</sup>, L. RAIMONDI<sup>4</sup>, M. MANFREDDA<sup>4</sup>, N. MAHNE<sup>4</sup>, P. PISERI<sup>6</sup>, M. ZANGRANDO<sup>4</sup>, L. GIANNESI<sup>4</sup>, T. MÖLLER<sup>3</sup>, R. FEIFEL<sup>5</sup>, K. C. PRINCE<sup>4</sup>, M. MUDRICH<sup>2</sup>, D. RUPP<sup>3</sup>, F. STIENKEMEIER<sup>2</sup>, and T. FENNEL<sup>1</sup> — <sup>1</sup>Uni Rostock, Germany — <sup>2</sup>Uni Freiburg, Germany — <sup>3</sup>TU Berlin, Germany — <sup>4</sup>ELETTRA-Sincrotrone Trieste, Italy — <sup>5</sup>Uni Gothenburg, Sweden — <sup>6</sup>Uni Milan, Italy

Atomic clusters enable the well-controlled generation of nanoscale plasmas allowing for the study of their ultrafast light-induced correlated and collective dynamics. In particular, short-wavelength FELs can probe these dynamics in a regime that is fundamentally different from the well-known near-infrared domain. Plasma processes like collisional plasma heating, collective resonance excitation, and ionization avalanching that are generic in the NIR are strongly suppressed in the XUV and soft X-ray domain. Instead, sequential direct photo- or Auger emission dominates the plasma generation and heating dynamics. Signatures of this multistep ionization are characteristic plateau-like electron spectra and frustration of direct photo-emission by the cluster potential. Here we report the first direct time-resolved measurement of the underlying cluster potential evolution using the nanoplasma oscilloscope method, implemented in a recent two-color XUV pump-probe experiment at the seeded, high gain harmonic generation FEL FERMI FEL-2 operating in double stage mode.

MO 19.2 Fri 11:30 N 3

**NIR-induced Auger decay in clusters** — ●BERND SCHÜTTE, MARC VRAKING, and ARNAUD ROUZÉE — Max-Born-Institut, Berlin

In nanoplasmas, which are formed by the interaction of clusters with intense laser pulses, Rydberg states are efficiently populated. While it is well known that some of the excited atoms and ions relax via the emission of photons [1], nonradiative relaxation mechanisms have only recently been discovered at moderate cluster ionization [2,3]. Here we show that nonradiative decay remains important at high ionization degrees, as is demonstrated for CH<sub>4</sub> clusters that interact with 400-fs NIR pulses ( $I = 1 \times 10^{14}$  W/cm<sup>2</sup>). We observe a clear peak in the electron spectrum at 7 eV that is assigned to Auger decay, and that is explained by a 3-step process: (i) Our calculations show that the laser pulse removes almost all electrons from the outer shells of C, resulting in a dominant C<sup>4+</sup> ion contribution at the end of the laser pulse. (ii) Rydberg and outer-vacancy shell levels are populated by recombination, and (iii) relax via Auger decay. This picture is consistent with a dominant C<sup>3+</sup> ion contribution observed in the experiment, whereas, surprisingly, C<sup>+</sup> and C<sup>4+</sup> ion contributions are negligible.

Our results could explain the high average ion charge states that have been observed in clusters in spite of highly efficient recombination processes. Furthermore, the observed population inversion may be exploited for the development of an XUV or X-ray laser.

- [1] A. McPherson *et al.*, *Nature* **370**, 631 (1994).
- [2] B. Schütte *et al.*, *Phys. Rev. Lett.* **114**, 123002 (2015).
- [3] B. Schütte *et al.*, *Nat. Commun.* **6**, 8596 (2015).

MO 19.3 Fri 11:45 N 3

**The 3D shapes of spinning helium nanodroplets** — ●B. LANGBEHN<sup>1</sup>, Y. OVCHARENKO<sup>1,2</sup>, D. RUPP<sup>1</sup>, K. SANDER<sup>3</sup>, C. PELTZ<sup>4</sup>, A. CLARK<sup>4</sup>, R. CUCINI<sup>5</sup>, P. FINETTI<sup>5</sup>, M. DI FRAIA<sup>5</sup>, D. IABLONSKY<sup>6</sup>, A. C. LAForge<sup>7</sup>, V. OLIVER ÁLVAREZ DE LARA<sup>4</sup>, O. PLEKAN<sup>5</sup>, P. PISERI<sup>8</sup>, T. NISHIYAMA<sup>9</sup>, C. CALLEGARI<sup>5</sup>, K. C. PRINCE<sup>5</sup>, K. UEDA<sup>6</sup>, F. STIENKEMEIER<sup>7</sup>, T. FENNEL<sup>3</sup>, and T. MÖLLER<sup>1</sup> — <sup>1</sup>TU Berlin — <sup>2</sup>European XFEL — <sup>3</sup>Univ. Rostock — <sup>4</sup>EPFL, Lausanne — <sup>5</sup>Elettra-Sincrotrone Trieste — <sup>6</sup>Tohoku Univ. Sendai — <sup>7</sup>Univ. Freiburg — <sup>8</sup>Univ. di Milano — <sup>9</sup>Kyoto Univ.

Scattering techniques using intense femtosecond short-wavelength pulses from free-electron lasers (FEL) have been developed to gain an insight into the structure of nanoparticles such as viruses or clusters. Recent pioneering experiments in the hard X-ray range revealed that superfluid helium nanodroplets can gain high angular momentum resulting in large centrifugal deformation [1]. While hard X-ray experiments push towards atomic resolution, full 3D information on the particle shape (and orientation) from a single scattering pattern

requires access to the wide-angle scattering signal available only at longer wavelength [2]. We have used intense XUV pulses from the FERMI-FEL to retrieve the 3D shapes of single helium nanodroplets. We follow the evolution from axisymmetric oblate over triaxial prolate to two-lobed droplets with increasing angular momentum, as predicted by the theoretical model of a classical spinning drop.

- [1] Gomez *et al.*, *Science* **345** (2014)
- [2] Barke *et al.*, *Nat. Comm.* **6** (2015)

MO 19.4 Fri 12:00 N 3

**Machine-learning assisted classification of diffraction images** — ●J. ZIMMERMANN<sup>1</sup>, M. SAUPPE<sup>1</sup>, A. ULMER<sup>1</sup>, B. LANGBEHN<sup>1</sup>, S. DOLD<sup>2</sup>, B. V. ISSENDORFF<sup>2</sup>, I. BARKE<sup>3</sup>, H. HARTMANN<sup>3</sup>, K. OLDENBURG<sup>3</sup>, F. MARTINEZ<sup>3</sup>, K.H. MEIWE-BROER<sup>3</sup>, B. ERK<sup>4</sup>, C. BOMME<sup>4</sup>, B. MANSCHWETUS<sup>4</sup>, J. CORREA<sup>4</sup>, S. DÜSTERER<sup>4</sup>, R. TREUSCH<sup>4</sup>, T. MÖLLER<sup>1</sup>, and D. RUPP<sup>1</sup> — <sup>1</sup>IOAP, TU Berlin — <sup>2</sup>Univ. Freiburg — <sup>3</sup>Univ. Rostock — <sup>4</sup>FLASH@DESY

Short wavelength Free-Electron-Lasers (FEL) enable diffractive imaging of individual nanosized objects with a single x-ray laser shot. Due to the high repetition rate, large data sets with up to several million diffraction pattern are typically obtained in FEL particle diffraction experiments, representing a severe problem for the data analysis. We here propose a workflow scheme to drastically reduce the amount of work needed for the categorization of large data-sets of diffraction patterns, with the ultimate goal of developing an unsupervised training procedure. With a first supervised approach a classification and viewer tool is used for classifying manually selected high quality diffraction pattern. These patterns are then used as training data for a Residual Convolutional Neural Network (RCNN). The RCNN is designed for the classification of data for efficient indexing and subsequent analysis. The residual learning framework is a new type of network structure that drastically increases the depth of neural networks [He, et al. Deep Residual Learning, 2015]. First performance evaluations are done using data from a single-shot wide-angle scattering CDI experiment on silver clusters conducted in 2015 at the FLASH facility in Hamburg.

MO 19.5 Fri 12:15 N 3

**The X-Ray Movie Camera: Time-Resolved Diffractive Imaging Of Individual Clusters** — ●M. SAUPPE<sup>1</sup>, T. BISCHOFF<sup>1</sup>, K. KOLATZKI<sup>1</sup>, B. LANGBEHN<sup>1</sup>, M. MÜLLER<sup>1</sup>, B. SENFFTLIEBEN<sup>1</sup>, A. ULMER<sup>1</sup>, J. ZIMBALSKI<sup>1</sup>, J. ZIMMERMANN<sup>1</sup>, L. FLÜCKIGER<sup>2</sup>, T. GORKHOVER<sup>1,3</sup>, C. BOSTEDT<sup>4,5</sup>, C. BOMME<sup>6</sup>, S. DÜSTERER<sup>6</sup>, B. ERK<sup>6</sup>, M. KUHLMANN<sup>6</sup>, D. ROLLES<sup>6</sup>, D. ROMPOTIS<sup>6</sup>, R. TREUSCH<sup>6</sup>, T. FEIGL<sup>7</sup>, T. MÖLLER<sup>1</sup>, and D. RUPP<sup>1</sup> — <sup>1</sup>IOAP, Technische Universität Berlin, Germany — <sup>2</sup>ARC CoAMI, LaTrobe University, Australia — <sup>3</sup>LCLS, Stanford Linear Accelerator Center, USA — <sup>4</sup>Argonne National Laboratory, USA — <sup>5</sup>DoP, Northwestern University, USA — <sup>6</sup>FLASH, Deutsches Elektronen-Synchrotron — <sup>7</sup>optiX fab, Germany

Coherent diffractive imaging has been developed as a powerful technique for uncovering the structure of nano-sized particles like viruses, aerosols and clusters, as well as laser-induced nanoparticle dynamics. So far in time-resolved imaging experiments either optical pump lasers have been used or pump- and probe-images were superimposed. Here we present a new experimental setup, were the image of the initial particle and the image of final state are spatially separated and recorded by a two detector system. From the pump-image we can extract informations like size, shape and exposed intensity, from the probe-image we gain insight into light-induced dynamics. Probe pulses can be delayed up to 650 ps, realized by the new split-and-delay unit DESC, which has been permanently installed at the CAMP end-station at the FLASH FEL. First results will be discussed.

MO 19.6 Fri 12:30 N 3

**Determination of average cluster sizes by fluorescence: proof of principle on Ne, Ar, and Kr clusters** — ●XAVIER HOLZAPFEL<sup>1</sup>, ANDREAS HANS<sup>1</sup>, PHILIPP SCHMIDT<sup>1</sup>, LTAIEF BEN LTAIEF<sup>1</sup>, PHILIPP REISS<sup>1</sup>, REINHARD DÖRNER<sup>2</sup>, ARNO EHRSMANN<sup>1</sup>, and ANDRÉ KNIE<sup>1</sup> — <sup>1</sup>University of Kassel, Institute of Physics and Center of Interdisciplinary Nanostructure Science and Technology (CINSaT), D-34132 Kassel, Germany — <sup>2</sup>Institute of Nuclear Physics, J. W. Goethe University, D-60438 Frankfurt, Germany

Finite aggregates like clusters cover the range between molecular and

condensed matter physics and are used to study microscopic phenomena. The most important quantity for fundamental investigations of clusters is their average size. For clusters produced by a supersonic expansion, however, the average cluster size is usually estimated by an empirical law involving the stagnation pressure of the expansion, a law whose validity is under debate since its introduction. Here we present an alternative method for determination of the mean cluster size by resonant excitation of outer valence electrons and the subsequent emission of fluorescence photons in Ne, Ar and Kr. This method has been compared to average cluster size determination by the empirical law essentially corroborating the latter and can be used in the future for independent average cluster size determination.

MO 19.7 Fri 12:45 N 3

**Signatures of Rabi cycling and excited state population in single-shot coherent diffractive imaging** — ●BJÖRN THORBEN KRUSE, CHRISTIAN PELTZ, and THOMAS FENNEL — Institute of Physics, University of Rostock, Germany

Single-shot coherent diffractive imaging (CDI) of individual free

nanoparticles enables the study of their three-dimensional shape and orientation [1] as well as their optical and electronic properties [2]. Recently, even signatures of quantum-mechanical vortices have been observed in scattering images of superfluid helium droplets [3], demonstrating that CDI provides access to the observation of rotational excitation in quantum liquids. Whereas the imaging of these vortices is an indirect detection of quantum effects, it remains an open question to what extent quantum effects can be observed directly and how their signatures would look like. Here we study the possibility to directly image the nonlinear quantum-mechanical few-level dynamics in laser-excited nanoparticles. For our theoretical analysis, we employ a Maxwell-Bloch type description of the scattering problem, where the polarization dynamics is described in local few-level approximation and field propagation is treated with the finite-difference time-domain method (FDTD). The origin of non-linear effects in resonant XUV scattering from droplets and possible routes to the spatiotemporal imaging of population dynamics will be discussed.

[1] I. Barke et al., Nat. Comm. **6**, 6187 (2015)

[2] C. Bostedt et al., Phys. Rev. Lett. **108**, 093401 (2012)

[3] L. F. Gomez et al., Science **345**, 6199:906-909 (2014)