MO 9: Atomic Clusters II (joint session A/MO)

Time: Tuesday 14:00–16:00 Location: K 2.016

MO 9.1 Tue 14:00 K 2.016

X-ray coherent diffractive imaging of quantum vortices in single helium droplets — •Rico Mayro Tanyag[1], Charles Bernando[1], Curtis Jones[1], Luis Gomez[1], Andrey Vilesov[1], Camila Bacellar[2], James Cryan[2], Oliver Gessner[2], Ken Ferguson[3], Sebastian Schorb[3], Christoph Bostedt[3,4], Daniel Rolles[5], and Artem Rudenko[5] — [1]University of Southern California, Los Angeles, California USA — [2]Chemical Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, California USA — [3]SLAC National Accelerator Laboratory, Menlo Park, California USA — [4]Argonne National Laboratory, Lemont, Illinois USA — [5]Kansas State University, Manhattan, Kansas USA

Free, single, rotating superfluid 4He nanodroplets (diameter D = 200-2000 nm, temperature T = 0.4 K) containing a number of quantum vortices have been studied via ultrafast X-ray coherent diffraction imaging using a free electron laser. The droplets were doped with Xe atoms, which collect on the vortex cores and serve as a contrast agent. In order to obtain the instantaneous positions and shapes of the vortices from the diffraction images, a phase retrieval algorithm has been developed, which utilizes the droplet boundary as a physical support. The algorithm also uses the droplet's scattering phase as an input for the iterative phase reconstruction. The obtained reconstructions reveal a plethora of transient vortex configurations within the droplet. The details of the algorithm and the possible origin of the observed vortex configuration will be discussed.

MO 9.2 Tue 14:15 K 2.016

Imaging the equilibrium shapes of spinning superfluid quantum droplets — •B. Langbehn[1], K. Sander[2], Y. Ovcharenko[1,3], C. Peltz[3], A. Clark[4], M. Coreno[5], R. Cucini[5], P. Finetti[5], M. Di Fraia[5], L. Giannessi[5], C. Grazioli[5], D. Iablonskyi[6], A. C. LaForge[7], T. Nishiyama[8], V. Oliver Álvarez de Lara[4], P. Piseri[9], O. Plekan[5], K. Ueda[6], K. C. Prince[5], F. Stienkemeier[7], C. Callegari[5], T. Fennel[3,10], D. Rupp[1,10], and T. Möller[1] — [1]TU Berlin — [2]Univ. Rostock — [3]European XFEL — [4]EPFL, Lausanne — [5]Elettra-Sincrotrone Trieste — [6]Tohoku Univ. Sendai — [7]Univ. Freiburg — [8]Kyoto Univ. — [9]Univ. di Milano — [10]MBI, Berlin

With the intense short-wavelength femtosecond light pulses from free-electron lasers (FELs) it is now possible to study the structure of unsupported nanoparticles, including superfluid helium nanodroplets. When produced by a free-jet expansion from the liquid phase, these droplets can gain angular momentum. As superfluid droplets cannot rotate in the classical hydrodynamic sense, quantized vortices accommodating the angular momentum are formed inside the droplets. These alter the equilibrium shapes known for normal liquid droplets. In an experiment at the FERMI FEL, we recorded wide-angle scattering images of individual helium nanodroplets. From the diffraction patterns, we reconstructed the full three-dimensional droplet shapes, enabling a comparison to a theoretical model of rotating normal liquid droplets. Surprisingly, the observed shapes of the superfluid droplets match their classical counterparts.

MO 9.3 Tue 14:30 K 2.016

Two-color diffraction imaging of helium nanodroplets — •L. Hecht[1], B. Langbehn[1], Y. Ovcharenko[1,2], M. Sauppe[1], J. Zimmermann[1], B. Kruse[3], C. Peltz[3], K. Sander[3], A. Colombo[4], P. Piseri[4], A. D' Elia[5], M. di Fraia[6], L. Giannessi[6], O. Plekan[6], K. Prince[6,7], M. Zangrando[6], C. Callegari[6], T. Möller[1], T. Fennel[3,8], and D. Rupp[1,8] — [1]IOAP, TU Berlin — [2]XFEL@DESY — [3]Univ. Rostock — [4]Univ. Milano — [5]Univ. Trieste — [6]FERMI@Elettra — [7]IOM, Trieste — [8]MBI, Berlin

Extremely intense femtosecond pulses produced by short-wavelength free-electron lasers open up the possibility to image non-depositable nanostructures like superfluid helium nanodroplets in a single shot [Gomez *et al.* Science **345** (2014)] and to follow the transient formation [Bostedt *et al.* PRL **108** (2012)] and disintegration [Gorkhover *et al.* Nat. Phot. **10** (2016)] of laser-excited matter. At the FERMI facility a two-color XUV beam [Ferrari *et al.* Nat. Comm. **7** (2016)] can be used to perform time-resolved imaging with the goal to investigate ultrafast excitation and plasma dynamics.

Two diffraction images, each generated by one color, of the same He droplet can be separated through filter foils in front of the scattering detector. A pulsed cryogenic cluster source produces these at a size of several hundred nanometers. A combination of around 21 and 42 eV is scanned for a resonant scattering response, as the singly (*1s2p*) and doubly (*2p3p*) excited states of atomic He lie close to these energies, and thereby spatially resolve the excitation profile of the nanodroplets. The experimental setup and first results will be presented.

MO 9.4 Tue 14:45 K 2.016

Coherent diffraction images of isolated helium nanodroplets obtained with a high harmonic source — N. Monserud[1], B. Langbehn[2], P. Nuñez von Voigt[2], M. Sauppe[2], A. Spanier[2], J. Zimmermann[1,2], Y. Ovcharenko[2,3], T. Möller[2], F. Frassetto[4], L. Poletto[4], A. Trabattoni[5], F. Calegari[5,6], M. Nisoli[6,7], K. Sander[8], C. Peltz[8], T. Fennel[1,8], B. Schütte[1], M.J.J. Vrakking[1], A. Rouzée[1], and •D. Rupp[1,2] — [1]Max-Born-Institut Berlin — [2]TU Berlin — [3]European XFEL — [4]CNR Padova — [5]CFEL@DESY — [6]CNR Milano — [7]Politecnico di Milano — [8]Universität Rostock

We recently demonstrated single-shot coherent diffractive imaging of individual gas-phase nanoparticles with a table-top light source (Nat. Comm. 8, 493 (2017)). In the present work, superfluid helium nanodroplets were irradiated by intense extreme ultraviolet (XUV) pulses from high-order harmonic generation (HHG). The single-shot XUV spectra of the multicolor pulses were measured in coincidence. From the diffraction images, a majority of spherical droplets and a small fraction of rotationally distorted prolate shapes could be identified. Further, the spherical diffraction images were analyzed via multicolor Mie fits and the refractive indices at the harmonic wavelengths were extracted. They are a sensitive measure of changes in the electronic structure and thus can serve as a handle for tracking ultrafast excitation and ionization dynamics in the droplets in time-resolved imaging approaches - ultimately with intense attosecond pulses, that are currently under development at HHG sources and free-electron lasers.

MO 9.5 Tue 15:00 K 2.016

Machine-Learning assisted classification of diffraction images — •J. Zimmermann[1], M. Sauppe[2], B. Langbehn[2], Y. Ovcharenko[2,4], LDM Collaboration[3], T. Fennel[1,5], T. Möller[2], and D. Rupp[1] — [1]MBI Berlin — [2]IOAP, TU Berlin — [3]FERMI@Elettra — [4]XFEL, Hamburg — [5]Univ. Rostock

Short wavelength Free-Electron-Laser (FEL) enable diffractive imaging of individual nano-sized objects with a single x-ray laser shot. Due to the high repetition rates, large data sets with up to several million diffraction patterns are typically obtained in FEL particle diffraction experiments, representing a severe problem for data analysis. Assuming a dataset of N diffraction patterns with M x K pixels, a high dimensional space (N x M x K) has to be analyzed. Thus feature selection is crucial as it reduces the dimensionality. This is typically achieved via custom-made algorithms that do not generalize well, e.g. feature extraction methods applicable to spherically shaped patterns but not to arbitrary shapes. This work exploits the possibility to utilize a neural network (NN) as a feature extractor. A workflow scheme is proposed based on a Residual Convolutional NN, that drastically reduces the amount of work needed for the classification of large diffraction datasets, only a fraction of the data has to be manually classified. As a next step a generalized and fully unsupervised approach is envisioned (no manual classification needed) using an auto-encoder NN. First performance evaluations are done using data obtained from an experiment on helium nanodroplets conducted at the LDM endstation of the FERMI free-electron laser in Trieste.

MO 9.6 Tue 15:15 K 2.016

Holography combined with iterative phase retrieval: advances in coherent diffractive imaging of single nanoparticles — •Felix Zimmermann[1], Tais Gorkhover[2], Daniela Rupp[1,3], Thomas Möller[1], and Anatoli Ulmer[1] — [1]TU Berlin — [2]LCLS@SLAC — [3]MBI Berlin

Free-Electron-Lasers open the door to high-resolution images of noncristallin nanoparticles such as viruses via coherent diffraction of single X-ray pulses. The phase loss problem impedes the extraction of high resolution structural information from the recorded diffraction patterns. Two major approaches address this problem. First, one can recover the phase during post processing using iterative algorithms. Second, one can directly encode the phase into the image using reference scatterers as common in holography. The limitations and advantages of iterative and holographic methods differ significantly: iterative algorithms can reconstruct the sample from its diffraction patterns alone, but require a priori constraints and rely on human input. Usually, a number of independent reconstructions using different starting values have to be performed. The final result is based on the average of these reconstructions. In holography, a unique reconstruction without prior knowledge can be performed via simple calculations based on the Fourier transformation, but the result is degraded by the transfer function given by the reference. This talk will discuss whether a combination of both approaches might be advantageous regarding stability, computational complexity and achievable fidelity.

MO 9.7 Tue 15:30 K 2.016

Optical focusing of isolated particles for diffractive imaging experiments — •Salah Awel[1,4], Daniel Horke[1,4], Rick Kirian[2], Xiaoyan Sun[1], Andrei Rode[3], Jochen Küpper[1,4,5], and Henry Chapman[1,4,5] — [1]Center for Free-Electron Laser Science, DESY, Hamburg, Germany — [2]Arizona State University, Tempe, AZ, USA — [3]Laser Physics Centre, Australian National University, Canberra, Australia — [4]Center for Ultrafast Imaging, Universität Hamburg, Germany — [5]Department of Physics, Universität Hamburg, Germany

Single-particle imaging (SPI) is emerging as a new techniques at x-ray free-electron lasers (XFELs) that consists of directing a stream of randomly oriented bioparticles across the focus of the XFEL beam aiming to construct high-resolution 3D structure from diffraction patterns of multiple identical particles. Presently, the difficulty of efficiently delivering isolated bioparticles to the sub-micrometer x-ray focus is the limiting factor in the development of SPI. In order to mitigate this problem, we have developed a technique for guiding aerosolized nanoparticles to the x-ray focus using spatially shaped optical laser beam [1]. Our current experiments aim at transversely confining streams of aerosolized particles as they exit an aerosol injector with a counter-propagating “hollow-core” quasi-Bessel beam. Through radiation pressure and thermal (photophoretic) forces that arise from the interaction of the particle with the surrounding gas molecules, the particles confine within the low-intensity core of the laser beam [2].

[1] Eckerskorn et al., *Opt. Exp.* **21**, 30492-30499 (2013).
[2] Eckerskorn et al., *Phys. Rev. Applied* **4**, 064001 (2015).

MO 9.8 Tue 15:45 K 2.016

Signatures of Rabi cycling and excited state population dynamics in single-shot coherent diffractive imaging — •Björn Kruse[1], Christian Peltz[1], and Thomas Fennel[1,2] — [1]University of Rostock, Albert-Einstein-Straße 23, D-18059 Rostock, Germany — [2]Max-Born-Institute, Max-Born-Straße 2A, D-12489 Berlin, Germany

Only recently, coherent single-shot diffractive imaging (CDI) of individual free nanoparticles has been demonstrated with a laser-based source using high harmonic generation [1], promising new applications and unprecedented insights into the ultrafast dynamics induced or probed via the single-shot scattering process. So far, CDI experiments have been analyzed via an effective classical linear response description, e.g. to reconstruct the shape and orientation of nanoparticles [2]. For strong laser fields and in particular for resonant excitations, both the linear and the classical description may no longer be valid as population depletion and stimulated emission become important. To what extent such processes may influence CDI scattering images is currently largely unknown. In our theoretical analysis, we describe the quantum-mechanical few-level bound state dynamics using a density matrix formalism and incorporate this into a 3D Maxwell solver based on the finite-difference time-domain method (FDTD). We discuss how and to which extend the spatio-temporal population dynamics influences the scattering images and analyze the observed trends.

[1] D. Rupp et al., Nat. Commun. **8**, 493 (2017)
[2] I. Barke et al., Nat. Commun. **6**, 6187 (2015)