Wide-angle diffraction reveals 3D shapes of superfluid helium nanodroplets

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Synopsis
Recent pioneering experiments on superfluid helium nanodroplets revealed strong deformations of the particles' shape projections that were attributed to high angular momenta [1, 2]. We have used intense XUV pulses from the FERMI free-electron laser to record diffraction patterns of helium nanodroplets up to large scattering angles that allow for a unique determination of the droplets' 3D shape. We find axisymmetric oblate, triaxial prolate and even two-lobed droplets, that are in line with the observed shapes of non-superfluid spinning drops.

With the advent of free-electron lasers (FEL) delivering femtosecond short-wavelength pulses, coherent diffractive imaging methods have been developed to gain insight into the structure of unsupported nanoparticles such as viruses or clusters. While experiments using light pulses in the X-ray regime aim at atomic resolution [3], full 3D information on the particle shape and orientation from a single diffraction pattern requires access to wide-angle scattering signal, available at longer wavelengths [4].

In our experiment at the FERMI-FEL's LDM endstation [5], diffraction patterns of single helium nanodroplets were recorded using intense 100 fs light pulses at XUV photon energies ranging from 19 to 62 eV.

While the majority of the 45,000 bright scattering images exhibit centrosymmetric rings, thus indicating spherical droplet shapes, about 10% of the images show diffraction patterns of non-spherical particles.

In particular, a tilt of a deformed droplet out of the scattering plane produces features in the wide-angle diffraction pattern that break the point symmetry (cf. Fig. 1). In order to simulate these features, a multi slice Fourier transform (MSFT) algorithm was employed, similar to that described in Ref. [4]. By assuming simple model droplet shapes and matching the MSFT simulations to our data, the droplets' axes and volume could be retrieved. When compared to a numerical model of non-superfluid rotating drops [6] our data show unexpectedly good agreement. This finding raises questions on the role and implications of superfluidity and vortices in helium nanodroplets with high angular momenta.

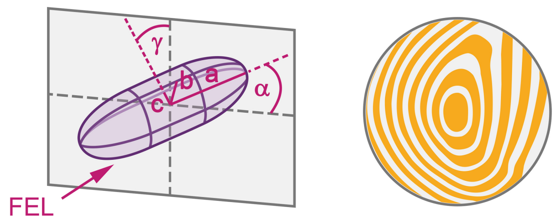

Figure 1. Tilting a deformed droplet out of the scattering plane (left) produces features in the diffraction pattern (right) that break the point symmetry.

References

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