Imaging the three-dimensional shapes and light induced dynamics of rotating helium nanodroplets

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Synopsis Intense short-wavelength femtosecond light pulses delivered by free-electron laser (FEL) facilities allow to investigate the structure and dynamics of nanometer-sized objects via coherent diffractive imaging (CDI). We studied single helium nanodroplets using extreme ultraviolet (XUV) femtosecond light pulses delivered by the FERMI-FEL and could uniquely determine the three-dimensional shapes of the droplets. Further, when irradiated by a strong optical laser pulse before being imaged with the XUV pulse, ultrafast dynamics inside the droplets can be observed in the diffraction patterns, thus enabling the investigation of plasma formation on a scale of nanometers and femtoseconds.

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 ultimately aim at atomic resolution [1], full three-dimensional information on the particle shape and orientation from a single diffraction pattern requires access to wide-angle scattering signal, especially available at longer wavelengths [2].

In our experiment, XUV diffraction patterns of single helium nanodroplets were recorded at the FERMI FEL's LDM endstation [3]. While the majority of the bright scattering images exhibit rings in concentric circles, thus indicating spherical droplet shapes [cf. Fig. 1(a)], 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 non-centrosymmetric features in the wide-angle diffraction pattern [cf. Fig. 1(b),(c)] that allow a unique determination of its three-dimensional shape. In a second set of measurements the droplets were doped with xenon atoms and a plasma was ignited with a high power infrared (IR) laser pulse. The nanoplasma propagation and destruction of the droplets was traced via single-particle imaging from femtoseconds up to hundreds of picoseconds after IR excitation [cf. Fig. 1(d)-(f)]. The reduced symmetry of the exploding particles complicates the interpretation of the diffraction patterns, demonstrating the need for 3D reconstruction algorithms.

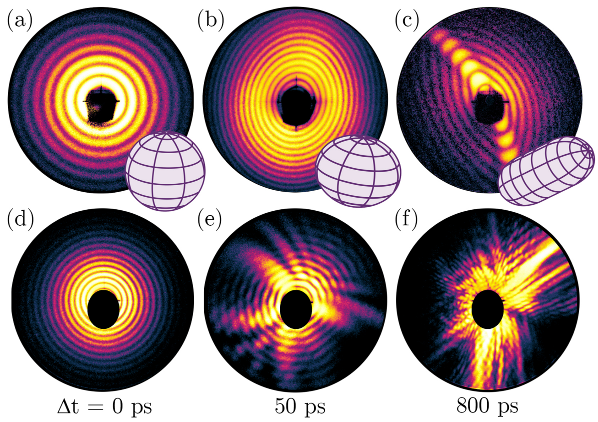

Figure 1. Top: Wide-angle scattering images of helium nanodroplets and corresponding model droplet shapes. Bottom: Droplet disintegration after IR excitation.

References

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[2] I. Barke *et al.* 2015 *Nat. Comm.* **6** 6187

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