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Coherent Imaging at FLASH

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Abstract. We have carried out high-resolution single-pulse coherent diffractive imaging at the FLASH free-electron laser. The intense focused FEL pulse gives a high-resolution low-noise coherent diffraction pattern of an object before that object turns into a plasma and explodes. In particular we are developing imaging of biological specimens beyond conventional radiation damage resolution limits, developing imaging of ultrafast processes, and testing methods to characterize and perform single-particle imaging.

1. Imaging at the FLASH Free-Electron Laser
The FLASH FEL, located at the Deutsches Elektronen-Synchrotron (DESY) laboratory in Hamburg, generates short, high-power soft-X-ray pulses by the principle of self-amplification of spontaneous emission [1]. This facility currently operates at fundamental wavelengths between 6 nm and above 50 nm, with pulse energies above 50 μJ and pulse durations less than 30 fs. Such short pulses permit the high-resolution and ultrafast imaging of a sample before the pulse destroys it [2].

In our coherent diffractive imaging experiments, we focus the FEL pulse to a 25-μm diameter spot to illuminate an isolated sample, which scatters the light to give a coherent and continuous diffraction pattern. We record this forward-directed diffraction pattern on a bare CCD after reflection from a multilayer-coated plane mirror angled by 45° to the incident beam [3]. The optical path length from the sample to the CCD is 53 mm. For a pixel size of 20 μm and a wavelength of 7 nm, for example, this camera length allows us to adequately sample (above the Nyquist rate) the intensity fringe pattern due to interference of waves scattered from two object points separated by 9.2 μm. These conditions
allow the diffraction pattern to be phased and then inverted to give a high-resolution image of the sample. There are advantages to this mode of microscopy when using FEL pulses: the retrieved image is complex-valued, giving image contrast both by absorption and phase-shifting or refractive properties of the sample; the diffraction pattern is insensitive to the transverse sample position; and “focusing” of the image is performed as part of the computational reconstruction process. These properties make diffractive imaging ideal for studying objects that are injected into vacuum on the fly, and for high-resolution single-shot imaging where the sample does not survive the beam.

2. High-Resolution Imaging

Our apparatus can be used to position fixed samples on silicon nitride window arrays into the beam, or to shoot particles across the beam. Examples are shown in Fig. 1. The test object was etched with a focused ion beam (FIB) after a few latex spheres of 145 nm were deposited on the silicon nitride membrane. Both the FIB object and the two latex spheres were reconstructed to a resolution of 40 nm. We also performed the first ever reconstruction of a single cell using a single FEL pulse. The single cell organism is *Spiroplasma melliferum*, an elongated helical bacteria, and the image was reconstructed to 70 nm resolution. We additionally developed nanofabricated coded apertures to increase the efficiency for single shot ultrafast holographic imaging of weakly scattering objects [4].

We successfully developed and demonstrated a particle injection system and diagnostics [5]. The injector operates as a continuously refreshed beam of free nanoparticles delivered through a differentially pumped aerodynamic lens, in a “shotgun” mode (unsynchronized to the FLASH pulses). We operated with pulse trains of 10 μs spacing. The particles, traveling faster than 100 m/s, were only hit by a single pulse. A summary of some results is shown in Fig. 2. During one run we operated the injector continuously for 18.7 hours, performing 26 sample changes with 14 different samples (Fig. 2 C). The CCD collected images continuously and 1873 of 16639 patterns contained particle scattering information, giving an average hit rate of 0.05 Hz (including all sample change down time). Samples included size-selected latex spheres, Ag cubes, DNA nanocomplexes, picoplankton, and lipid micelles. We have reconstructed images of injected whole hydrated picoplankton cells and latex dimers. Our results confirm the feasibility of single-particle diffractive imaging.

3. Ultrafast Imaging

The ultrafast duration of FLASH pulses allows the study of processes with a temporal resolution of tens of femtoseconds. The soft-X-ray FEL source gives significantly higher spatial resolution than achievable with optical lasers and a higher penetration and larger signal to noise than short-pulse electron microscopy. These capabilities open up the detailed study of non-repetitive processes, such as crack formation, phase separation and nucleation, plasma formation, and spallation and ablation. We demonstrated ultrafast coherent X-ray diffraction of ablating foils pumped with a visible laser synchronized to the FEL pulses.

We have phased time-resolved coherent diffraction patterns of laser-ablating microstructures [6]. Many identical structures were manufactured by focused ion-beam milling. We see that, from the
reconstructed images, and correlations of patterns at various time delays to the pattern of the
undisturbed object, order in the structure is progressively lost starting from short length scales. This
structural rearrangement progresses at close to the sound speed in the material [6]. Our initial
experiments show the ability to capture single-shot images of nanoscale dynamics at timescales of
individual atomic motion

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