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Nanoscale imaging with table-top coherent extreme ultraviolet source based on high harmonic generation

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ABSTRACT

A table-top coherent diffractive imaging experiment on a sample with biological-like characteristics using a focused narrow-bandwidth high harmonic source around 30 nm is performed. An approach involving a beam stop and a new reconstruction algorithm to enhance the quality of reconstructed the image is described.

1. Introduction

Microscopy is a critical enabling technology for visualizing objects with high resolution imaging down to the nanometer scale in order to study dynamic processes in material and biological systems. Conventional visible light microscopy can image living cells with a resolution as high as 200 nm [1]. However, its resolution is typically limited to λ/2NA, where λ is the wavelength of the light source and NA is the numerical aperture. To significantly improve resolution, scanning electron microscopy (SEM) and transmission electron microscopy (TEM) have been widely used and atomic resolution has been demonstrated [2]. Unfortunately, electron microscopes are limited by the mean free path of the charged particles, and therefore this technique is restricted to imaging thin samples, typically < 500 nm. For thickness as larger than 500 nm, because of inelastic scattering, this technique produces a blurred image and low resolution. Therefore, new techniques for high resolution imaging of thick samples are of great interest. Coherent x-ray diffractive imaging (CDI) using short wavelength light in the extreme ultraviolet or soft x-ray regions has emerged as a promising alternative approach to address the above problem [3–7]. Especially, CDI is a very useful method to investigate biological samples. Coherent diffractive imaging (CDI) is a powerful tool for imaging in which the optical lens used to reconstruct the sample's image is replaced by a computer-based reconstruction algorithm [3–7]. When an object is exposed to light from a coherent source, a diffraction pattern of the object is captured and based on diffraction and propagation theory the complex electric field of the light diffraction can be considered as a Fourier transform of the object. The object's image is then reconstructed by performing an inverse Fourier transform. Because only the intensity of the diffraction pattern is recorded a Fourier-based iterative phase-retrieval algorithm combined with an over-sampling method is used to recover the phase for image reconstruction process. This "lens-less" technique is aberration-free so that it is suitable for use at extreme UV and soft X-ray wavelengths and the theoretical spatial resolution is limited only by the radiation wavelength. In addition, because x-ray radiation can penetrate thicker samples, the CDI technique can overcome the limitations of an electron microscope and can be used as a promising approach for high resolution imaging of thick samples.

Besides the radiation from synchrotrons [4] and free electron x-ray lasers [5], high harmonic generation (HHG) sources which are generally produced by focusing a high intensity laser beam into a nonlinear medium [8–10] provide a new illuminating source for XUV and soft x-ray imaging, with their ultra-short pulses, excellent coherence properties and high degree of tunability [6,11–16]. Moreover, the generation of this source only requires a compact table-top setup, which enables small scale x-ray microscopy. The radiation of the harmonics can be explained by a three-step model [8–10], in which, free electrons produced by ionization by the laser field and then accelerated by the laser field recombine with their parent ions, releasing energy as single high energy photons. Basically, in order to meet the requirements of the image reconstruction algorithm, a monochromatic wave field CDI is conducted with a single harmonic order which can be selected using XUV focusing mirrors. In addition, since the single harmonic beam is focused into a tiny area (< 50 μm) comparable to the sample size the effective photon flux illuminating the sample strongly increases. Consequently, the acquisition time of a high-dynamic range diffraction image can be dramatically reduced. By

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using two concave spherical multilayer mirrors to collimate and then focus a single harmonic at 33.2 nm into the sample, M. Zurch et al. have reported real time XUV imaging at sub-70 nm spatial resolution in less than one second [16]. Recently, we have also conducted CDI microscopy using a focused harmonic beam around 30 nm generated from argon gas. We have shown that for samples in which the opaque area to the illuminating beam is much larger than the transmission area, high-angle diffractive patterns are easily captured and thus a good spatial resolution (up to ~ 45 nm) can be achieved [17,18].

In this paper, we demonstrate extreme ultraviolet CDI microscopy for a sample with biological-like characteristics which is fabricated by evaporating a solution of 2 µm-Carboxylated Polystyrene and 400 nm-Amino Polystyrene nano-particles on a 30 nm-thick Si_3N_4 membrane. The density of particles is very low, so the reduction in transmission is less than 30% which quickly leads to saturation of the detector. Therefore, a beam stop is implemented to block the high intensity region in the centre of the diffraction pattern to allow the high-angle data to be captured and then a new approach in processing diffraction patterns is implemented to increase the resolution of the reconstructed image.

2. Theory of coherent diffractive imaging

The diffracted field of a sample illuminated by a coherent plane wave at the detector plane can be approximated by [19,20]

$$U_i(x, y, z) = \frac{\exp(ikz)}{i\lambda z} \int\int_{-\infty}^{+\infty} U_0 \exp\left(\frac{ik}{2z}[(x_i - x_0)^2 + (y_i - y_0)^2]\right) dx_0 dy_0 \tag{1}$$

where U_O, x_O, y_O and U_i, x_i, y_i are the electric fields and coordinates in the sample plane and detector plane, respectively. z is the propagation coordinate ($z=0$ at the sample plane) and $k = 2\pi/\lambda$ is the wave vector of the incident wavelength λ. U_O is the electric field after propagating through the sample and this is also referred to as the "exit surface wave field" which contains information on the structure of the sample.

Expanding the exponential term in the integral and taking a Fourier transform operation, (1) can be rewritten as

$$U_i(x, y, z) = \frac{\exp(ikz)}{ik\lambda} \exp\left[\frac{ik}{2z}(x_i^2 + y_i^2)\right] F\left\{U_0 \exp\left[\frac{ik}{2z}(x_0^2 + y_0^2)\right]\right\} \tag{2}$$

If the distance z is much larger than the size of the sample ($z >> \frac{k}{2}(x_{0\,\max}^2 + y_{0\,\max}^2)$), then $\exp\left[\frac{ik}{2z}(x_0^2 + y_0^2)\right] \approx 1$, and (2) can be simplified to a far-field Fraunhofer approximation.

$$U_i(x, y, z) = \frac{\exp(ikz)}{ik\lambda} \exp\left[\frac{ik}{2z}(x_i^2 + y_i^2)\right] F\{U_0\} \tag{3}$$

An inverse Fourier transform is applied to reconstruct the sample image from the intensity distribution of the diffraction pattern and the phase information. However, in our experiment, only the intensity of the diffraction field given by the modulus of squared $U_i(x, y, z)$ is recorded by the CCD camera and all the phase information is lost. Therefore, the phase information must be recovered by using phase-retrieval algorithms to perform the reconstruction process. Generally, an iterative phase retrieval algorithm consists of the following four basic steps involving iterative Fourier transformation back and forth between the object and Fourier domains and application of the measured data or known constraints in each domain [21,22].

1) The sample's image is initialized with a random guess, or a support image, *g(u)*.
2) The magnitude and phase at the detector plane is calculated by taking a Fourier transform of the initialized sample image as described by *G(u)*.
3) The magnitude of *G(u)* is replaced by the Fourier modulus (square root of the diffraction intensity recorded by the detector). Then the new sample's image in real space is calculated by taking an inverse Fourier transform of the updated *G(u)*.
4) A support constraint is applied to the image from step 3 to obtain the updated sample's image, including the ER (Error Reduction) and HIO (Hybrid Input Output) algorithms.

The ER algorithm is based on the assumption that the object is of finite size and is isolated in empty space, i.e., the ER operation sets all the intensities outside the support to zero while it keeps the intensities which are inside. Although ER helps to clean up the noise around the object, the operation may stagnate at a local minimum and prevent further convergence. On the other hand, the HIO operation takes into account feedback of the previous reconstructed image to the current image and allows non-zero amplitudes outside the support. This algorithm helps to escape the local minima. Therefore, combining HIO and ER is an effective method in the phase retrieval process.

In order to obtain a high-quality reconstruction from the diffraction pattern of the illuminated sample experimental parameters consisting of the quality of the harmonic emission, the distance between the sample and the detector and the sample size need to meet the following requirements [22]. First, the illuminating beam must be coherent and have a reasonably narrow bandwidth ($\lambda/\Delta\lambda > 100$) and its spatial coherence length is about as large as the sample size times the linear oversampling factor. In addition, the harmonic photon flux density needs to be sufficiently high to capture diffractive patterns at high diffraction angles, but not too high as to damage the sample. Furthermore, to record a far-field diffraction pattern and thus satisfy the Fraunhofer approximation in illumination the sample is positioned far enough from the detector. In this case, the theoretical resolution of the CDI method can be estimated from $\delta=0.61\lambda/NA$, where NA is the effective "numerical aperture" of the scattered radiation.

3. Experimental results and discussion

3.1. Experiment

The high harmonic source is driven by a laser system consisting of a 1 kHz multi-pass chirped-pulse amplifier (Quantronix Odin-II HE) that is seeded by a Ti:Sapphire oscillator (Venteon). This laser system generates 30 fs pulses centred at 805 nm and the unapertured and unfocused beam has an average diameter of 12 mm and an energy of 2 mJ per pulse.

The laser beam is focused into a 150 mm-long argon gas cell by a lens of focal length 300 mm. which is mounted on a precision x/y/z translation stage to allow proper alignment with respect to the centre of the laser beam and to move the laser focus longitudinally with respect to the exit plane of the gas cell. The pressure in the gas cell is controlled by a stabilization system, which consists of a single valve pressure regulator (Alicat), a ceramic capacitance pressure gauge (Pfeiffer CMR 361), a gauge controller (Pfeiffer TPG 256 A MaxiGauge) and a LabView program on a computer. The gas cell has a glass window for entry of the laser pulses and a 0.1 mm pinhole at the exit to isolate the vacuum chamber from the argon gas-filled cell and to out-couple the harmonic emission. An ultra-thin aluminium filter which has high transmission in the wavelength range of 17–60 nm and is mounted on a filter wheel controlled by an electric motor is used to remove the fundamental beam. After that, the harmonic beam passes through a spatial filter set consisting of several pinholes from 200 µm to 2 mm to provide precise spatial filtering for the illuminating signal. This filter set is placed precisely along the beam path and is controlled by piezo linear motors. The harmonic beam is then focused into the sample by a pair of mirrors comprising a plane mirror (M1) and a 20 cm-radius focusing mirror (M2) (commercial Optix Fab multilayer Mo/Si mirrors with 2 nm bandwidth and 35% reflectivity at 30 nm for each mirror) which are installed at a reflection angle of ~10° in a Z-configuration to