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High contrast 3D imaging of surfaces near the wavelength limit using tabletop EUV ptychography

Bosheng Zhang*, Dennis F. Gardner, Matthew D. Seaberg, Elisabeth R. Shanblatt, Henry C. Kapteyn, Margaret M. Murnane, Daniel E. Adams

JILA, University of Colorado, 440 UCB, Boulder, CO 80309-0440, USA

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ABSTRACT

Scanning electron microscopy and atomic force microscopy are well-established techniques for imaging surfaces with nanometer resolution. Here we demonstrate a complementary and powerful approach based on tabletop extreme-ultraviolet ptychography that enables quantitative full field imaging with higher contrast than other techniques, and with compositional and topographical information. Using a high numerical aperture reflection-mode microscope illuminated by a tabletop 30 nm high harmonic source, we retrieve high quality, high contrast, full field images with 40 nm by 80 nm lateral resolution ($\approx 1.3\lambda$), with a total exposure time of less than 1 min. Finally, quantitative phase information enables surface profilometry with ultra-high, 6 Å axial resolution. In the future, this work will enable dynamic imaging of functioning nanosystems with unprecedented combined spatial (<10 nm) and temporal (<10 fs) resolution, in thick opaque samples, with elemental, chemical and magnetic sensitivity.

Microscopic imaging is critical for discovery and innovation in science and technology, accelerating advances in materials, bio, nano, and energy sciences, as well as nanoelectronics, data storage, and medicine. X-ray crystallography revolutionized many fields by determining crystalline structure on an atomic scale. Electron, X-ray, and scanning-probe microscopies can image complex matter with atomic-level spatial resolution. Super-resolved fluorescence microscopy can generate beautiful images of cellular organelles and structures. However, even these advanced imaging capabilities have limitations. Current imaging techniques are nowhere near their fundamental limits in terms of spatial and temporal resolution. Most techniques require extensive sample preparation, can damage the sample, are not applicable in situ, require invasive labeling, are chemically unspecific, or suffer from limited speed and field-of-view. Opaque, scattering, and disordered (non-crystalline) samples present a formidable challenge using any imaging modality.

Coherent diffractive imaging (CDI) is an important new full field imaging technique that can achieve very high spatial and temporal resolution simultaneously. In CDI, a spatially coherent beam illuminates an object, and the intensity of the scattered light is then collected on a pixel array detector. A generalized projection algorithm can then essentially replace any image-forming optics by solving for the complex-valued map of the sample that satisfies both the detector plane constraint (i.e. the magnitude of the retrieved scatter pattern must be consistent with the measured scattered pattern) and one or more a-priori sample plane constraints. The resulting image contains quantitative amplitude (material composition) and phase (thickness/height) contrast, providing more information about a surface or object than most traditional imaging techniques. Ptychography CDI has proven to be particularly robust when imaging objects containing complicated and/or large phase and amplitude variations [1–3]. This is because instead of collecting a single diffraction pattern as in traditional CDI, ptychography CDI acquires diffraction patterns from several adjacent overlapping positions. Although this requires that the sample be scanned, the resulting information redundancy from overlapping scatter patterns makes it possible to robustly and reliably solve the phase retrieval problem.

Coincident with the development of CDI, phase matching of the high harmonic generation (HHG) [4–9] process produces bright spatially coherent beams on a tabletop, spanning the entire VUV, EUV and soft X-ray regions of the spectrum, to photon energies >1.6 keV [9]. HHG takes advantage of the intense electric field created at the focus of a femtosecond laser that ionizes an atom and creates a nanoscale quantum antenna. Some laser driven electrons can acquire a large oscillation energy, which is then released in the form of an HHG photon after the electron recombines with the parent ion. HHG sources, when implemented in an optimal phase-matched geometry, can achieve full spatial and

* Corresponding author.
E-mail address: Bosheng.Zhang@colorado.edu (B. Zhang).

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temporal coherence, with attosecond to femtosecond pulse durations [10,11]. Recently, HHG sources have been successfully used to implement a variety of CDI techniques [12–18]. However, while EUV and X-ray CDI microscopy has been used for a wide range of applications, most experiments to date were performed in a transmission geometry. Although several attempts were made to apply CDI in reflection geometr [16,19–25] achieving high fidelity surface imaging comparable to scanning electron microscopy (SEM) [26–28] and atomic force microscopy (AFM) [29] in 3D has been challenging. In previous work [24], we demonstrated the first, general reflection mode result, working at an arbitrary angle of illumination and arbitrary angle of diffraction onto the detector however, in that work the numerical aperture was relatively low, (NA ~ 0.1) resulting in a transverse resolution of 150 nm.

Here we demonstrate high contrast, high quality, full field 3D imaging of surfaces by combining a tabletop HHG source at a wavelength of 30 nm with high NA (NA = 0.4) reflection mode ptychographic coherent diffraction imaging (CDI). We achieve record lateral spatial resolution of $\approx 1.3\lambda$ (40 nm), with 6 Å axial resolution. Our image quality compares very favorably with scanning electron microscopy (SEM) and atomic force microscopy (AFM), resulting in higher-contrast imaging with less sample damage. Moreover, the working distance is long at 3–10 cm. When combined with the femtosecond time resolution (≈ 10 fs) and shorter wavelengths (≈ 1–30 nm) readily available from HHG sources, [9] and in a pump-probe geometry to probe periodic phenomena, this work will make it possible to image the fastest charge, spin and phonon dynamics in functioning nanosystems in real space and time.

We note that compared to similar imaging techniques, EUV-based reflection CDI provides a powerful contrast mechanism by combining both material and geometric information. While zone plate based microscopes [30,31] can achieve ≈ 10–25 nm spatial resolution using shorter wavelength light, they result in amplitude-only images and thus contain less information about the object. Compared with AFM, where image contrast comes solely from height differences regardless of material properties, CDI derives its contrast from the reflected complex exit surface wave, which includes material dependent information. Finally, compared with SEM, which provides little information about height variation, the high phase contrast mechanism in reflection CDI inherently records even slight topographical features.

1. Experiment

To generate HHG beams, we focus ultrashort pulses from a Ti:sapphire laser amplifier system (central wavelength of 780 nm, pulse energy of 1.4 mJ, 5 kHz repetition rate and 22 fs pulse width) into a 200 μm diameter, 5 cm long, hollow waveguide filled with argon (36 Torr backing pressure). These experimental conditions are optimal for generating phase-matched, spatially coherent harmonics at wavelengths near 30 nm. After the waveguide, the CDI microscope is maintained at high vacuum ($\approx 10^{-6}$ Torr) to avoid absorption of the EUV light. We use a pair of silicon rejecter mirrors set near Brewster's angle for 780 nm, together with two 200 nm-thick aluminum filters, to completely eliminate the fundamental laser light. As shown in Fig. 1a, two 45° angle-of-incidence multilayer mirrors select the 27th harmonic (30 nm) which is then focused near the sample by an ellipsoidal nickel-coated mirror set at 5° grazing incidence. The beam then illuminates the sample at an angle of 50.5°, and has a slight negative phase curvature due to the position of the sample relative to the focus.

The sample consists of titanium shapes patterned with e-beam lithography onto a silicon substrate (SEM image shown in Fig. 1b). During the period after the sample was fabricated, surface contamination resulted in very fine height variations across the sample. To demonstrate excellent quantitative contrast of CDI, we measure the height of these variations and compare them to AFM and SEM for verification, as discussed below.

An EUV-sensitive CCD (Andor iKon-L, 2048 × 2048 pixel array, 13.5 × 13.5 μm^2 pixel size) is positioned 3.17 cm away from the sample and perpendicular to the specular reflection, resulting in a captured light NA of ≈ 0.4. Using a knife-edge scan, the beam size was measured at ≈ 12 μm in both *x*- and *y*-directions. Therefore, we chose a scanning step size of ≈ 3 μm in both *x*- and *y*-directions to ensure overlapping sampled areas, on an 18 × 11

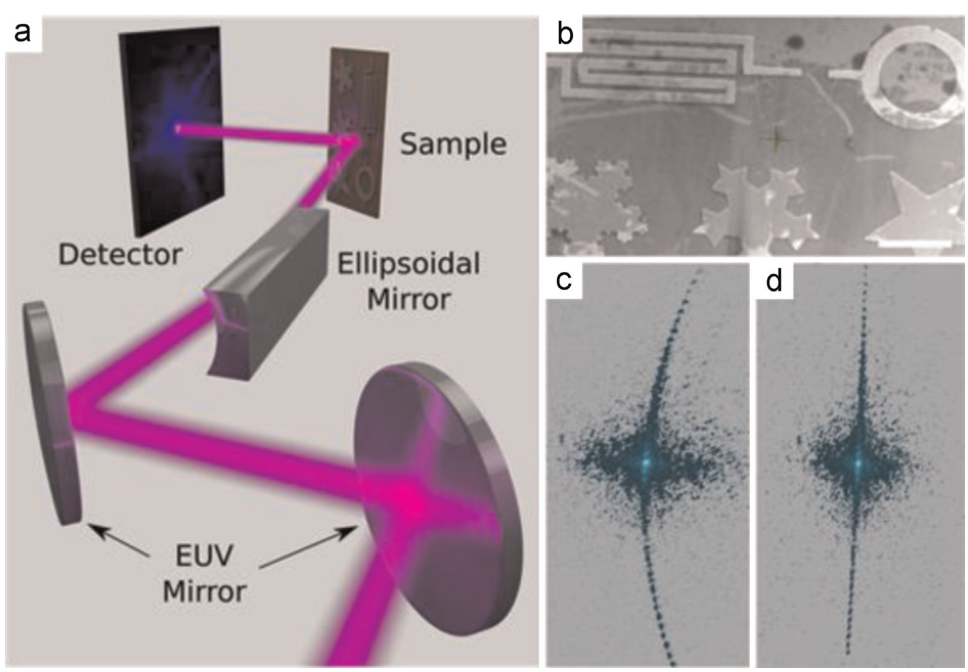

Fig. 1. Tabletop EUV ptychography. (a) Schematic of the tabletop EUV microscope. (b) SEM of the sample with a scale bar is 10 um. (c) Representative diffraction pattern from the ptychographic scan. (d) Diffraction pattern from (c) after tilted plane correction.