Tabletop coherent diffractive microscopy with extreme ultraviolet light from high harmonic generation

Daisy A. Raymondson*[a], Richard L. Sandberg[a], William F. Schlotter[b], Kevin S. Raines[c], Chan La-o-Vorakiat[a], Ethan Townsend[a], Anne Sakdinawat[d], Ariel Paul[a], Jianwei Miao[c], Margaret M. Murnane[a], and Henry C. Kapteyn[a]

[a]Department of Physics and JILA, University of Colorado and NIST, 440 UCB, Boulder, CO, USA 80309; [b]Institute for Experimental Physics, University of Hamburg, Luruper Chausee 149,22761 Hamburg, Germany; [c]Department of Physics and Astronomy and California NanoSystems Institute, University of California, Los Angeles, California, USA; Center for X-ray Optics, Lawrence Berkeley National Lab, University of California at Berkeley, Berkeley, California

ABSTRACT

We demonstrate lensless diffractive microscopy using a tabletop source of extreme ultraviolet (EUV) light from high harmonic generation at 29 nm and 13.5 nm. High harmonic generation has been shown to produce fully spatially coherent EUV light when the conversion process is well phase-matched in a hollow-core waveguide. We use this spatial coherence for two related diffractive imaging techniques which circumvent the need for lossy imaging optics in the EUV region of the spectrum. Holography with a reference beam gives sub-100 nm resolution in short exposure times with fast image retrieval. Application of the Guided Hybrid Input-Output phase retrieval algorithm refines the image resolution to 53 nm with 29 nm light. Initial images using the technologically important 13.5 nm wavelength give 92-nm resolution in a 10-minute exposure. Straightforward extensions of this work should also allow near-wavelength resolution with the 13.5 nm source. Diffractive imaging techniques provide eased alignment and focusing requirements as compared with zone plate or multilayer mirror imaging systems. The short-pulsed nature of the extreme ultraviolet source will allow pump-probe imaging of materials dynamics with time resolutions down to the pulse duration of the EUV.

Keywords: Coherent diffractive imaging, x-ray microscopy, ultrafast optics, high harmonic generation

1. INTRODUCTION

X-ray and extreme ultraviolet imaging offers several advantages over optical imaging, including higher resolution proportional to wavelength and the ability to access numerous chemical absorption edges in the soft x-ray and extreme ultraviolet (EUV) region of the spectrum. Soft x-ray microscopy on a synchrotron source has shown resolution down to 15 nm[1]. Two limitations to imaging in the EUV are the limited availability of high-brightness sources for applications, and the limited options for imaging optics. Since materials are highly absorbing in this region of the spectrum, no refractive lenses are available. Available optics include diffractive zone plate lenses and reflective multilayer mirror optics. Both are very lossy, 10% to 70% throughput per optical element, and both are prone to aberrations. Compact sources for EUV imaging are crucial to expanding access for applications such as biological microscopy and lithographic mask inspection. For compact sources such as high harmonic generation (HHG), coherent diffractive imaging without the use of imaging optics provides an attractive means of making use of the available flux.

1.1 Coherent diffractive “lensless” imaging

Coherent diffractive imaging of nonperiodic samples was first proposed by David Sayre in 1952[2] as an extension of x-ray crystallography. By sampling the Bragg peaks from a sample at higher than the Nyquist frequency for the desired spatial resolution, a condition known as oversampling, it is possible to capture the information necessary to uniquely

*daisy.raymondson@colorado.edu; phone 1 303 492-7764; fax 1 303 492-5235; http://jilawww.colorado.edu/kmgroup/

Metrology, Inspection, and Process Control for Microlithography XXIII, edited by John A. Allgair, Christopher J. Raymond
Proc. of SPIE Vol. 7272, 72720F · © 2009 SPIE · CCC code: 0277-786X/09/$18 · doi: 10.1117/12.814313

determine the amplitude and phase of the illumination field for each pixel in object space, thereby reconstructing an image of the sample. Mathematically, oversampling in frequency space corresponds to sampling a larger region in object space. By surrounding the sample with a region of no scatterers, a constraint is provided in object space. More details on the phase retrieval algorithms were given in 1982 by Fienup[3]. An experimental implementation of lensless diffractive imaging with phase retrieval was not realized until 1999[4], when computing power had advanced sufficiently for reconstruction of images containing hundreds of pixels square.

X-ray coherent diffractive imaging requires full spatial coherence of the illumination, as well as monochromaticity defined by

$$\frac{\lambda}{\Delta\lambda} \geq \frac{OD}{r},$$

where λ is the illuminating wavelength, Δλ is the bandwidth, O is the linear oversampling ratio, D is the sample diameter, and r is the desired resolution. The bandwidth and spatial coherence requirements mean that synchrotron sources must be spatially filtered in order for coherent diffractive imaging to work. The linear oversampling ratio must be ≥2 in order to capture the information necessary to reconstruct both amplitude and phase. Finally, the CCD must be in the far field, defined as

$$d >> \frac{\pi D^2}{\lambda},$$

where d is the distance from sample to CCD.

The reconstruction is accomplished by an iterative process. The square root of the measured diffraction pattern is seeded with an initial random phase set. An inverse Fourier transform gives a first guess at the sample density distribution. The oversampling constraint is applied in object space, then a Fourier transform generates a new diffraction pattern with an updated phase set. Here an error is generated by comparing the new diffraction pattern with the measured data. The amplitudes of the updated diffraction pattern are normalized to the measured values, and then this process is repeated, typically for several thousand iterations, until the image converges.

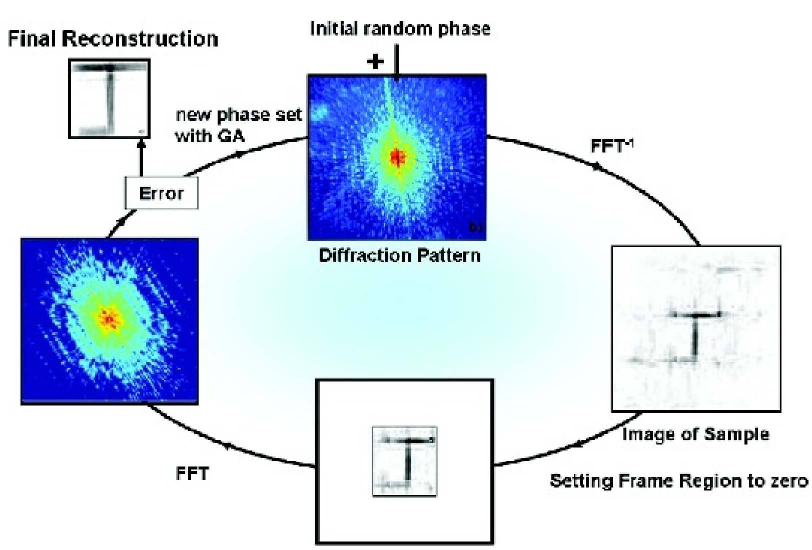

Fig. 1. Schematic of the phase retrieval process for lensless diffractive imaging. The 2D phase retrieval algorithm, known as Guided Hybrid Input-Output (GHIO), uses the oversampling condition as a constraint in object space and the amplitudes of the measured diffraction pattern as a constraint in the Fourier plane to iteratively converge on an image of the sample.

1.2 X-ray holography with a reference beam

A related imaging technique is holography with a reference beam, which is well established for optical light. X-ray holography is typically implemented with an integrated sample and reference aperture[5]. The resolution for this geometry scales as ~70% of the reference hole diameter. An advantage to holography is the quick image retrieval- since the relative phase information is encoded directly in the interference pattern, a simple Fourier transform retrieves the image, allowing for near-real-time imaging with moderate spatial resolution. Recently, Schlotter et al.[6] showed that multiple reference holes used with the same sample can be used to improve the image taken from a single exposure without increasing the dose to the sample. This technique is potentially valuable for imaging at short wavelengths where HHG flux is limited.

1.3 Lensless imaging with a tabletop source

Very recently, our group showed the first implementation of tabletop coherent diffractive imaging using a tabletop high harmonic generation source[9,10]. Resolutions down to 94 nm were shown using 29 nm illumination and resolution of 70 nm was obtained using 47 nm illumination. These results required imaging in a high-NA (> 0.6) geometry and applying a field curvature correction to the collected diffraction patterns, a first for extreme ultraviolet imaging.

2. EXTREME ULTRAVIOLET LIGHT FROM HIGH HARMONIC GENERATION

High harmonic generation (HHG) is an extreme nonlinear frequency conversion technique using ultrafast laser systems. It can be described semi-classically in a three-step process, shown schematically in Figure 1a, wherein (1) the strong electric field of a focused short-pulse laser beam ionizes an atom, (2) the electron is accelerated in the laser field, and (3) when the field reverses, the electron is accelerated back toward the atom and recombines, giving up its accumulated kinetic energy as a single high-energy photon. The high harmonic light is produced in a series of short bursts at each half-cycle of the laser pulse, which corresponds to a comb of narrow lines in frequency space. We have observed

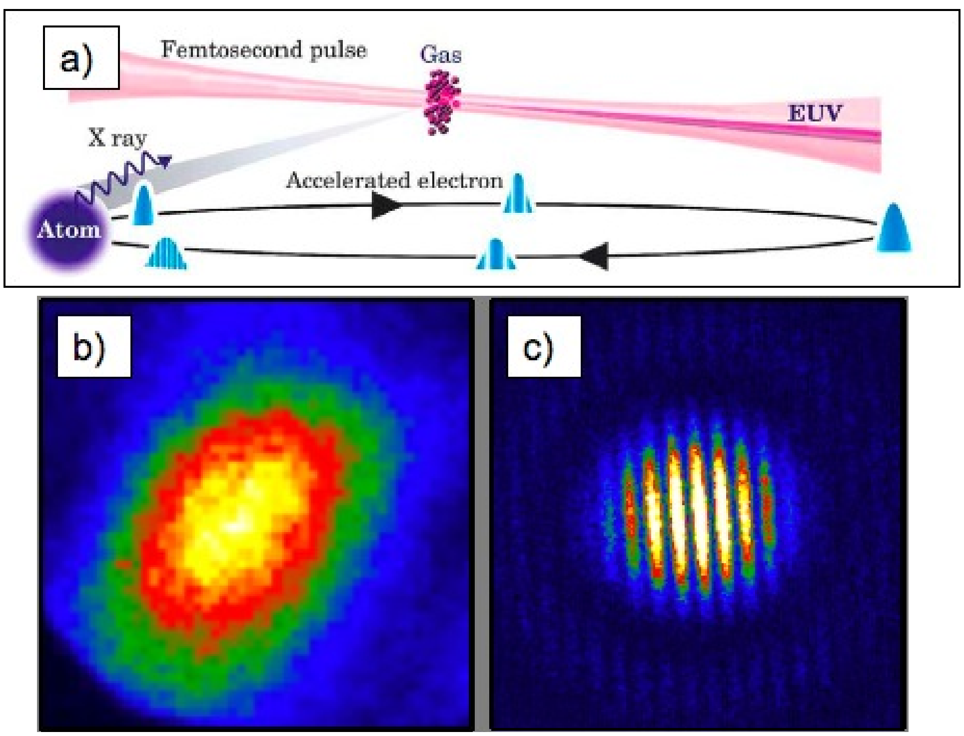

Fig. 2. a) Schematic of the high harmonic generation (HHG) process. b) Beam profile of the 13.5 nm beam as measured on the x-ray CCD camera. c) Young's double slit interference pattern with 13.5 nm HHG beam showing full spatial coherence.

linewidths of $\lambda/\Delta\lambda > 300$. It has been shown that HHG can be phase matched in a hollow core waveguide up to ~100eV photon energies. In practice, for a given gas pressure, several adjacent harmonics are produced simultaneously, near the phase matching peak. When the conversion process is phase matched, the EUV mode quality is excellent (see Figure 1b) and the beam has full spatial coherence[7,8] (Figure 1c). The mode quality allows for uniform illumination for imaging applications, and the spatial coherence means that phase-matched HHG is ideally suited for coherent imaging techniques. Finally, the EUV light retains the short-pulsed nature of the driving laser light, allowing for pump-probe experiments with time resolutions down to the ~10 fs pulse duration of the EUV.

3. EXPERIMENTAL SETUP

In both coherent imaging techniques, 1.4 mJ pulses of 800 nm light with ~20 fs pulse duration are produced in a Ti:sapphire oscillator/amplifier system (KM Labs Dragon) operating at 3 kHz repetition rate. The laser light is focused into a hollow-core waveguide filled with a noble gas for high harmonic generation. Thin metal filters separate the harmonics from the fundamental laser light. For 29 nm light, Ar is used as the nonlinear medium and a pair of 200 nm thick Al filters are used to filter out the laser light. For 13.5 nm, the He is used as the nonlinear medium and a pair of Zr filters are used. A pair of multilayer mirrors acts as a monochromator and condensor, selecting a single harmonic at the desired wavelength and focusing the beam onto a sample. The sample is mounted on a piezo-controlled x-y stage for alignment with the beam while under vacuum. Diffracted light from the sample is collected on an EUV-sensitive CCD camera mounted 2-5 cm from the sample.

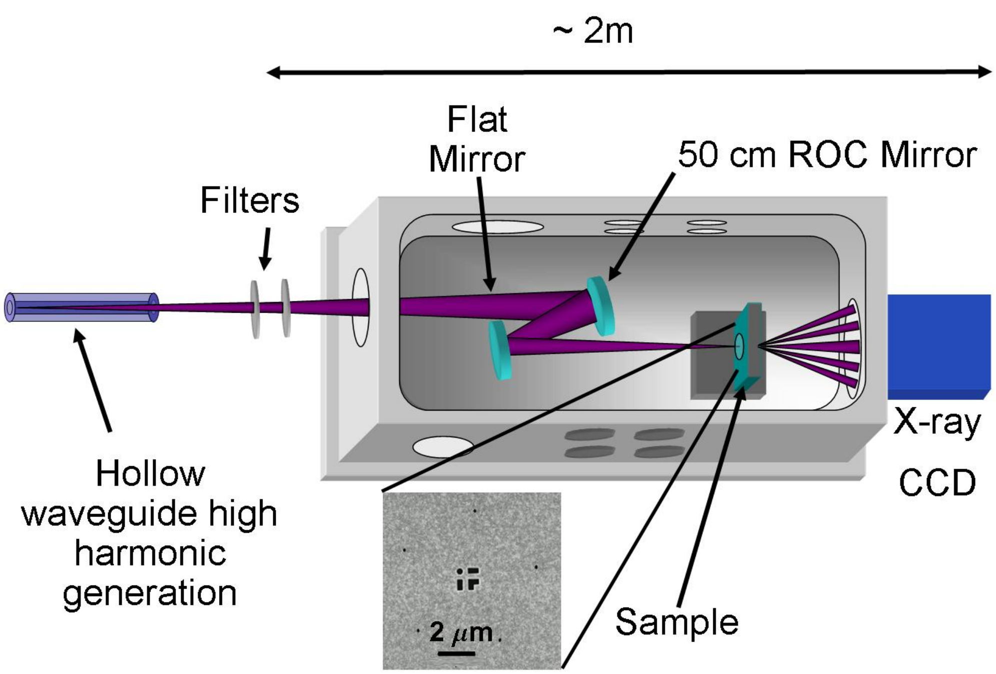

Fig. 3. Experimental setup for coherent diffractive microscopy with high harmonic generation. Extreme ultraviolet light is created through high harmonic generation in a hollow core waveguide. Thin metal filters block the residual fundamental laser light while allowing the EUV light to pass. A pair of multilayer mirrors acts as a monochromator to select out a single harmonic while focusing the beam onto the sample. A sample is mounted on a motorized x-y stage a few cm before an x-ray CCD, which captures the diffraction pattern from the sample.

4. RESULTS

4.1 Imaging with 29 nm light

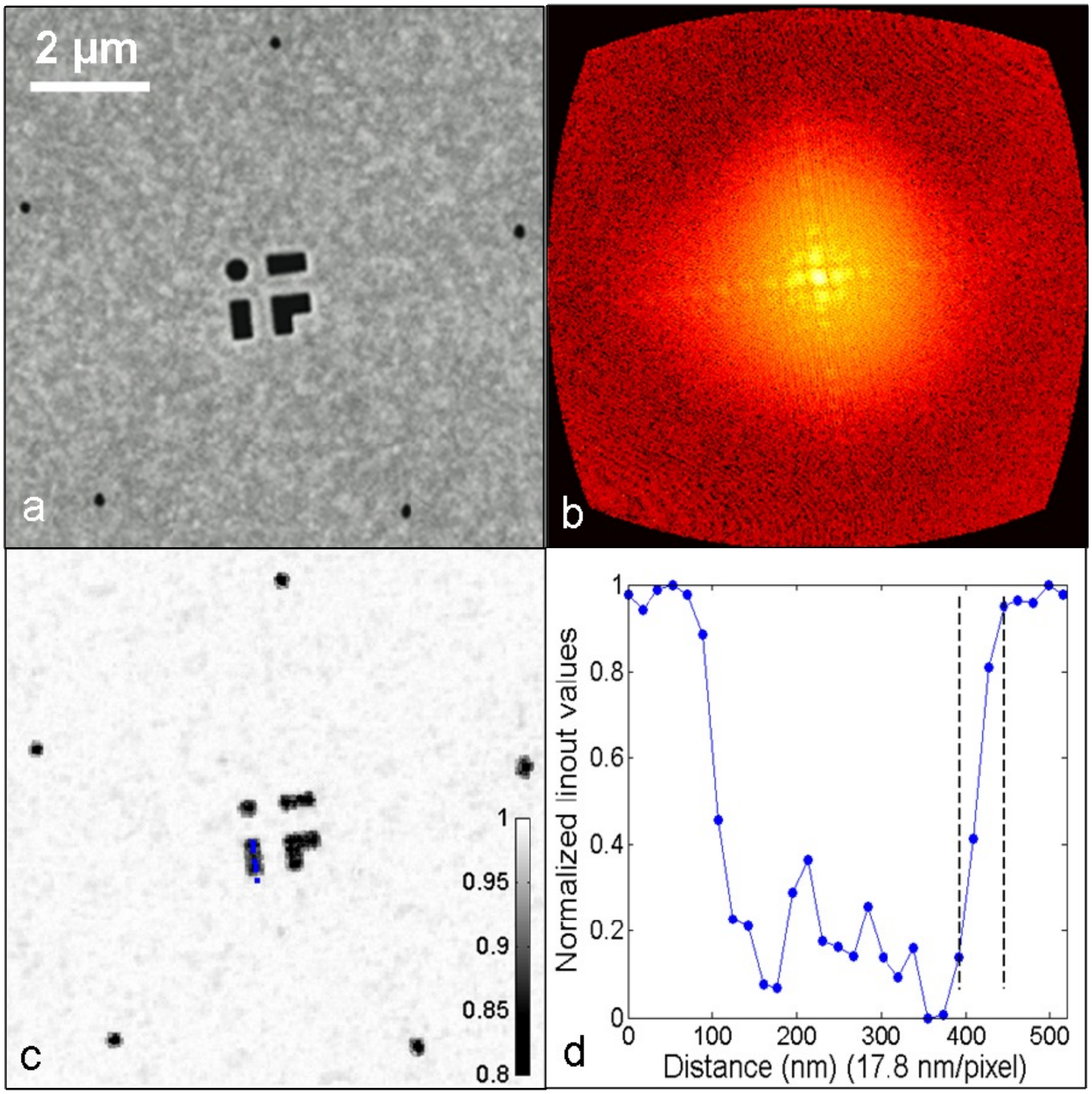

Fig. 4. a) Scanning electron microscope image of the five-reference-hole sample. b) Log of the curvature corrected hologram from 29 nm illumination, used in the iterative phase retrieval (maximum momentum transfer of q=±0.211 nm^{-1}). c) Reconstructed image of the sample using the GHIO phase retrieval algorithm to refine the resolution to 53 nm, as demonstrated in the lineout (blue dotted line) shown in d).

The sample used for imaging with 29 nm illumination is shown in Figure 4a. The substrate is 200nm of Si_3N_4 coated with 400 nm of Au. A test pattern inspired by the 1951 US Air Force test pattern was cut out using focused ion beam milling. The central test pattern was surrounded by 5 reference apertures of 130 nm diameter. The log scale diffraction pattern is shown in Figure 4b. Since this data was taken at NA > 0.65, a field curvature correction was applied to the pattern to map the detected diffraction on the flat CCD onto a curved surface. A simple Fourier transform of the background-subtracted diffraction pattern gave an image of the sample with 89 nm resolution, at the theoretical limit for the reference hole size. Recognizable images of the sample could be seen after exposures as short as 30s. The GHIO algorithm was applied to a longer exposure to make use of the detected light beyond the Airy disc of the reference and refine the resolution. The resulting image, with resolution 53 nm, is shown in Figure 4c, and the resolution lineout is shown in 4d.

4.2 Holography with 13.5 nm light

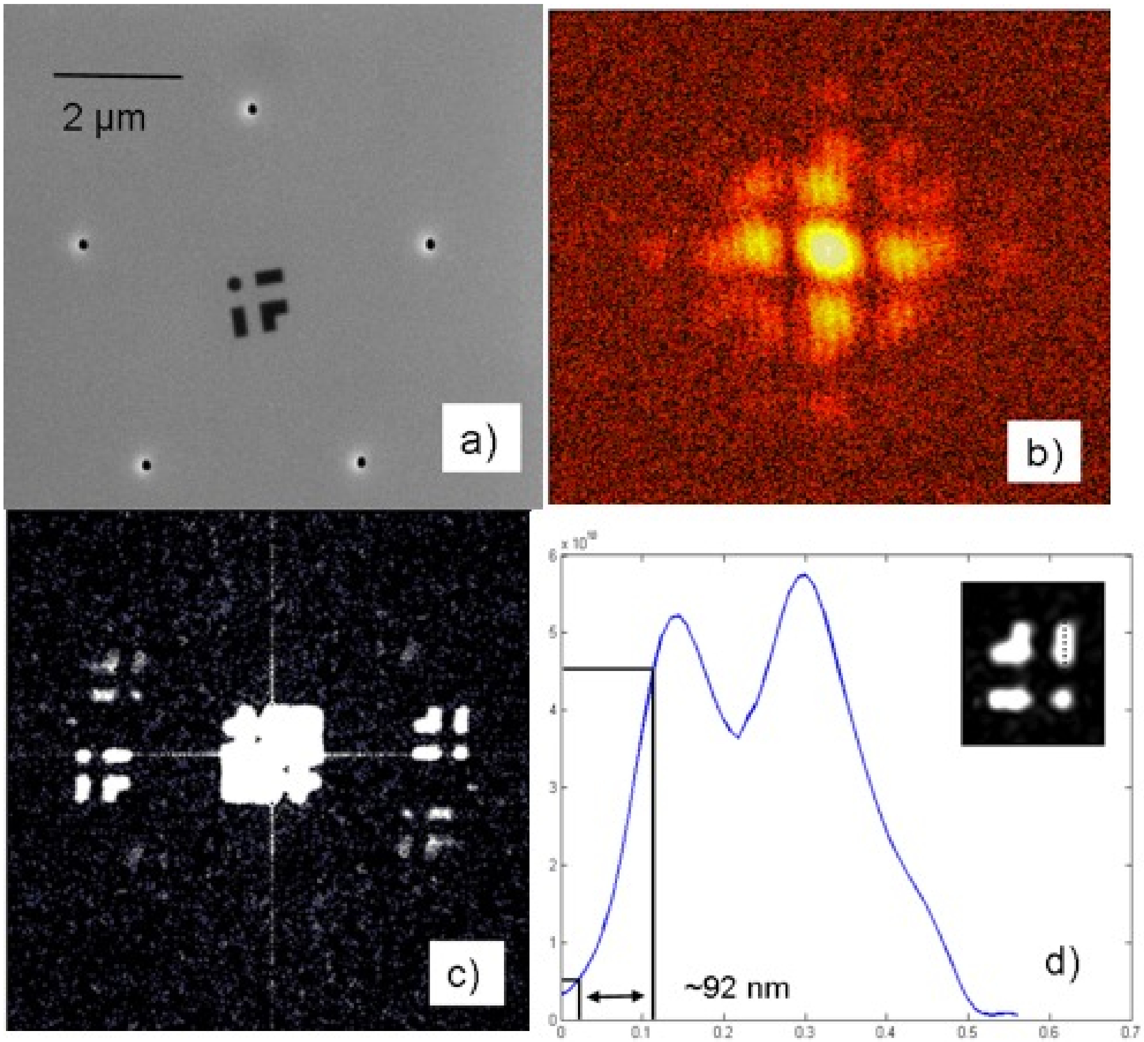

Figure 5. a) SEM image of FTH sample with five reference holes b) hologram after curvature correction (log scale), taken with the 13 nm HHG source, c) autocorrelation reconstruction of the sample, d) line cut through one sub-image showing 92 nm resolution

The same holography sample was used with the 13.5 nm source. The high harmonic flux at 13.5 nm is ~20x less than at 29 nm, in part because of re-absorption of the EUV in the gas used as the nonlinear medium. However, holography stil produced images of the sample after 10 min, with improved signal-to-noise with longer exposures up to 2 hours. The variation in intensity of the sub-images in the autocorrelation reconstruction is due to uneven illumination of the reference apertures.

4.3 Phase retrieval with 13.5 nm light

We have also obtained the first images using pure phase retrieval with a tabletop 13.5 nm source. Results are shown in Figure 4. The sample used was a “waving stick girl” figure milled out of a 200nm Si_3N_4 membrane coated with 400 nm of Au. With this experiment no reference beam was used, and the image was retrieved from the magnitude of the diffraction pattern alone. The diffraction pattern from 13.5 nm illumination and the resulting reconstruction, currently with >100 nm resolution, are shown. Pure phase retrieval with 13.5 nm HHG light is a crucial step toward scanning coherent imaging applications with high harmonics.

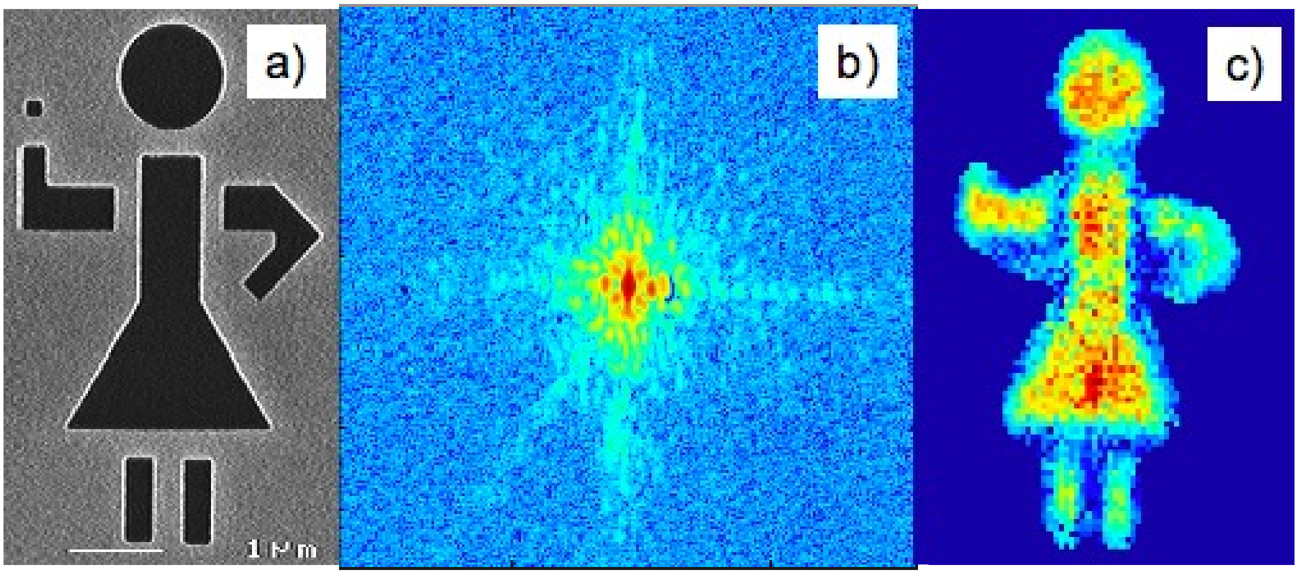

Fig. 6. a) Scanning electron microscope image of the waving stick girl sample. b) Log of the curvature corrected hologram from 13.5 nm illumination, used in the iterative phase retrieval. c) Reconstructed image of the sample using the GHIO phase retrieval algorithm.

4.4 Future directions

Several technical improvements in the microscope implementation and the laser sources should allow more efficient generation of the 13.5 nm light and higher throughput of that light through the optical system. These steps will bring down acquisition times as well as improving the image quality, making near-wavelength-limited resolution possible with the 13.5 nm source. With the addition of advanced phase matching techniques under development[11,12], scanning operation in reflection mode will be feasible.

ACKNOWLEDGMENTS

This work was supported by the NSF Engineering Research Center for Extreme Ultraviolet Technologies under NSF Award No. 0310717 and a grant from the DOE NNSA and the office of Basic Energy Sciences. The authors thank the JILA Instrument Shop and the Lehnert Labs for valuable assistance.

REFERENCES

[1] Chao, W., Harteneck, B. D., Liddle, J. A., Anderson, E. H., and Attwood, D. T., "Soft X-ray microscopy at a spatial resolution better than 15 nm," Nature **435**(7046), 1210-1213 (2005).

[2] Sayre, D., “The squaring method: a new method for phase determination,” Acta Cryst. **5**, 60-65 (1952).

[3] Fienup, J. R., "Phase retrieval algorithms: a comparison," Applied Optics **21**(15), 2758-2769 (1982).

[4] J. Miao, J., Charalambous, P., Kirz, J., and Sayre, D., "Extending the methodology of X-ray crystallography to allow imaging of micrometer-sized non-crystalline specimens," Nature **400**, 342 (1999).

[5] Eisebitt, S., Lüning, J., Schlotter, W. F., Lörgen, M., Hellwig, O., Eberhardt, W., and Stöhr, J., "Lensless imaging of magnetic nanostructures by X-ray spectro-holography," Nature(London) **432**(7019), 885-888 (2004).

[6] Schlotter, W. F., Rick, R., Chen, K., Scherz, A., Stohr, J., Luning, J., Eisebitt, S., Gunther, C., Eberhardt, W., Hellwig, O., and McNulty, I., "Multiple reference Fourier transform holography with soft x rays," Applied Physics Letters **89**(16), 163112-163113 (2006).

[7] Bartels, R. A., Paul, A., Green, H., Kapteyn, H.C., Murnane, M. M., Backus, S., Christov, I. P., Liu, Y., Attwood, D., and Jacobsen, C., "Generation of spatially coherent light at extreme ultraviolet wavelengths," Science **297**(5580), 376-378 (2002).

[8] Zhang, X., Libertun, A. R., Paul, A., Gagnon, E., Backus, S., Christov, I. P., Murnane, M. M., Kapteyn, H. C., Bartels, R. A., and Liu, Y., "Highly coherent light at 13 nm generated by use of quasi-phase-matched high-harmonic generation," Optics Letters **29**(12), 1357-1359 (2004).

[9] Sandberg, R. L., Paul, A., Raymondson, D. A., Hadrich, S., Gaudiosi, D. M., Holtsnider, J. M., Tobey, R. I., Cohen, O., Murnane, M. M., Kapteyn, H. C., Song, C., Miao, J., Liu, Y., and Salmassi, F., "Lensless diffractive imaging using tabletop coherent high-harmonic soft-X-ray beams," Phys Rev Lett **99**(9), 098103 (2007).

[10] Sandberg, R. L., Song, C. Wachulak, P., Raymondson, D., Paul, A., Amirbekian, B., Lee, E., Sakdinawat, A., La-o-vorakiat, C., Marconi, M., Menoni, C. Murnane, M. M., Rocca, J. J., Kapteyn, H. C., and Miao, J., “High numerical aperture tabletop soft x-ray diffraction microscopy with 70-nm resolution,” Proceedings of the National Academy of Sciences, **105** 1, 24-27 (2008).

[11] Lytle, A. L., Zhang, X., Sandberg, R. L., Cohen, O., Kapteyn, H. C., and Murnane, M. M., "Quasi-phase matching and characterization of high-order harmonic generation in hollow waveguides using counterpropagating light," Optics Express **16**(9), 6544-6566 (2008).

[12] Popmintchev, T., Chen, M.-C., Cohen, O., Grisham, M. E., Rocca, J. J., Murnane, M. M., and Kapteyn, H. C., “Extended phase matching of high harmonics driven by mid-infrared light,” Optics Letters **33**, 18, 2128-2130 (2008).