2

Conf 890502--14

BNL--43374

DE90 002291

SOFT X-RAY MICROSCOPE USING FOURIER TRANSFORM HOLOGRAPHY

I. McNulty, J. Kirz and C. Jacobsen
Physics Department, State University of New York,
Stony Brook, NY 11794-3800, USA

E. Anderson and M.R. Howells
Lawrence Berkeley Laboratory,
Berkeley, CA 94720, USA

H. Rarback
National Synchrotron Light Source, Brookhaven National Laboratory,
Upton, NY 11973, USA

Abstract

A Fourier transform holographic microscope with an anticipated resolution of better than 100 nm has been built. Extensive testing of the apparatus has begun. Preliminary results include the recording of interference fringes using 3.6 nm x-rays. The microscope employs a charge-coupled device (CCD) detector array of 576 × 384 elements. The system is illuminated by soft x-rays from a high brightness undulator. The reference point source is formed by a Fresnel zone plate with a finest outer zone width of 50 nm. Sufficient temporal coherence for hologram formation is obtained by a spherical grating monochromator. The x-ray hologram intensities at the recording plane are to be collected, digitized and reconstructed by computer. Data acquisition is under CAMAC control, while image display and off-line processing takes place on a VAX graphics workstation. Computational models of Fourier transform hologram synthesis, and reconstruction in the presence of noise, have demonstrated the feasibility of numerical methods in two dimensions, and that three-dimensional information is potentially recoverable.

1. Introduction

Early work in x-ray holography [1,2,3] has been limited by detector resolution, sensitivity, and the long exposure times necessary to obtain holograms of good contrast. Only recently has the field benefitted from the bright sources, high resolution optics and photoresists now available [4,5,6]. The advent of soft x-ray undulator sources at synchrotron facilities, and x-ray lasers, have made feasible a number of holographic imaging techniques hitherto difficult or impossible. This paper describes an experimental x-ray microscope based on Fourier transform holography (FTH), mounted on the long wavelength branch of the X1A soft x-ray undulator beamline, at the National Synchrotron Light Source. It uses a spherical grating monochromator to coherently illuminate a Fresnel zone plate, which forms a reference point source with an intensity exceeding 10^6 photons/second into a 60 nm focal spot. The detector is composed of a down-converting phosphor screen and cooled charge-coupled device (CCD). Using the FTH microscope, we expect to record holograms of good contrast with exposure times on the order of minutes.

Fourier transform holography, first proposed by Stroke and Falconer, and later by Stroke [7] in the "lensless" geometry used here, has been suggested as a practical method capable of yielding high resolution images that are relatively independent of the recording medium grain size [8]. Previous x-ray FTH experiments involved the use of photographic film as a detector, and thus introduced considerable noise in the process of recording the hologram. In addition, visible light reconstructions of the x-ray holograms suffered geometrical aberrations. In order to avoid these problems, we have chosen to record holograms with a CCD, and to reconstruct them numerically. Advances in electronic image processing and display technology also encourage a move in this direction. CCDs are appealing detectors for FTH, because their spatially quantized pixels naturally lend themselves to digital signal acquisition and processing. The CCD's linear response and wide dynamic range are similarly desirable for computational holography.

We are testing the CCD based FTH microscope at the X1A beamline, following a period of design and fabrication of the experimental apparatus. Software has been developed to direct the exposure and readout of the CCD camera, and to store the images on disk. Reconstruction software has been tested on numerical models. An analysis of the performance of the CCD camera is in progress, and preliminary results that demonstrate the recording of soft x-ray diffraction and interference fringes are promising.

2. Optical Design

In holography, interference with a reference beam is used to record both the phase and amplitude of light scattered by an object. Fourier transform holography differs from other holographic processes in that spherical waves are used for the phase reference. The optics for the FTH microscope are shown schematically in figure 1. The reference source and specimen illumination are derived from the same beam.

The reference is formed by a gold zone plate supported on a Si_3N_4 membrane. Zone plates with a finest outer zone width of 50 nm, have been fabricated for the Fourier transform holography project in a collaboration between LBL's Center for X-ray Optics and the Nanostructure Fabrication group at the IBM T.J. Watson Research Laboratory [9]. Ultra high resolution electron beam lithography and electroplating are the key techniques used. The zone plate produces a diffraction-limited focus when illuminated with a monochromatic x-ray beam ($\lambda/\Delta\lambda > 300$), that has a spatial coherence length comparable to the zone plate radius. The undiffracted zero-order beam passing through the open areas of the zone plate illuminates the object, which is placed in the first-order focal plane approximately a specimen-radius away from the focal point. Interference between the reference point source and waves scattered by the object result in an intensity pattern in space which may be measured by a CCD detector placed downstream.

The optimum working distance z between the focal and recording planes is determined by one's choice of spatial sampling frequency, and is ultimately set by the detector photoelelement pitch and acceptance aperture. Given a unity sampling condition, detector elements of size $\Delta_{x'}$, and approximating the object resolution element size Δ_x by the transverse resolution δ_t of a zone plate,

$$z = \frac{1.22\, N\delta_r\Delta_{x'}}{\lambda} \qquad (1)$$

for a smallest outer zone of width δ_r and an $N \times N$ array of recording plane elements. By recording the hologram at a distance of 100 mm we expect to obtain a 15 μm by 15 μm object field of view when a 256 × 256 pixel reconstruction is performed. The greater resolution of the CCD imager will allow pixel averaging, and eventually, an expansion of the field of view.

3. The Camera

We have developed a low noise, high resolution CCD camera for Fourier transform holography. An aluminum vacuum vessel with standard electrical feedthrough and vacuum ports houses the camera. It is maintained at a pressure of less than 10^{-2} torr to minimize x-ray absorption and vapor condensation as well as to insulate thermally the cooled CCD. Internally mounted miniature stages provide for small translations of the detector. X-ray photons enter the camera through a narrow snout, adjustable in length, which is sealed by a 120 nm thick, Si_3N_4 window made optically opaque by metallization with 80 nm of aluminum. At a wavelength of 3.6 nm, x-ray absorption in the aluminized window is about 60%, and 0.2% in residual air molecules within the camera, over a maximum working distance of 200 mm. Adaptability of the camera to the Stony Brook/NSLS Scanning X-ray Transmission Microscope (STXM) has been incorporated into the design [10]. The camera is readily substituted for the optical microscope normally used for pre-alignment of the STXM, and the STXM x-ray detector may be withdrawn from the beam when exposing the CCD. While not essential to the experimental arrangement, this feature greatly simplifies the task of aligning a zone plate lens, and the CCD camera in the x-ray beam, and then manipulating a specimen into the focal plane by using the scanning hardware and software. Initial alignment of the camera with the optic axis is aided by locating the CCD output signal peak via oscilloscope, when reading out the CCD array at a slow-scan rate. The effective collection time between successive reads of the same pixel is long enough to make this realtime alignment method possible with the available x-ray flux. Once aligned, camera exposure times are controlled by a manual or CAMAC controlled mechanical shutter that operates in the beamline vacuum, upstream of the zone plate. The camera alignment stage, as for the STXM, is mounted on a granite optical bench supported pneumatically for vibration isolation.

The imaging device consists of a scientific grade, Thomson-CSF type TH7883 CCD, which has a 576 × 384 array of 23 μm square pixels organized in a full frame format with no memory zone. When externally shuttered it is well suited to variable integration time, low light level imaging. The device is cooled below ambient temperatures to decrease the thermal background. We have used a thermoelectric cooler, and are now constructing a liquid nitrogen cooled system to further reduce the dark current. Two fine gauge type-K thermocouple elements permit cooling efficiency measurements and monitoring of the CCD temperature. The small power dissipation of the CCD supplies a fixed heat input for rewarming when necessary. All electrical connections to the chip are made with 5 mil, low thermal conductivity, constantan wire to decrease parasitic heating. The cooling rate is kept to less than 3 ° C/minute to avoid irreparable damage to the CCD. A visible LED test illumination source is used to calibrate the camera output. The source is located inside the vacuum envelope and is remotely controlled by the data collection system. The CCD is slow-scanned at a 45 Khz rate with correlated double sampling (clamp-and-sample technique) of the video signal. At present the noise in the CCD readout circuit is approximately 50 e^-. After refinements to the signal preamplifier and sampling circuit we expect to reach a minimum read noise figure of 12 e^-. Data acquisition from the readout electronics is accomplished with a 12-bit, 256K-word transient digitizer operated under CAMAC control by a VAXstation 3200 computer. Following a short image area cleaning period to remove any residual charge, then integration of an image, readout and digitization of the complete CCD array takes about six seconds.

The CCD was made sensitive to soft x-rays by depositing a thin layer of P31 phosphor (ZnS:Cu) directly onto its front surface. The composite detector spatial resolution is limited by the CCD pixel size. With the 5 – 10 μm diameter phosphor grains in close proximity to the CCD defocusing and optical scattering in the screen is not a problem. A nearly single-crystal thick layer, exhibiting good areal coverage, was obtained using filtration and sedimentation in distilled water. The phosphor screen forms a radiation damage resistant interface between incident x-rays and the CCD active surface. On the other hand, the screen is reasonably transparent to visible light and has a peak emission wavelength at 520 nm, where the quantum efficiency of the CCD is 35%. The detective efficiency for soft x-rays would be reduced by allowing absorptive materials to cover the phosphor, so little or no binder is used.

4. Test Objects, Noise, and Numerical Reconstructions

A series of well-defined imaging test objects suited to x-ray Fourier transform holography experiments were fabricated. They will be used to characterize the performance of the holographic microscope. The objects were produced by the same method used to make the zone plates that form the reference source. These two-dimensional masks doubly serve as "shadow foils", i.e. order-selecting apertures, and as resolution test patterns, integrated together onto single 260 μm square, Si_3N_4 membranes on 6.4 mm square silicon substrates. A test object imaged with the STXM is shown in figure 2. The 120 nm thick membranes are plated with 100 nm of gold, except at the location of a 2 μm aperture for the reference point source and a 10 – 20 μm aperture nearby, which contains one of a variety of linear gratings, grids, zone plates or radial star patterns spanning a range in spatial periods from 60 nm to 2000 nm. The same computer procedures used to generate the test patterns can be implemented in our synthetic hologram formation and reconstruction software to model the actual experiment.

Several independent sources of noise may degrade the quality of a recorded hologram, and therefore the fidelity of the reconstructed image. Vibration of the apparatus during the course of a hologram exposure will decrease fringe visibility. Mechanical vibration of the camera is much smaller than the CCD pixel size. Measurements using the STXM sample stage interferometer (least count size is 31.6 nm) indicate that specimen vibrations are less than 50 nm in magnitude, and drift is under 100 nm/hour. Fixed pattern variations in the down-converting phosphor screen response, and CCD photosite response nonuniformities are largely normalizable by subtraction of the flat-field image. Inherent to the CCD readout electronics are quantifiable noise sources arising from kTC noise in the MOSFET output node capacitance on the CCD chip itself, noise in the output signal preamplifier, and clock and correlated sampler timing jitter. Considerable effort is required to reduce the latter sources to acceptable levels. Fortunately, these only contribute to the total noise each time a pixel is read, and are for the most part independent of the integration time and the quantity of charge collected in a photosite. The dark current, and therefore its shot noise contribution, is minimized by cooling the detector, since it is a steep function [11] of the temperature T:

$$S_{dc} = S_0 \, T^{3/2} \, e^{-\frac{W}{2kT}} \tag{2}$$

where $W \approx 1.27\,\mathrm{eV}$ and $S_0 = 2/h^3(2\pi mk)^{3/2} \approx 5 \times 10^{15}\,\mathrm{cm}^{-3}$. At a temperature of 150° K, CCD integration periods of the order of hours result in only a few dark charges per pixel. Lastly, the charge transfer efficiency (CTE) of the CCD may be adversely affected by low temperatures and signal levels, causing an observable decline in the output signal for the last pixels to be read out. The manufacturer of our CCDs claim a transfer inefficiency of at most 10^{-6} at 25 ° C; even so, the last pixel read will have only 80% of its integrated charge remaining after 221, 184 transfers. In this application we do not supply a "fat zero" (low level optical or electrical bias) to improve the CTE. Compared to earlier CCDs, the TH7883 devices used contain a wider image zone to output register transfer channel, and separate transfer gate clock. These modifications facilitate better charge transfer efficiency in low light conditions and preclude the need for such a bias charge.

Hologram reconstructions are to be done numerically on the same computer used to acquire the raw data. The reconstruction software has been tested on computer-generated holograms of model objects which are distributed in one or more planes transverse to the optic axis. The simulation results, which include effects of noise in the hologram, indicate that successful numerical reconstructions can be carried out. On our system a 256 × 256 pixel reconstruction takes about 15 cpu – seconds. The algorithm was developed from Fresnel diffraction theory and is based on standard discrete fast Fourier transforms [12]. Applying the Fresnel-Kirchhoff relation for a general diffractive field condition, we may write for the complex amplitudes in the recording plane,

$$\Psi(x,\, y) \;=\; \Psi_0 \, e^{-\frac{i\pi}{\lambda z}(x'^2+y'^2)} \, T(x',\, y') \quad , \tag{3}$$

where $T(x',\, y')$ is the Fourier transform of the product of the object transmittance $t(x,\, y)$ and the factor $e^{-\frac{i\pi}{\lambda z}(x^2+y^2)}$. The general case includes object points not in the plane of the reference source. Strictly defined, holograms formed without a lens by a coplanar point reference source and two-dimensional object are known as lensless Fourier transform holograms. When the magnitude squared of the complex amplitudes in the hologram is recorded, the spherical (quadratic in the transverse coordinates of the hologram) phase term cancels in equation (3). Reconstruction of a real, three-dimensional object requires treatment of the phase dependence on differences in curvature of the object and reference wavefronts. The phase factor will likewise be important when considering the longitudinal extent of a focused source. Reference sources departing from the ideal may also be considered if their spatial distribution is known. Reconstruction by Stroke's correlative source compensation technique [13] offers a potential way to improve on the resolution normally limited by the size of an extended reference source.

5. Results, and the Future

The initial design and construction of the major elements of the Fourier transform holography microscope is complete. The basic components of the apparatus have been characterized, and testing with soft x-rays has begun with resolution test objects. Recently, we recorded fringes with 3.6 nm x-rays using the microscope, formed by interference between a zone plate generated reference source, and the light through a 5 μm pinhole illuminated by the zero-order radiation from the zone plate. Images such as these (see figure 3) were typically obtained with thirty minute exposures. Parts of the x-ray diffraction patterns from test objects were also recorded with the camera at a wavelength of 3.6 nm. Further improvements to the CCD cooling and readout noise, camera entrance window transmissivity, and phosphor coating uniformity, will permit higher contrast fringes to be recorded. When holograms of sufficient contrast have been collected we will begin numerical reconstructions. After completion of the hardware and careful characterization of the entire system, we will turn our attention to imaging biological specimens, and to further development with generalized reference sources.

6. Acknowledgements

We gratefully acknowledge the help of Steve Lindaas and Fernando Camilo. This work was supported in part by the Air Force Office of Scientific Research, under Contract F-49620-87-K-0001, and the Department of Energy, under grant DE-FG02-89ER60858 and Contract DE-AC03-76SF00098. It was carried out at the National Synchrotron Light Source, which is supported by the Department of Energy, under Contract DE-AC02-76CH00016.

References

[1] S. Aoki, Y. Ichihara and S. Kikuta, *Japan J. Appl. Phys.* **11** (1972) 1857

[2] B. Reuter and H. Mahr, *J. Phys. E* **9** (1976) 746

[3] M.R. Howells, M.A. Iarocci and J. Kirz, *J. Opt. Soc. Am. A* **3** (1986) 2171

[4] M. Howells, C. Jacobsen, J. Kirz, R. Feder, K. McQuaid, and S. Rothman, in: *Science* **238** (1987) 514; and C. Jacobsen, M. Howells, J. Kirz, K. McQuaid and S. Rothman, in: **OSA Proceedings on Short Wavelength Coherent Radiation: Generation and Applications**, eds., R.W. Falcone and J. Kirz (Opt. Soc. Am., Wash. D.C., 1988), Vol. 2, p. 303

[5] J.E. Trebes, S.B. Brown, E.M. Campbell, D.L. Matthews, D.G. Nilson, G.F. Stone, and D.A. Whelan, *Science* **238** (1987) 517

[6] D. Joyeux, S. Lowenthal, F. Polack, and A. Bernstein, in: **X-Ray Microscopy II** (Springer-Verlag, Berlin, 1988), p. 246; and D. Joyeux, and F. Polack, in: **OSA Proceedings on Short Wavelength Coherent Radiation: Generation and Applications**, eds., R.W. Falcone and J. Kirz (Opt. Soc. Am., Wash. D.C., 1988), Vol. 2, p. 295

[7] G.W. Stroke, in: **Optique des Rayons X et Microanalyse**, eds., R. Castaing, P. Deschamps and J. Philibert (Hermann, Paris, 1966) p. 30.

[8] See, for example, J.C. Solem and G.F. Chapline, *Opt. Engr.* **23** (1984) 193 and W.S. Haddad, D. Cullen, J.C. Solem, K. Boyer and C.K. Rhodes, in: **OSA Proceedings on Short Wavelength Coherent Radiation: Generation and Applications**, eds., R.W. Falcone and J. Kirz (Opt. Soc. Am., Wash. D.C., 1988), Vol. 2, p. 284

[9] E. Anderson, "Fabrication Technology and Applications of Zone Plates", in: **X-ray/EUV Optics for Astronomy and Microscopy**, ed., R.B. Hoover, *SPIE Proceedings* **1160** (1989) 2

[10] H. Rarback, et al., "The Scanning Transmission X-Ray Microscope at the N.S.L.S.", this volume.

[11] G. Beal, G. Boucharlat, J. Chabbal, J.P. Dupin, B. Fort, Y. Mellier, *Opt. Engr.* **26** (1987) 902

[12] C. Jacobsen, "X-ray Holographic Microscopy of Biological Specimens Using an Undulator", Ph.D. dissertation, Department of Physics, State University of New York at Stony Brook (Stony Brook, 1988) pp. 206-213

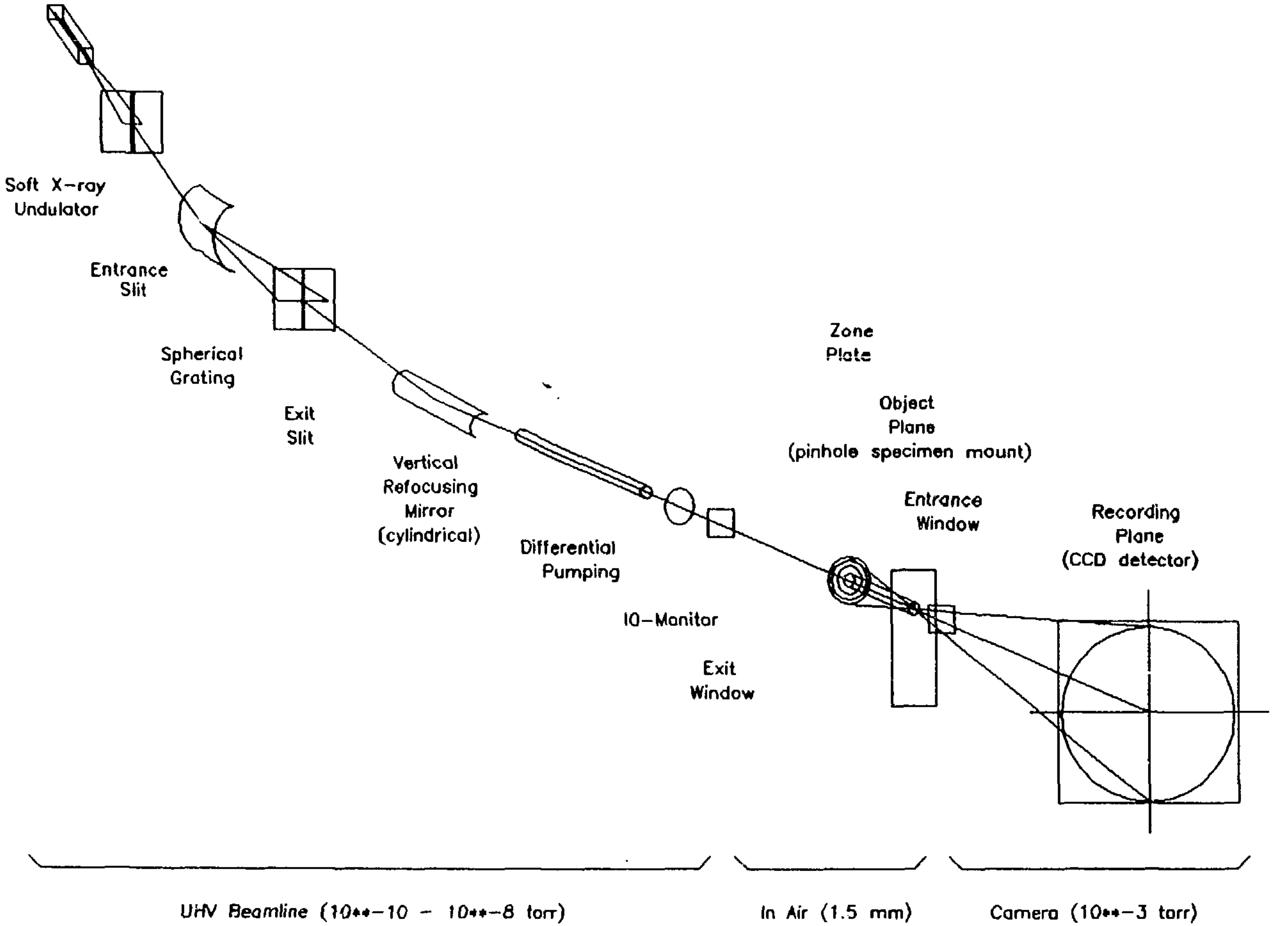
[13] G.W. Stroke, R. Restrick, A. Funkhouser and D. Brumm, *Phys. Lett.* **18** (1965) 274

Figure Captions

Fig. 1. Schematic diagram showing elements of the X1A soft x-ray undulator beamline optics for Fourier transform holography.

Fig. 2. High resolution test objects for Fourier transform holography, imaged with the Stony Brook/NSLS Scanning Transmission X-ray Microscope.

Fig. 3. Fringes formed with 3.6 nm x-rays and recorded with the CCD camera. The object illuminated was a 5 μm pinhole in a platinum foil. The reference source was formed with a zone plate and is located to one side of the pinhole center.


Soft X–ray
Undulator
Entrance
Slit
Spherical
Grating
Exit
Slit
Vertical
Refocusing
Mirror
(cylindrical)
Differential
Pumping
I0–Monitor
Exit
Window
Zone
Plate
Object
Plane
(pinhole specimen mount)
Entrance
Window
Recording
Plane
(CCD detector)
UHV Beamline (10**–10 – 10**–8 torr)
In Air (1.5 mm)
Camera (10**–3 torr)