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X-Ray Holographic Microscopy Experiments at the Brookhaven Synchrotron Light Source

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Abstract

Soft x-ray holographic microscopy is discussed from an experimental point of view. Three series of measurements have been carried out using the Brookhven 750 MeV storage ring as an x-ray source. Young slits fringes, Gabor (in line) holograms and various data pertaining to the soft x-ray performance of photographic plates are reported. The measurements are discussed in terms of the technique for recording them and the experimental limitations in effect. Some discussion is also given of the issues involved in reconstruction using visible light.

Introduction

The idea of recording a hologram with short wavelength radiation and reconstructing it in the visible is a very old one. It was at the heart of Gabor's[1] thinking when he first proposed the holographic method and was always intended to be a form of microscopy. The magnification would under ideal conditions be equal to the ratio of the reconstructing wavelength to the recording wavelength. The practical realization of holographic microscopy using X-rays has been attempted[2] since the earliest days of holography but with very limited success. The X-ray sources used in the early experiments had insufficient brightness to allow a reasonable flux of spatially coherent photons to be obtained for illumination of the sample. The brightest sources available during this period (the 1950's and 60's) were microfocus X-ray tubes and some holograms were made[3] with these, the best of which were those of Aoki, et al[4]. However, the long exposure times (around an hour or so) and low resolution (4 μm in the best case) gave little hope that useful scientific information about the sample could be obtained by this technique or any imaginable elaboration of it.

The harbinger of a change in this situation was the introduction of synchrotron radiation (SR) X-ray sources. Aoki and Kikuta[5] reported the first reconstructible hologram made with synchrotron radiation in 1972 and since that time there has been a steady growth in the brightness of synchrotron X-ray sources. The brightest ones currently in use are bending magnet sources on storage rings dedicated to the production of SR. If we confine our interest to the biologically useful region between the oxygen and carbon k edges (24 Å < λ < 44 Å) then the brightest sources are presently the 750 MeV ring at Brookhaven and the 800 MeV ring at Berlin. These sources are about 2-3 orders of magnitude brighter than the earlier sources and may be expected to give considerably better results. However, the real reason for the present reawakening of interest in X-ray holography is not the expected capabilities of bending magnet sources (in fact, even the improved quality of the X-ray beams they provide is still not expected to lead to scientifically useful holographic microscopy) but rather the even greater promise of undulator sources[6,7]. These will give a further 3-4 orders of magnitude improvement in source brightness compared to bending magnets and should allow the vast potential of three dimensional holographic imaging with soft x-rays finally to be realized.

The present authors have begun a series of investigations of the X-ray holographic imaging process using the 750 MeV storage ring at Brookhaven. We do not expect to discover useful biological information in our present experiments which are mainly concerned with imaging test objects at modest resolution. However, we do expect to be able to image objects which are simple enough that their holographic recording and reconstruction can be calculated. This should provide evidence for the validity of our present physical picture[8] of the image forming process. It will also allow us to rehearse the holographic technique, evaluate detector materials and so on.

In this paper we report experimental aspects of the program and the results obtained so far. We do not discuss the general character of X-ray holography or its theoretical basis and expected limitations since these questions are considered elsewhere[8].

Experimental Arrangements

We have carried out three one-week experimental runs on the U15[9] beam line at the Brookhaven 750 MeV storage ring. These were directed toward the following goals:

(i) The recording of Young's slits interference fringes. Comparison of various photographic films. Verification that the apparatus was sufficiently geometrically stable for X-ray interferometry.

(ii) Recording of Gabor in-line holograms of simple objects such as cylinders and spheres.

(iii) Same as (ii) but using a smaller source size, higher resolution film and smaller objects.

The arrangement for experiment (i) is shown in Fig. 1 and that for the other two experiments in Fig. 2. Both were supplied with soft x-rays of $\lambda \sim 30$ Å and $\Delta\lambda/\lambda \sim 10^{-2}$ from the U15 beam line system. The beam line contains a contamination barrier (9) which does allow non ultra high vacuum equipment to be connected as in Fig. 1, but it is far more convenient to have the arrangement of Fig. 2 where the holography experiment is separated from the beam line by a silicon nitride window which can withstand atmospheric pressure.

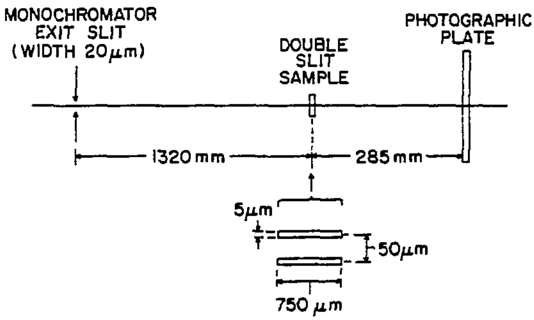


Figure 1. Schematic of experimental set-up for recording Youngs frings (experiment (i)).

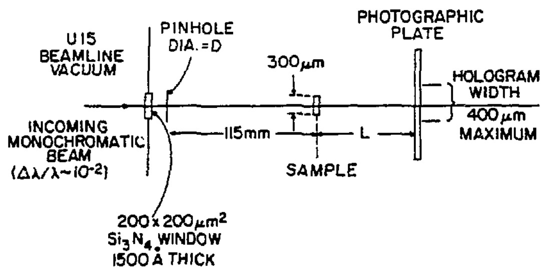
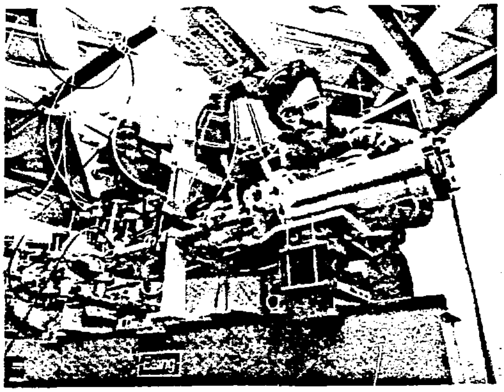


Figure 2. Schematic of experimental holograms. Parameters are as follows: Experiment (ii) D = 2 μm, L = 58mm, photographic plate is Kodak 131-02 with quoted resolution of 2500 lines/mm. Experiment (iii) D = 1 μm, L = 29mm, photographic plate

The general layout of the beam line with the holography sample chamber attached is illustrated in Fig. 3. The large aluminium tube on the right is a spacer piece. To its left is the sample chamber, to its right, the camera. The latter is a McPherson spectrometer camera which operates normally in vacuum but the film transport and shutter are both operable from outside the vacuum. Using these controls, we can record up to a dozen or so holograms without breaking vacuum. The turn-around time to break vacuum, develop a plate, load another one and pump down again to working pressure (10^{-2} torr) is about half an hour. The spacer piece can readily be interchanged for a different length piece and values of the sample to plate distance in the range 29-285mm can be selected. Immediately upstream of the sample chamber is a pinhole holder. This enables external interchange of pinholes with high reproducibility. The choices are 0.5, 1, 2, 10, 25 μm[10] or fully open.

The sample chamber is shown in Fig. 4. It incorporates a sample holder with X-Y manipulation and three viewing windows. One of these allows the sample to be viewed using a 40X visible light microscope. In order to achieve this an adjustable and retractable viewing mirror is provided.



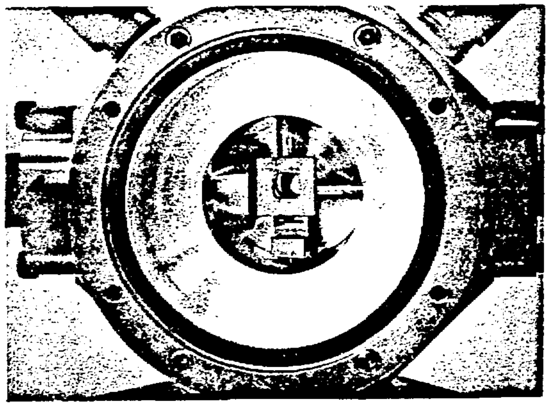


Figure 3. U15 beam line, showing holography experiment mounted on vibration isolated optical bench. The octagonal aluminum chamber at center is the sample chamber. The black object to the right is the camera.

Figure 4. Holography sample chamber: The sample is mounted to the square block in the center. This receives micrometer X-Y drives from the two shafts (above and right) to which it sticks magnetically. The sample is viewed by a 40x visible light microscope (left) with the aid of the 45° slotted mirror (below sample block). The latter is rotatable and retractable.

Alignment

Since it is necessary to provide a spatially coherent beam to illuminate the sample and provide the reference beam it is inevitable[8] that very small pinholes and x-ray beams need to be aligned. This is one of the main difficulties of the experiment. Our approach to this follows that of the x-ray microscope that also operates on U15[9]. The beam is followed by its visible light fluorescence emitted by a phosphor[11] viewed through various vacuum windows. For beams passing through pinholes less than about 25 μm diameter the light emitted by the phosphor becomes hard to see even with the monochromator set to zero order. For this case, we have designed a retractable light pipe system which passes through the edge of a 2-3/4 in. Conflat flange. The phosphor is on one end of the light pipe and the fluorescence is detected at the other by a 1/2 in. photomultiplier. With this system, we can easily 'see' monochromatic x-rays emerging from a 0.5 μm pinhole. To set the sample in the beam we select a 25 μm pinhole, tune the monochromator to zeroorder and shift the sample mounting block so that a region of the block which is coated with phosphor receives the beam. The position of the bright spot is then noted relative to the microscope graticule and the sample driven to that position. The desired pinhole for the experiment is then selected and centered using the light pipe system.

Experiment (i)

This was the crudest of the three experiments. Recordings were made of one dimensional Youngs interference fringes from a double slit sample with the monochromator exit slit as source. We show in Fig. 5 an example of the fringe patterns we recorded. The form of the pattern is very close to expectation and the contrast is good. It is clear from Fig. 5 that, at least for the fringe frequency involved in these recordings, the vibrational and thermal stability of the apparatus was quite adequate. In fact, we have had no problems of stability even at much higher fringe frequencies.

Figure 5. Youngs slits recording using the experimental set-up of Fig. 1 and a Kodak 131-02 plate. The fringe spacing is about 15 μm. Exposure = 285 mA.min, magnification = 70.

After a number of recordings, we began to get a feeling for the properties of the various films we were using. Our data are really only qualitative but soft x-ray data on photographic plates are so rare that we consider it useful to try to communicate them as best we can. In Table I we list six photographic plates that we used and the exposure needed to achieve a 'good' looking fringe pattern. The relative accuracy is estimated to be about a factor 3 and the absolute accuracy a factor of 10 or more. We note the following points:

1. The absolute exposures expressed in energy units are larger than the corresponding figures for visible light by about 1-2 orders of magnitude. This is not unexpected.

2. Kodak 131 is normally quoted as about 30 times faster than Agfa 8E56 HD in visible light. We estimate it fairly reliably as only 2 times faster for soft x-rays. This again is not very surprising in view of the very different statistics of x-ray recording (one photon per activated grain, etc.).

3. The figure of merit for x-ray holographic recording would appear to be the detective quantum efficiency (DQE) times the dynamic range (number of gray levels). The only x-ray DQE measurements on film that we know of are those of Niemann[13] and these data do not include any of the films that we have used. We may speculate that our 'normal' films may be similar to those measured by Neimann, i.e., DQE ~ 0.2. However, it is not obvious how the low gelatin, VUV films would fit in. They lose fewer photons by absorption in gelatin but seem to add photographic noise via infectious and spontaneous development. The most interesting of them appears to be the Ionomet material which has no gelatin and essentially no developed grains in unexposed areas. Thus, it should have very high DQE. This has to be set against its total lack of dynamic range, i.e., no gray levels.

4. A useful set of measurements of film speed at various soft x-ray energies were made by Koppell[14]. The nearest useful data point we can get from this is that density one was obtained using TiL_α x-rays (453 eV) and Kodak 101 film with an exposure of 0.13 ergs/cm^2. 101 film is quoted to be about 10 times faster than SWR. This suggests that our measurements of speed are very low.

TABLE I - PHOTOGRAPHIC PLATE PROPERTIES

Material	Development	Exposure (ergs/cm^2)	Quoted Resolution (μm)	Noise
Kodak TE	5 min. D-19 (undiluted)	29,000	< 0.5	Good
Kodak 131	4 min. D-19 (undiluted)	2,200	0.4	Good
Kodak SWR*	2 min. D-19 (diluted 1:1)	56	2-5	Very Bad
Ilford Q*	2 min. D-19 (diluted 1:1)	14	2-5	Bad
Ionomet[12] Gel** Free Photoplates	As per Mfg. instructions	50	2-5	Good
Agfa 8E56HD	4 min. D-19	4,400	0.2	Good

* Low Gelatin VUV Film
** Gelatin Free Silver Halide

Experiment (ii)

In this experiment, we recorded Gabor in-line holograms of simple objects using the arrangement shown in Fig. 2. Once a correct alignment was achieved, the experiment was surprisingly easy. The recordings were all made using a 2 μm pinhole source and Kodak 131-02 high speed panchromatic holographic film which has a quoted resolution of 2500 lines/mm.

At the time of writing we do not know how well the finest fringes will reproduce in this publication. Our comments are based on the original recordings as viewed with the visible light microscope.

Fig. 6 shows the hologram of several 10 μm diameter wires. We see a rich fringe structure superimposed on the central peak of the Airy Disk of the 2 μm pinhole. Fig. 7 shows a similar recording of a group of 3-5 μm diameter spherical, glass beads mounted on a Si_3N_4 window. Again, a complex fringe structure is seen, this time resembling more closely the 'blotchy' appearance of highly magnified visible light holograms. Fig. 8 shows the hologram of a single wire. It is very reminiscent of the analagous pictures taken in visible light by Tyler and Thompson[16] and of the famous first-ever hologram recorded in 1932 by Kellstrom[15] using Aℓ Kα X-rays and reconstructed by El Sum in 1952[2].

Experiment (iii)

In this experiment, we tried to press the Gabor in-line method further toward its limit for X-ray holography using a bending magnet source. We changed to a 1 μm pinhole and Agfa 8E56 HD film which has a quoted resolution of 5000 lines/mm. In light of the higher resolution of the film we reduced the sample-to-film distance by a factor of two to 29mm. This should give us a 16-fold disadvantage in speed due to the pinhole change, a four-fold advantage due to the distance change and two-fold disadvantage due to film speed or an 8-fold disadvantage overall. This was about what we observed.

As a preliminary, we did an intermediate experiment using the same geometry as experiment (ii) and Agfa 10E56 NAH film. With this, we repeated the recording of Fig. 6. We see (Fig. 9) that the graininess of the film is considerably improved but the recorded fringe pattern if anything, seems less complete. We then switched to the full configuration of experiment (iii) and recorded a number of holograms of samples of amosite asbestos needles. The needles (Fig. 10) had diameters in the range 0.5 - 2 μm. An example is shown in Fig. 11. This picture demonstrates that complex patterns of fringes including many overlaps can be recorded by this method and we see in some cases dozens of fringes, apparently due to one needle. The uneven illumination is a problem as before and we notice for this case that the pinhole seems to be oval to some degree. We also repeated the single wire experiment for the experiment (iii) geometry (Fig. 12), this being the best case from the point of view of comparison with theory.

Figure 6. Gabor hologram of several 12.5 μm wires made using the experiment (ii) set up and using Kodak 131-02, exposure = 250 mA.min., magnification = 175.

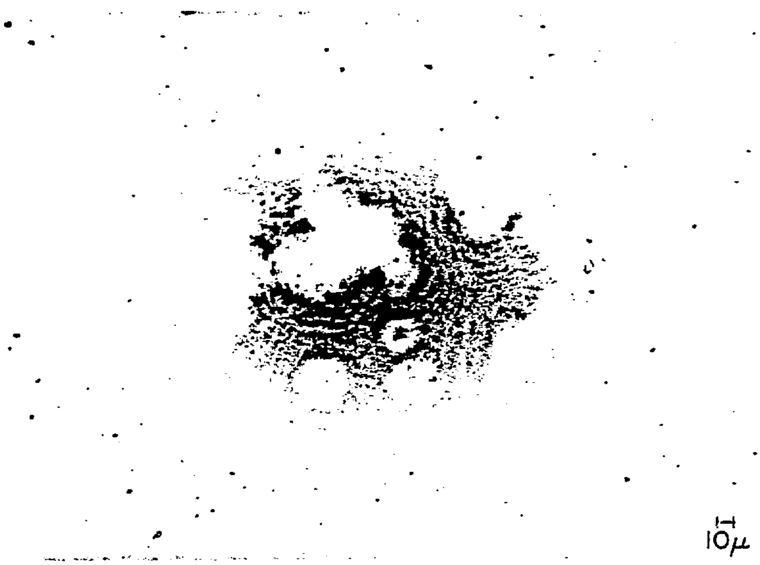


Figure 7. Gabor hologram of a number of glass spheres 3-5 μm diameter using experiment (ii) geometry. Kodak 131-02, exposure = 350 mA.min, magnification = 300.

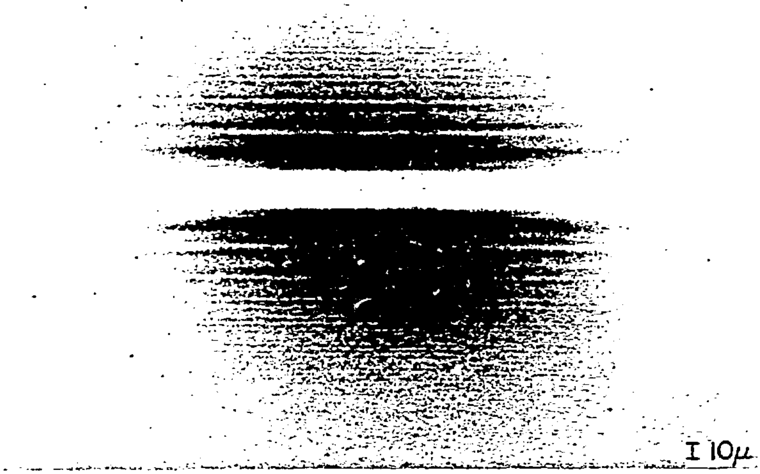


Figure 8. Gabor hologram of a single wire using experiment (ii) geometry. Kodak 131-02, exposure = 165 mA.min., magnification = 300.

Figure 9. Gabor hologram of several wires using experiment (ii) geometry. Agfa 10E56NAH exposure = 1200 mA.min., magnification = 175.

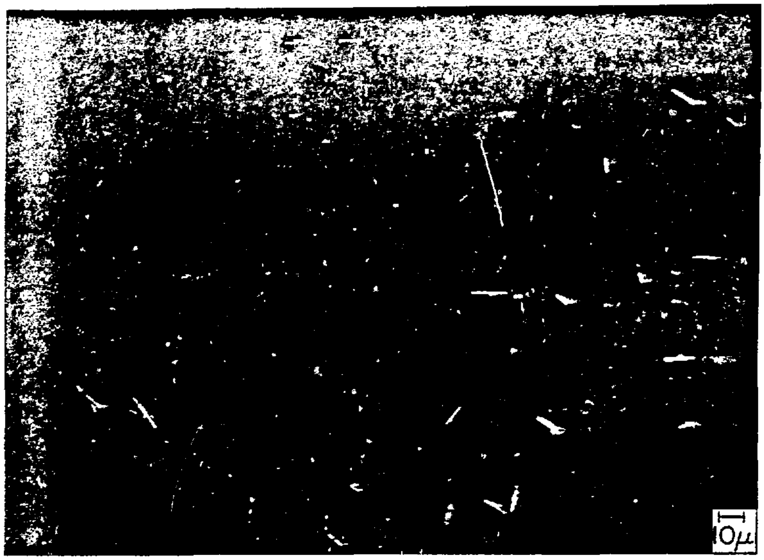


Figure 10. Visible light micrograph of a typical selection of amosite asbestos fibres diameter 0.5-2mm as used in the hologram in Fig. 13. Magnification = 450.

Figure 11. Gabor hologram of amosite asbestos fibres as shown in Fig. 12 using experiment (iii) geometry. Fibre diameter is 0.5-2 μm. Afga 8E56HD, exposure = 2800 mA.min., magnification = 300.

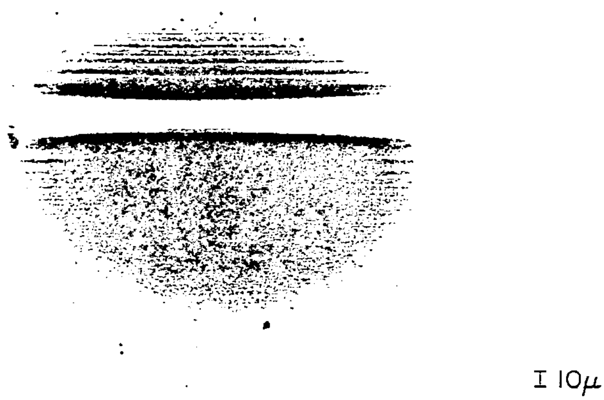


Figure 12. Gabor hologram of a single 12.5 μm diameter using experiment (iii) geometry. Agfa 8E56HD, exposure = 3800 mA.min., magnification = 300.

In all the holograms the fringe contrast diminishes as the order of the fringe becomes higher until eventually the fringes become invisible. It is not clear why this happens. For the holograms recorded on 131 film it is obvious that the fringe spacing is quite close to the resolution limit of the film even though the spacing is about 1 μm or wider. The quoted 2500 lines/mm resolution for this film is hard to believe for these pictures. It is also clear that a limit due to signal to noise considerations is being approached and this is related to the very uneven illumination provided by the central maximum of the Airy pattern of the pinhole. The latter is related, in turn, to the use of light transmitted through the sample for providing a reference beam for this type of hologram. The situation could, in principle, be improved by using a smaller pinhole. The Airy Disc would then be wider, however, the cost in throughput is severe. For an n-fold smaller pinhole, the pinhole transmission is worse by a factor n^2. The diffracted light is also spread over a film area which is n^2 times larger, leading to an n^4-fold loss of intensity at the film. An additional effect caused by the use of the Airy Disk of the pinhole is incomplete spatial coherence for fringe patterns that extend over a significant fraction of the width of the Disk. Again the result is a worsening of fringe contrast.

<u>Reconstructions: Theory</u>

We have made various attempts at reconstruction of our holograms using visible light lasers. There are a number of problems that must be resolved if we are to achieve good results doing this:

(i) Aberrations: The change of wavelength by a factor of 4416/31 for He:Cd or 6328/31 for He:Ne brings about a huge degree of spherical aberration. Other aberrations are small for nearly on axis points. One can easily calculate the effect of the aberrations using the theory given by, for example, Meier[17] or Smith.[18] Results show that aberrations are substantial for reconstruction of the original hologram with a He:Cd laser. For a He:Ne laser they are even worse.

The only way to reduce the spherical aberration without changing either of the wavelengths is to scale up the hologram by a factor of at least twenty or so. This is an unwelcome step in an otherwise lensless imaging process. but it is certainly possible using standard photolithographic equipment. It was part of the procedure originally envisioned by Gabor.

(ii) Limitations of resolution due to recording conditions: It has long been known[19] that the finite source size and finite film resolution will limit the resolution of the reconstructed image using this type of hologram. Following the method of Baez[19], we calculate that the resolution limit in our experiments due to these causes is about 0.8 μm for experiment (i) geometry and 0.25 μm for experiment (ii).

(iii) Limitation of resolution due to signal to noise ratio: It is well known that the resolution of a zone plate is equal to the width (Δ) of the finest zone. Thus, in order to achieve the resolution limits found in (ii) above, it is necessary to record zone plate fringes out as far as a spacing of 0.8 μm and 0.25 μm respectively for the two experiments. This requires that the signal to noise of the recordings be good enough to record this information and as shown in Reference 8 the total number of photons needed to record the required patterns is proportional to Δ^{-8}. This is the reason that none of our holograms will achieve the resolution predicted by the Baez equations. The resolution they will achieve can be found from standard zone plate theory by noting that $\Delta = r_n/2n$ where r_n is the radius of the nth zone of the pattern. If we set r_n equal to the distance from the center fringe to the last discernible fringe and n equal to the number of fringes then we arrive at the likely resolution of a well reconstructed image. One could also simply estimate the narrowest zone width by eye, but the $r_n/2n$ relationship reassures us that the expected 1/n dependence for the resolution of an n-component diffracting structure does, indeed, appear.

Reconstructions: Experimental

We are presently attempting to carry out visible light reconstructions of our holograms using both the original hologram and various magnified forms both positive and negative. We have had some success, at least as judged by eye, for the case of the original holograms and He:Cd laser light. The reconstructed images are extremely close to the hologram and there is a difficulty distinguishing between them and the shadow of the hologram itself. There is also a difficulty in photographing such an image. Our efforts in making reconstructions using magnified holograms have not been successful so far. We believe that success is possible. We have, after all, many more fringes in our holograms than any previous x-ray holographers due to our superior source qualities. We expect therefore that when we can magnify our holograms with sufficiently good contrast and dimensional linearity we will be able to produce a reconstruction commensurate with the quality of the original hologram. Our efforts are continuing on this question.

Acknowledgments

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