CONF-871124--21-Rev.1

UCRL- 96783, Revision 1
PREPRINT

FABRICATION AND EVALUATION OF TRANSMISSIVE MULTILAYER OPTICS FOR 8 keV X RAYS

R.M. Bionta
A.F. Jankowski
D.M. Makowiecki

This paper was prepared for submittal to
1987 Fall Meeting of the Materials Research Society
November 30 - December 5, 1987
Boston, Massachusetts

January 21, 1988

Lawrence
Livermore
National
Laboratory

FABRICATION AND EVALUATION OF TRANSMISSIVE MULTILAYER OPTICS FOR 8 keV X RAYS

R.M. Bionta, A.F. Jankowski, and D.M. Makowiecki, Lawrence Livermore National Laboratory, P.O. Box 5503, M.S. L-278, Livermore, CA 94550.

UCRL--96783-Rev.1

DE88 005644

ABSTRACT

We have made and tested several sliced multilayer structures which can function as transmissive x-ray optical elements (diffraction gratings, zone plates, and phase gratings) at 8 keV. Our automated multilayer sputtering system is optimized to sputter layers of arbitrary thickness for very large total deposits at high deposition rates. Diffraction patterns produced by the multilayer devices closely match theoretical predictions. Such transmissive optics have the potential for wide application in high resolution microscope and spectrometer systems.

Introduction

Zone plates are focusing devices constructed of alternating transparent and opaque layers [1]. Constructive interference occurs at the focal point of the zone plate when the zones are properly spaced. Using electron beam and holographic lithography, zone plates have been constructed that operate at x-ray energies below 1 keV [2-6]. Unfortunately, for higher x-ray energies of interest in the industrial laboratory, namely 5 to 20 keV, zone plates cannot be fabricated by the holographic and electron beam lithography techniques, because such energies require large aspect ratios and very fine zones. To date, the only way to manipulate x-ray energies above 5 keV is by the use of grazing incidence mirrors or near-grazing incidence mirrors coated with multilayer coatings to increase the reflectivity.

We have investigated a different technique foı fabricating zone plates for use in the 5 to 10 keV regime. Ultimately we plan to make zone plates by sputtering alternating layers of opaque and transparent materials onto a thin wire core, then slicing perpendicular to the core axis to produce many zone plates. This technique shows promise for making x-ray optical elements that can be used in industrial crystallography, microprobe, and radiography equipment. In a previous publication [7] we reported on the good agreement between the measured performance of an Al/Ta diffraction grating and our numerical simulation. Details of the test bed used to evaluate the gratings and zone plates may be found elsewhere [8]. In this report we concentrate on the fabrication techniques used tc produce diffraction gratings and linear zone plates.

In the past 40 years, we have seen impressive advances in the multilayer technology used to enhance the reflectivity of grazing and near-grazing incidence mirrors [9-12]. Even so, transmissive multilayer optics is sufficiently novel to require considerable improvement in the techniques of multilayer sputtering. In general, transmissive optics require much thicker layers, ruling out techniques such as molecular beam epitaxy which is mainly useful for making very thin layers. Also, multilayer transmissive optics usually require many more layers than reflective optics. Because the number of layers often exceeds 10,000, to make transmissive multilayer optics requires computer control of the process. Futhermore, since the total thickness of the sputtered multilayer determines the aperture of the lens, very thick coatings are required. This means that the sputtering system must run for several days in order to achieve thicknesses approaching 500 μm, requiring extensive computer-controlled monitoring and logging of the sputtering process. In addition, absolute thickness control is required to achieve proper constructive interference. And finally, to make a wide variety of optics with different focal lengths requires a sputtering system capable of sputtering layers of arbitrary thickness. This means that there must be some way of individually specifying the thickness of several thousand layers.

Sputtering

For simplicity, we have avoided the problems of sputtering on a wire core by sputtering onto a flat substrate and then slicing the coatings into thin slabs. This technique allows us to produce linear zone plates which focus x rays from a point source into a line, much as a cylindrical lens does in ordinary optics. The sputtering system, shown in Figure 1, is optimized to produce transmissive linear optics. The system has two sputtering sources positioned 90° apart with respect to a rotating mandrel. Two substrates, measuring 5 x 20 mm, are mounted on the mandrel, and are alternately positioned in front of each source. Cylindrical shutters cover the opposing source during deposition, and the substrate is heated to ~300°C by a quartz lamp inside the mandrel. Two quartz crystal microbalances track the deposition rate during the process. The sputtering guns, shown in Figure 2, are of our own design. They employ neodymium iron boron magnets to produce very high field strengths. The magnets are configured into a small cylinder located in the center of the gun and a ring magnet surrounding it. The field strength produced by these magnets are sufficiently high that targets up to 0.75 inch thick may be employed. These very thick targets allow us to continuously sputter for several days in order to build up very thick coatings. The guns are powered by 10 kW power supplies in a DC mode.

Deposition is under the control of an HP9000 series computer (CAD/CAM), which is used to design and build the multilayer. To use the lens-making software, the user enters the focal length of the lens, its diameter, and the materials to be used. Then the program calculates the thickness of each layer and the material type. The thicknesses and materials are written into a file stored on a floppy disk. Both zone plates with variable spacing and diffraction gratings with constant spacing can be built. The software has provisions for inverting and concatenating the lens files to make more complex designs. The sputtering software is divided into an initialization section, a sputtering section, and a wrap-up section. During initialization, the user has the option of changing the preprogrammed sputtering parameters before the user inserts the diskette containing the lens file. These parameters include sputtering power, pressure, warm up time, and tooling factor, which is defined as the ratio of the thickness on the substrate to the thickness measured by the crystal.

To begin sputtering, the program first turns on the sputtering gas and purges the system with argon for about an hour. Then the program goes into the layer loop. For each layer, the program reads the layer thickness and material from the diskette, sets the pressure of the sputtering gas, brings up the sputtering gun to the desired sputtering power, moves the substrate to face the sputtering gun, and then opens the shutter. During deposition the substrate is made to oscillate back and forth in front of the sputtering gun (ensuring a uniform layer thickness). The computer continuously reads the value of the thickness on the crystal until the desired value is reached, taking into account the crystal tooling factor. When the desired thickness is reached, the computer closes the shutter, rotates the substrate to a neutral position, and powers down the gun. After all layers are complete, the program goes into the wrap-up mode, during which it shuts down the high voltage on the guns, turns off the argon, and prints out a summary sheet of the sputtering run.

Since the sputtering system is intended to run for many days without supervision, it is necessary to have emergency monitoring and shut-down procedures programmed into the computer. This is accomplished by having the computer read the current in the sputtering guns during deposition of the layers. The current is read once every second. If the current

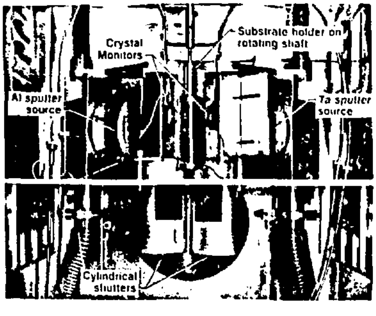


Figure 1. Automated sputtering system for multilayer deposition.

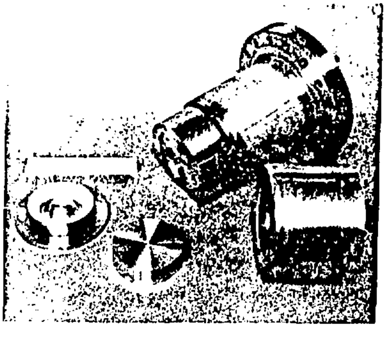


Figure 2. Sputtering source guns and targets (eroded and new).

exceeds a predetermined value (10 amps) for more than 4 seconds, the computer shuts off all power supplies and stops the process. Another fault check involves the measurement of the amount of time it takes to sputter a layer of a given thickness. If for a given layer the deposition time is more than 600% of the time that it should take based on the thickness of the current layer and the sputtering rate achieved on previous layers, the computer shuts down the operation. This is useful if the shutter sticks or the crystal becomes broken. In addition, the computer monitors the deposition process by recording all deposition parameters, including the substrate temperature, the sputtering rate, the current, the voltage, and the pressure, once each minute onto a log file on the disk. After the deposition run is over, plots of these recorded parameters are helpful in understanding what happened during the run.

The crystal tooling factors are obtained in special calibration runs. A cross-section of a calibration run, shown in Figure 3, consists of 4 μm of Ta, followed by 20 μm of Al, 20 μm of Ta, 4 μm of Al, then 4 μm of Ta. For calibration purposes we measure the actual thickness of the 20 μm layers and correct the crystal tooling factor for any discrepancies. The accuracy of the calibration is limited by our ability to measure the 20-μm-thick layers. In particular, it is limited by the sharpness of the interface between adjacent layers. In practice, we can achieve thickness measurement errors of less than 0.5 μm, which with a thickness of 20 μm gives a calibration error of $\leq$ 2.5%. Since the crystals are further away from the sputtering source than the substrate, the tooling factors are large, and generally vary between 8 and 15. The tooling factors are very sensitive to the distance between the target and the crystal. Figure 4 shows a plot of the tooling factor as a function of distance from the target on a log scale. The variation of tooling factor of distance in this sputtering system is seen to be an exponential, with a slope of about 37 mm. Thus an error of 1 mm in crystal position corresponds to a 3% error in calibration.

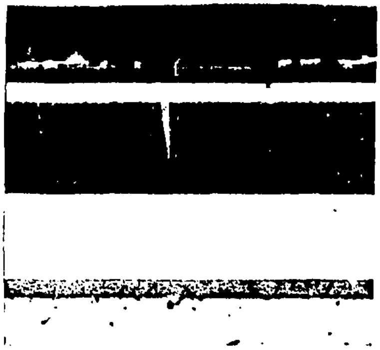


Figure 3. Cross section of Al/Ta calibration run.

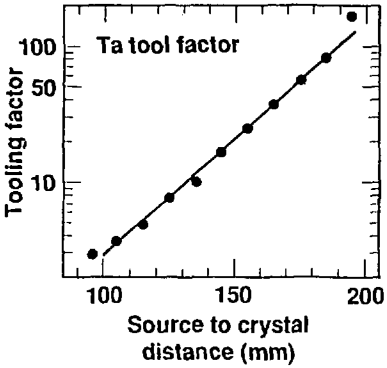


Figure 4. Typical plot of crystal tooling factor (defined as the ratio of the deposition rate on the substrate to the deposition rate on the crystal) vs. distance from sputtering source.

With a 0.75-inch-thick Al target and 1 kW power, we can achieve deposition rates of 100 Å per second. The influence of the strong magnetic field can be seen in Figure 5, which shows a log-log plot of the sputtering voltage versus the sputtering current for the Al gun. The plot shows the results for several layers as the gun voltage is raised and lowered for each layer. For magnetron operation, the current should be proportional to some power of the voltage, which would produce a straight line in the log-log plot. The slope of the line is related to the efficiency of the system for retaining the plasma. In the beginning of the run, when the target is thick, the slope is very shallow. It takes a large change in voltage to change the current. Towards the end of the run, the slope is very steep, because the target has eroded away considerably and the sputtered surface is now much closer to the magnetic field.

Another effect of target erosion is shown in Figure 6, which shows the sputtering rate versus time for a six day run at constant power. It can be seen that over a period of six days, the sputtering rate for Al drops by more than a factor of two as the target erodes.

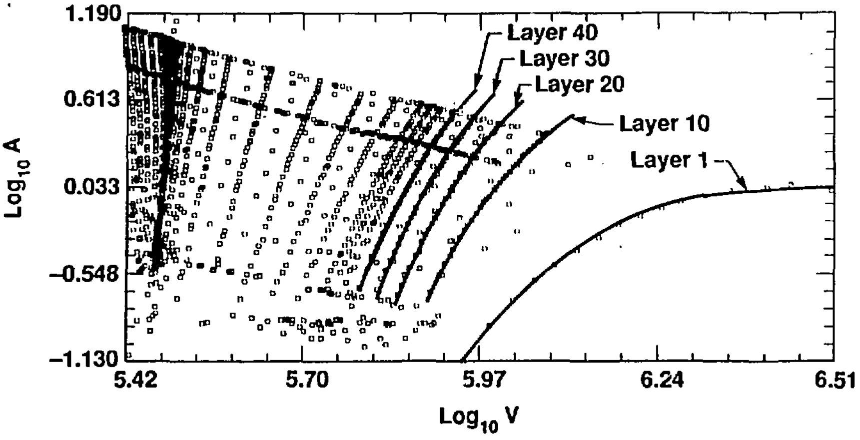


Figure 5. Current voltage relationship for magnetron sputtering of Al. Data was recorded one per minute during deposition as well as every tenth layer during the power up of the sputtering gun.

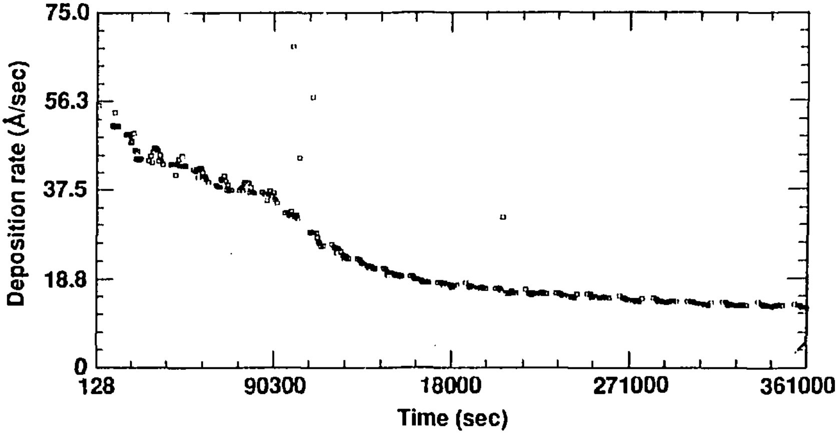
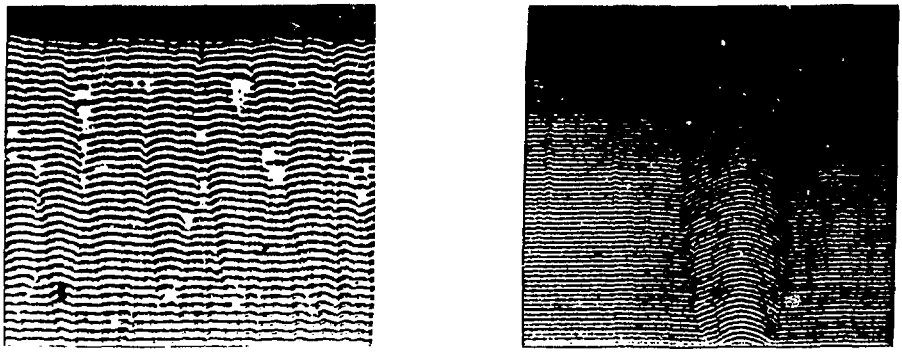


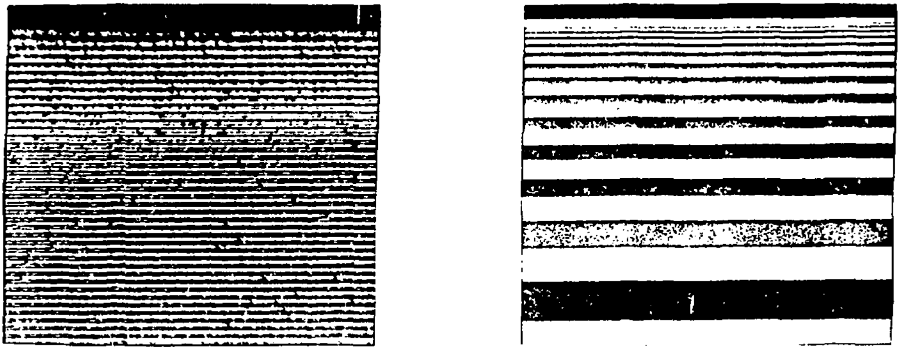
Figure 6. Reduction of Al sputtering rate during 6-day run due to target erosion.

Several improvements had to be made to the system in order to obtain extremely smooth layers of Al and Ta. Figure 7 shows several Al/Ta multilayers sputtered under various conditions. Our first coatings were done with the mandrel continuously rotating at 60 rpm and an argon pressure of 5 mTorr. *Continuously rotating the mandrel would ultimately allow us to* make as-deposited blended coatings by co-sputtering the two materials simultaneously. Unfortunately, rotating the substrate caused most of the coating to be deposited at oblique angles, promoting columnar defects as shown in Figure 7a. Changing to normal incidence coating by positioning, then holding the substrate in front of each gun produced smoother coatings as shown in Figure 7b. The coatings still had a few gross defects which were traced to fine particles of Ta that drifted onto the substrate. We found that at 5 mTorr the Ta coatings were highly stressed and tended to flake off of the shutters and surfaces of the chamber, causing very fine debris to land on the substrate and introduce defects. To overcome this problem we made test runs of Ta at various pressures in order to determine the pressure at which the Ta coatings had the least amount of stress. This was done by sputtering Ta films on very thin Ta substrates at various pressures and observing the stress-induced curling of the substrates. At low pressures, between 5 and 30 mTorr, the coatings were compressive and tended to curl inward. Above 60 mTorr, the coatings were tensile and tended to curl outward. At 50 mTorr, however, the coating seemed to have very little stress and did not curl the substrate, indicating that 50 mTorr was the pressure we should use to sputter Ta. Unfortunately, at 50 mTorr the Al deposition rate is very low, so it is necessary to sputter Al at 5 mTorr and Ta at 50 mTorr. We subsequently modified our control system so that the computer had control of the pressure. Figure 7c shows a thick multilayer coating made under these conditions, where the Al layers are sputtered at 5 mTorr and the Ta layers are sputtered at 50 mTorr. There is a considerable improvement in the quality of the multilayer as compared to Figure 7b, chiefly due to the fact that the Ta sputtered at 50 mTorr sticks very well to all of the surfaces inside



7a. Rotating substrate produces columnar defects.

7b. Nonmoving substrate, with gross defects due to particle contamination.



7c. Coatings sputtered at optimal pressures, with columnar defects in aluminum.

7d. Substrate temperature of 300°C results in extremely smooth coatings.

Figure 7. Al/Ta multilayers sputtered under various conditions.

of the chamber, and no debris is introduced onto the substrate. The remaining defects in Figure 7c are due to the columnar growth that is a property of Al coatings. This growth can be minimized by sputtering Al at a high substrate temperature, around 300°C [13]. To accomplish this, we located a quartz heater behind the substrate with a temperature control system that keeps the temperature of the substrate at 300°C. The resulting coatings were extremely smooth, as shown in Figure 7d.

Finishing

To make even thicker structures, we sputter two coatings at once onto two separate substrates and then bond them together in the middle. This is possible because the diffraction gratings are periodic, and the zone plates are symmetric about their axis. We use a solid state bonding technique to bond the two coated substrates together. The last layer of each coating is Al. The two substrates are placed facing each other in a vacuum chamber where they are brought up to a temperature of 550°C and then pressed together at a pressure of 3000 psi. Figure 8 shows a zone plate before and after bonding. The bonds are strong and durable, and the bonded Al layers behave as if they are one coating.

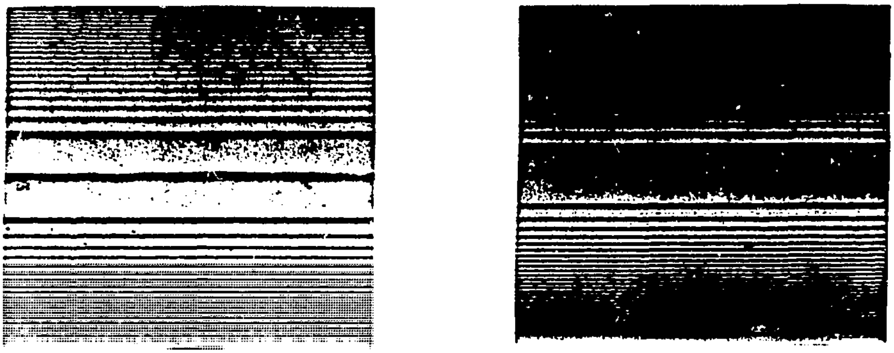


Figure 8. Lens bonding process. Al/Ta zone plate before bonding (a) and after bonding (b).

After bonding, the coated substrates are sliced into wafers perpendicular to the layers. These wafers are about 0.050 inch thick. The sliced wafers are polished on one side and then glued down to a thinning fixture, where they can be polished from the other side. The fixture has boron carbide shims which stop the polishing at the desired thickness. Figure 9 shows a thinned mounted lens.

X-Ray Testing

The thinned lenses are tested in our x-ray test bed, which has a microfocus x-ray source with a copper anode. The x-ray source emits x rays at 8 keV. The lenses are positioned according to the thin lens law to image the x-ray source onto our scanning pinhole detector, which enables us to record the image. Diffraction gratings are mounted midway between the source and detector where the interference pattern is observed. Figure 10 shows the diffraction pattern produced by one of our diffraction gratings in this apparatus.

Future Work

Several improvements in our process are envisioned in order to make improved x-ray optics. First, the 2.5% calibration error is too large to produce high quality zone plates, which require an absolute calibration of < 1%. To achieve this, we plan to install an interferometer in the sputtering system which will register the thickness change of the coating during the deposition process. To achieve very-high-efficiency focusing devices requires the use of blazed structures having a continuously varying concentration of the two materials across each zone, in a saw-tooth like pattern. This will involve co-depositing the two materials simultaneously. We plan to use the interferometer to determine the thickness of the coating, and use the crystals to set the ratios of the two materials. Finally, we are constructing a sputtering system that will deposit onto thin strands of optical fiber in order to make circular lenses. The circular lenses will be very similar to transmissive lenses in visible optics, and will avail a wide assortment of optical techniques to the x-ray regime.

The authors are indebted to the technical staff of 'O' Division's X-Ray Microscopy Project who made this work possible, including: E. Ables, K. Cook, D. Coufal, P. Gabriele, H. Highstone, L. Kennedy, K. Miller, H. Olson, L. Ott, R. Tilley, R. Vital, and T. Viada, as well as the useful advice given by T. Barbee and L. Wood.

This work was performed under the auspices of the United States Department of Energy by Lawrence Livermore National Laboratory under Contract W-7405-Eng-48.

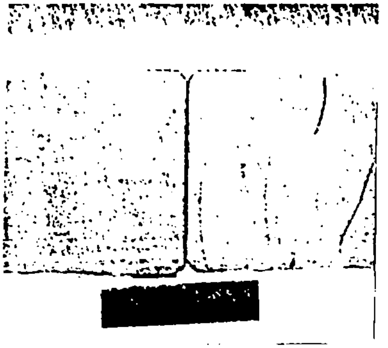


Figure 9. Thinned mounted lens.

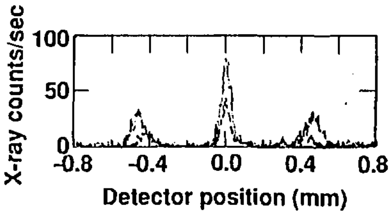


Figure 10. X-ray diffraction pattern produced by a diffraction grating.

REFERENCES

1. J.L. Soret, Arch. Sci. Phys. Nat. 52, 320 (1875).
2. A.V. Baez, J. Opt. Soc. Am. 42, 756 (1952); 51, 405 (1961).
3. G. Schmahl and D. Rudolph, Optik 29, 577 (1969).
4. J. Kirz, J. Opt. Soc. Am. 64, 301 (1974).
5. N.M. Ceglio, A.M. Hawryluk, and M. Schattenburg, J. Vac. Sci. Technol. 1, 1285 (1983).
6. R. Tatchyn, in X-Ray Microscopy (Springer, Berlin, 1984) 40.
7. R.M. Bionta, Appl. Phys. Lett., 51, 725, (1987).
8. R.M. Bionta, A.F. Jankowski, and D.M. Makowiecki, in X-Ray Microscopy, Springer-Berlin, 1988.
9. J. DuMond and J.P. Youtz, J. Appl. Phys. 11, 357 (1940).
10. J. Dinklage, J. Appl. Phys. 38, 3781 (1967).
11. E. Spiller, Appl. Phys. Lett. 20, 365 (1972); Appl. Opt. 15, 2333 (1976).
12. J.H. Underwood, T.W. Barbee, and D.C. Keith, Proc. Soc. Photo-Opt. Instrum. Eng. 184, 123 (1979).
13. J.A. Thornton, Ann. Rev. Mater. Sci. 7, 239 (1977).

DISCLAIMER