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# Profile coatings and their applications*

Chian Liu [a], R. Conley, L. Assoufid, and A. T. Macrander

Experimental Facilities Division, Argonne National Laboratory, Argonne, Illinois 60439

G. E. Ice and J. Z. Tischler

Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831

K. Zhang

Department of Biological, Chemical and Physical Sciences, Illinois Institute of Technology, Chicago, Illinois 60616

[a] Electronic mail: cliu@aps.anl.gov

We report a method of profile coating to achieve a certain selected thickness profile of a thin film coating using dc magnetron sputtering. In profile coatings, the substrate is passed over a contoured mask at a constant speed to obtain a desired profile along the direction perpendicular to the substrate-moving direction. The shape of the contour depends on the desired profile and the thickness distribution directly above the gun at the substrate level. Four-inch-diameter Si wafers were coated through a 100 x 152 mm$^2$

aperture on the top of the shield can. The thickness distribution was then obtained using a spectroscopic ellipsometer with computer-controlled X-Y stages. A model has been developed to fit the measured thickness distribution. The relative thickness weightings are then obtained at every point 1 mm apart for the entire open area of the aperture. When the substrate is moving across the shield can during depositions, the film thickness is directly proportional to the length of the opening on the can along the moving direction. By equating the summation of relative weighting to the required relative thickness at the same position, the length of the opening at that position can be determined. By repeating the same process for the whole length of the required profile, a contour can be obtained for a desired thickness profile. The contoured mask is then placed very close (~1 mm) to the substrate level on the shield-can opening. The number of passes and the moving speed of the substrate are determined according to the required thickness and the growth rate calibration. This method of profile coating has been applied to coat laterally graded W/C multilayers. It has also been applied to coat Au on a cylindrical mirror to obtain an elliptical mirror for x-ray focusing applications. Test results for these applications will be presented.

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## I. INTRODUCTION

In science and technology, it is often necessary to obtain certain selected thickness profiles for thin-film studies or applications. In nanoscience and surface magnetism for example, finding out how the physical properties of a thin film will change when the film thickness goes to the ultrathin limit is essential.[1, 2] In x-ray optics, an x-ray mirror is often coated with a uniform metal film to increase the critical angle of the mirror. To better focus the x-ray beam, an elliptical surface profile is desired for x-ray mirrors. Also, multilayer optical components are widely used in x-ray optics. [3-5] Multilayers usually consist of alternating layers of high electron density and low electron density to simulate the structure of a natural crystal, as in crystal optics. Generally each layer in a multilayer has a uniform thickness. In some applications laterally graded multilayers are more desirable. [6] A laterally graded multilayer has a continuously varying d spacing along one direction of the mirror. One material in the multilayer is uniform in thickness, and the other has a specified lateral thickness profile. In all these applications, one needs a capability to do profile coating.

In recent years, we have developed a method of profile coating for a variety of applications using dc magnetron sputtering. In this paper, we summarize the details of the technique and report its applications in obtaining elliptical x-ray mirrors, in improving film uniformity, in graded multilayers, and in studying small d-spacing x-ray multilayers.

## II. PROFILE-COATING TECHNIQUES

The profile-coating technique evolved naturally at the Advanced Photon Source (APS) deposition lab. The deposition facility consists of four large vacuum chambers, each 16 inches in diameter and 66 inches long. Three CTI model CT-8 cryo pumps and an Alcatel ADP 81 dry pump provide a base pressure of < 1 x $10^{-8}$ Torr for the system. Samples on a sample holder can be loaded into a carrier, which can be moved from chamber to chamber by a computer-controlled transport system. Four 3-inch-diameter magnetron sputter guns are deployed in the deposition chamber. The sputter targets are facing up, and the substrates are facing down. During the deposition, the substrates are usually moving. We have used masks and the linear substrate motion to improve the uniformity of coatings. The mask is placed close to the substrate level on a shield can over the sputter gun. Uniform deposition can be achieved through the design of a shaped-aperture mask over the sputter gun. Later we used this technique to make laterally graded W/C multilayers, where the W layer was kept uniform in thickness and the C layer had a wedge shape.[6] In both cases of uniform and graded coatings, the profile of the film thickness is known and well defined. The same technique is improved to make profile coatings for elliptical mirrors. In this application, the desired surface profile after the profile coating on a cylindrical mirror should be an ideal elliptical surface. The coating profile is determined by the difference between the ellipse profile and the measured profile of the cylindrical substrate from a long trace profiler (LTP). Every mirror requires a different coating profile. Also, the profile is usually not mathematically well defined, so that it can only be determined through point-by-point calculations. According to our experience, a profile calculated for each position 1 mm apart is sufficient. A spline fit of the calculated data points then provides a smooth curve.

In order to calculate the mask profile for profile coating, one needs to know how the sputtered atoms are distributed on the area above the sputter gun at the substrate level. Film-thickness distribution in magnetron sputtering has been extensively studied over the past few decades.[7, 8] Generally the thickness $t$ of the film deposited from a ring source onto a flat stationary substrate can be expressed as

$$t = m_x h^2 (h^2 + r^2 + a^2)/[\rho\pi (h^2 + r^2 + a^2 + 2ar)^{1.5} (h^2 + r^2 + a^2 - 2ar)^{1.5}], \qquad (1)$$

where $m_x$ is the mass of emitted material on the ring source, $\rho$ is the density of the material, $h$ the source-to-substrate distance, $r$ the radius of the source ring, and $a$ the position on the substrate.[8] The relative thickness distribution is simply $t/t_0$, where $t_0$ is the thickness at $a$=0. The value of $m_x$ can be calculated from a measured erosion depth profile and is directly proportional to the erosion depth at the ring radius $x$. But in $t/t_0$, $m_x$ is cancelled out and $h$ and $r$ are known from the experimental setup and the erosion profile of the target. For simplicity, we take the center ring of the erosion donut on the target to represent the source ring. In the case of Au coatings, we had $h$ = 11.4 cm and $r$ = 2.3 cm. The ratio of $t/t_0$ is thus a fixed function of $a$. In practice, however, there are usually some deviations from measured thickness profiles.

The experimental data for thickness distribution were obtained as follows. First, any existing mask from the shield can was removed. Then a thin film (~40 nm thick or less) was grown on a stationary 4"-diameter Si wafer at a level where the substrate would be coated through the 100 x 152 mm$^2$ aperture on the top of the shield can. The film was coated in a 2.3 mTorr Ar atmosphere, with the power supply of the sputter gun in a constant current mode of 0.5 ampere. The film thickness distribution was obtained by using a M-44 spectroscopic ellipsometer equipped with an automated X-Y translation station.[9] Figure 1 shows such a thickness distribution for a gold film.

Figure 2 compares the normalized experimental result of a gold film to that of the model calculation using Eqn. (1) in one dimension. It was obtained by measuring the thickness every 1 mm apart along the y direction (perpendicular to the moving direction). The agreement between the experimental data and the model calculation is quite good over all. But in detail the experimental data is slightly higher than the model calculation when $a$ is larger than ~3 cm and slightly lower when $a$ is less than ~3 cm. Thus it is not possible to perfectly fit the experimental data even if we adjust $h$ and $r$ in the model. Calculations that consider several weighted rings to better approximate the target erosion area did not change the overall shape of the calculated profile either. We notice that in the model calculation of Eqn. (1), the possible influence of the shield can is not considered. In other words, we have assumed that the sticking coefficient of gold atoms on the shield can is one. In practice, we have noticed that this is not always the case. For example, we have found traces of gold on the back of the substrate. These Au atoms were reflected from a aluminum foil that was loosely covered over the back of the substrate. It is thus possible that some gold atoms would also scatter from the inner wall of the shield can and enhance the thickness on the outer area of the Si wafer. Other evidence is that, when the sputter gun was asymmetrically located in the shield can, the Au thickness distribution was also asymmetrical, with a slightly thicker film on the side that was closer to the wall. We have since relocated the Au target to the center symmetric position. We have also added additional mechanical support to the gun so that it will not wobble around from the center position.

To better fit the experimental data, we simply added an extra term of $f(a)$ in the model. This term is obtained from a polynomial fit of the difference between the experimental curve and that of $t/t_0$ from Eqn. (1). Then $[1+f(a)]$ represents a first-order correction of the model. The final model we used for the relative thickness distribution is $[1+f(a)] * t/t_0$.

Using the above model, one can then obtain the deposition weighting for any position above the aperture on the top of the shield can at the substrate level. The length of the aperture is 100 mm along the moving direction, and the width is 152 mm. When the substrate is moving across the shield can, the film thickness is directly proportional to the length of the opening on the can along the moving direction (with a maximum of 100 mm). By equating the summation of relative weighting to the required relative thickness at the same position, the length of the opening at that position can be determined. By repeating the same process for the whole length of a required profile, a contour can be obtained for a desired thickness profile. For example, the thickness profile for a uniform coating is a straight horizontal line in a thickness vs. position plot. The mask profile is then determined by choosing a length of 100 mm (or less) at positions of ±76 mm, calculating the total weighting at this position, and calculating the length needed at other positions in order to have the same total weighting as that at the 76 mm position. To simplify matters, symmetry is used in the calculations and should be ensured in the experimental setup. Figure 3 shows a mask for uniform Au coating on top of a shield can over a gold target. The masks are cut from aluminum plates. Figure 4 shows the result of a test run measured using the ellipsometer. Good uniformity within ±0.15% is achieved.

## III. PROFILE COATING OF ELLIPTICAL X-RAY MIRRORS

Major efforts are currently underway world wide to improve x-ray microfocusing, through the use of Kirkpatrick-Baez (KB) mirrors. A KB mirror pair[10] consists of two concave mirrors at glancing angles to the x-ray beam and arranged 90° to each other to successively focus x-rays in the vertical and horizontal directions. For microfocusing, it is essential that the mirrors are elliptical, where a ray in any direction from one focal point in an ellipsoid will be reflected into the other focal point.[11, 12] Spherical mirrors unavoidably introduce aberrations. Monolithic KB mirrors are much easier to use, but the desired elliptical surface profile is usually difficult to fabricate. To overcome this problem, a differential deposition technique on selected areas of a well-polished cylindrical mirror has been previously reported.[13] This technique uses a narrow slit in front of the mirror while varying the power of the sputter gun as the mirror is passing across the slit. The power of the sputter gun for each mirror position is determined according to the thickness requirement calculated from a LTP measurement for that position. To be effective, the programmable ramp of the power supply is limited to its linear range. The slit width cannot be too small because of the diffraction effect between the sputtered atoms. To increase the controllability of the process, the target is kept ~8" away from the mirror surface.[14] This method requires a fine control of the overlap of the neighboring Au coatings and several cycles of measuring and deposition tries. We can achieve the same goal by using the profile-coating technique.

The coating profile for elliptical mirrors is obtained from the difference between the measured slope and that of a perfect ellipse. The LTP measurements were carried out at the APS metrology lab. The mirror substrate is a 40-mm-high, 20-mm-wide and 90-mm-long Si block. It has a spherical surface profile with a radius of ~87 m along the long direction. The LTP sampling period on the surface was 1 mm, with the scan length usually set to start and end 2 mm from the ends of the mirror. The input parameters of the ellipse correspond to the UNI-CAT 34-ID beamline at the APS. These parameters are: 64.5 m for the source-to-mirror distance, 2.6 mrad for the mirror glancing angle, and 130 mm for the mirror-to-focus distance. [15]

Since the mirror angle is adjustable, one may choose a coating profile for minimum gradient at the center of the mirror or for minimum coating thickness. [13] For the previously reported differential deposition, better performance was achieved using the profile where the deposition gradient is minimized at the center of the mirror. For better efficiency, we choose the coating profile where the deposition thickness is minimized. We have chosen gold as the coating material. We found that, although Au initially grows as small islands on a Si substrate, thick Au films are usually smooth, especially when a thin Cr underlayer is first coated on the Si mirror. [16, 17] In our experiments, a thin Cr film of ~5 nm was used as a "glue" layer for better adhesion of the subsequent Au coating. For profile coatings, it is also extremely important to load the mirror at the right position on the substrate holder. A test coating of a maximum Au thickness of ~40 nm on a ~12.5 mm x 100 mm Si strip was performed prior to the mirror coating. Then the film thickness was measured using the ellipsometer and normalized to compare with the required coating profile. Figure 5 shows the result of such a test run.

The test run serves two purposes. One is to check the profile with the required coating profile and pinpoint the maximum and minimum positions for mirror loading. The other is to obtain the scaling parameters for the final mirror coating. To obtain an elliptical profile, it is important to put down the right amount of Au at the right positions. Since ellipsometry can measure only a limited thickness range, the measurement can be done only on thin test samples. Fortunately, the magnetron sputtering process can be controlled to be very stable, and a linear scaling of the sample passing speed and number of passes is sufficient to achieve the right total thickness from the test results. This point of view has been confirmed by measuring a scaled-up, thick Au/Si sample using a TOPO interferometer.[18] A thickness of 715±9 nm was obtained from TOPO measurements. This result is in good agreement with the scaled-up number of 714±8 nm from the result of an ellipsometer measurement on a thin Au/Si sample.

When the test was done, the mirror was carefully mounted on a mirror holder and loaded on the carrier so that the mirror surface is ~0.5 mm above the mask during the deposition. The whole deposition took less than an hour to complete. The coated mirror was then evaluated using the LTP measurements.

Figure 6 shows the LTP result obtained after the coating as compared to an ideal elliptical surface. The desired elliptical shape is achieved with an overall rms slope error of 1.66 μrad from an ideal ellipse. The surface figure is even better than that of the original cylindrical mirror, which had a 2.6 μrad rms slope error from an ideal cylinder. This result means that, by using the profile-coating technique, we can not only convert the slope of a mirror but also improve its figure error. If we ignore the large slope error at the right end, an even better figure value can be obtained.

## IV. SMALL D-SPACING LIMIT AND GRADED MULTILAYERS

Laterally graded multilayers have many potential applications and can be made using the profile-coating technique. [6] Here we demonstrate that it can also be used to study the small d-spacing limit in multilayers. In many applications, such as x-ray fluorescence detection and large-incident-angle x-ray monochromators, it is desirable to have multilayers with a small d spacing to decrease the absorption of x-rays in the multilayer and to increase the Bragg angle. The resolution of multilayers also depends on the total number of layers that are effectively involved in x-ray reflection. Thus multilayers with a smaller d-spacing can provide better resolution and higher incident angle compared with those with larger d-spacings. However, fabricating small d-spacing multilayers is a challenge. As d decreases, the interfacial roughness becomes more dominant and the reflectivity decreases. The interfacial roughness is related to the substrate roughness, as well as to interlayer diffusion/chemical reaction at the interface. To search for the best small-d-spacing multilayer system, both the material system and the substrate smoothness need to be explored. Using a laterally graded multilayer is an efficient way to study this topic.

The method of using a wedge-shaped Cr spacer between two Fe layers to study the ferromagnetic-to-antiferromagnetic exchange coupling was very successful and well documented. [19] Although a wedge-shaped sample has a continuously varying thickness, as long as the measurement has a relatively high spatial resolution, reliable thickness-dependent information can still be achieved.

Figure 7 shows the measured d spacing and reflectivity as a function of lateral distance along a W/C graded multilayer. The measurements were done at 6.5 keV on the Bio-CAT undulator x-ray beamline at the APS. The multilayer consists of 60 bilayers of uniform W layer (~1.0 nm thick) and wedge-shaped C layer grown on an ordered 100 mm x 100 mm x 2 mm Si substrate. A linear gradient of d spacing from 3.5 to 7 nm over 85 mm range was designed. The undulator x-ray beam is narrow enough (<0.1 mm) to give a high spatial resolution for the measurement. The substrate had a 0.7 nm rms roughness as determined by a TOPO interferometer. [18] A drop in reflectivity at d=~3 nm is clearly seen. A similar W/C multilayer system grown on a smoother Si substrate (~0.3 nm rms) showed a higher reflectivity of 85-89% with d varying from 3.5 to 6.0 nm. [20] The W layer thickness is also a contributing factor. It has a lower limit as well. Recently we have made graded W/C multilayer samples with different W thicknesses on a smoother Si substrates (better than 0.2 nm rms). These samples will be studied when the next run of undulator x-ray beam is available. By using graded multilayers, we are able to check the substrate quality quickly and to determine the right W thickness to use.

## V. SUMMARY

We have demonstrated that a new profile-coating technique can be used to convert an ordinary cylindrical mirror into a x-ray quality elliptical mirror, to grow uniform thin films and laterally graded multilayers, and to study small d-spacing multilayer systems. Details of the profile-coating technique have been outlined in this paper. The excellent results obtained demonstrate that this technique is very promising for exploring the limits of achievable focus in x-ray optics and the achievable small d-spacing multilayers.

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**FIGURE CAPTIONS**

Fig. 1 Thickness distribution obtained from ellipsometer measurements of a gold film on a Si wafer placed directly above the Au target. The units are angstrom for the vertical axis and cm for the horizontal axes.

Fig. 2 Normalized thickness distribution from ellipsometry measurements (circles) of a gold film compared with that calculated from Eqn. (1) (solid line) as a function of position *a*. The distance from the target to the substrate level is 114 mm, and the radius of the center erosion profile on the target is 23 mm.

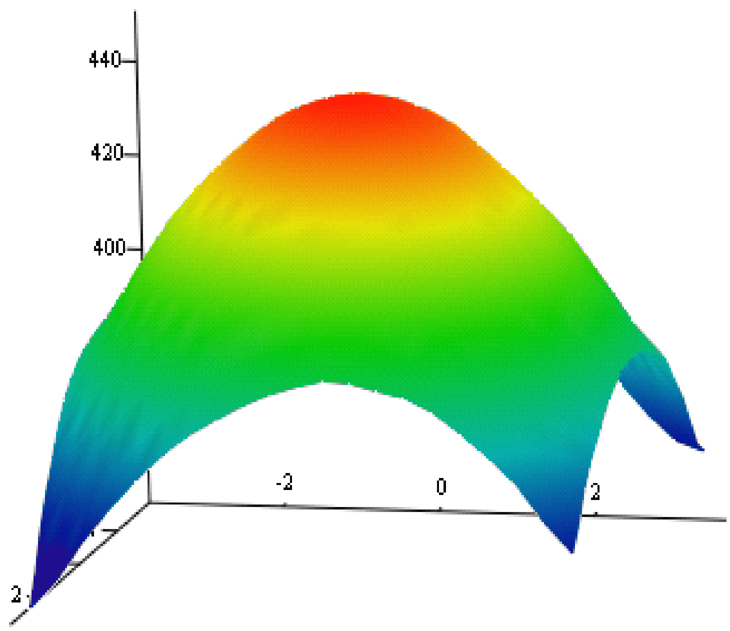
Fig. 3 A mask placed on top of the shield can above the Au target to achieve a uniform coating.

Fig. 4 Measured thickness profile for a gold thin film obtained by using the mask shown in Fig. 3.

Fig. 5 Normalized required and measured thickness profiles for a test run of profile coating of elliptical mirrors.

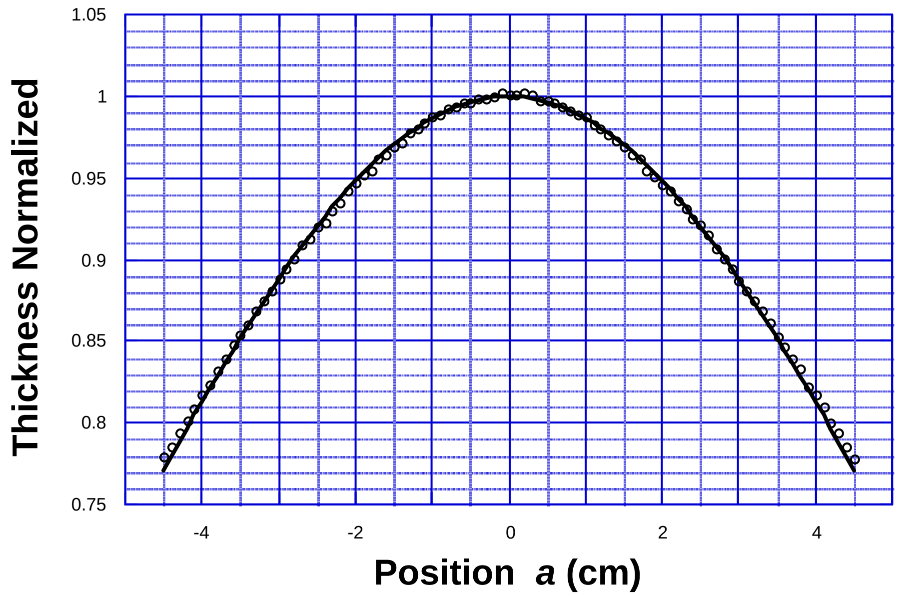
Fig. 6 A typical LTP measurement result showing: 1) at the bottom, the measured slope as compared with an ideal slope, and 2) on the top, the residual slope error as well as the rms number and the projected spot size of a focused beam. This figure shows the result for a mirror after one profile coating. The solid line on the bottom half is the slope of an ideal ellipse. The top shows the difference between the measured slope and the ideal slope, or the residual slope error. The large slope error on the right side is due to the edge effect of the mask.

Fig. 7 Measured d spacing and reflectivity as a function of position along a W/C graded multilayer of 60 bilayers of uniform W layers and wedge-shaped C layers. The measurements were done at 6.5 keV on the Bio-CAT undulator x-ray beamline at the APS. A desired linear gradient of d spacing from 3.5 to 7 nm over 85 mm range was achieved.



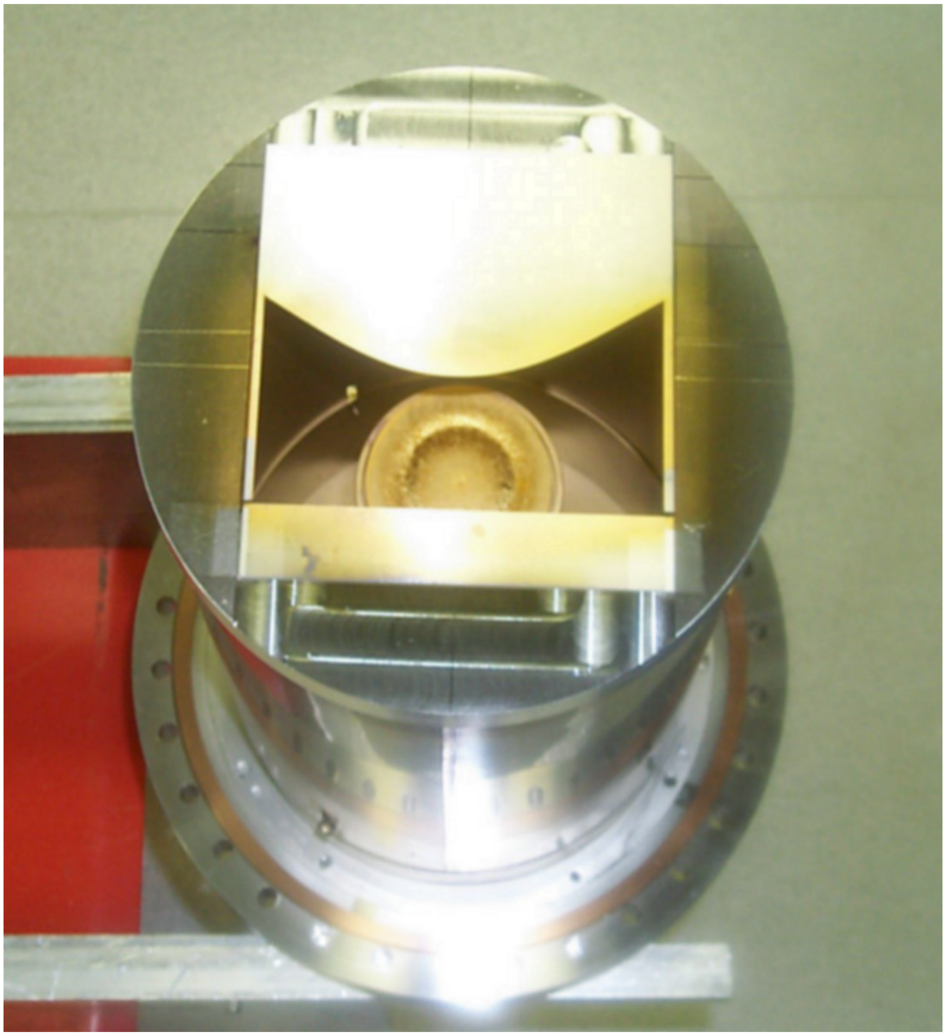

(x, y, t)

**Liu, Figure 1**

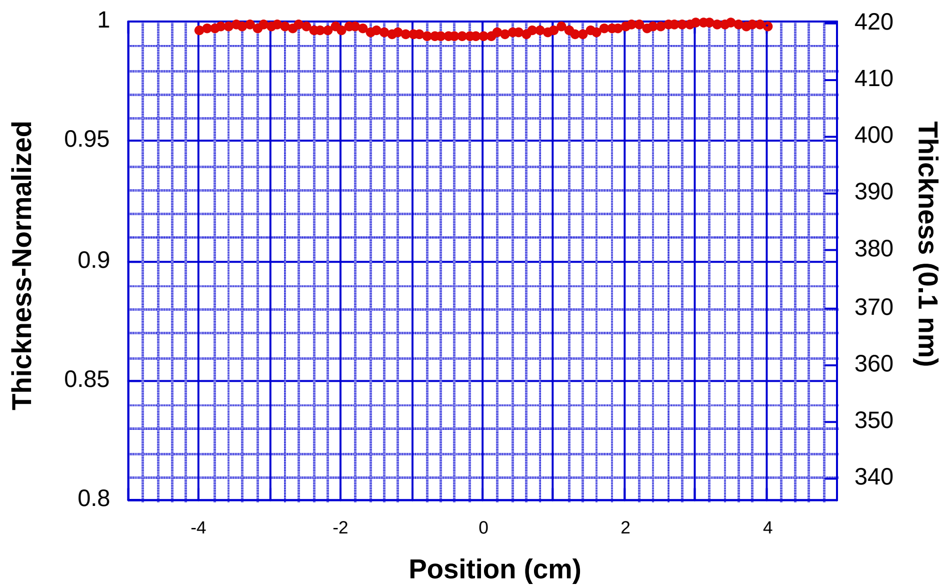


1.05
1
0.95
0.9
0.85
0.8
0.75
Thickness Normalized
-4
-2
0
2
4
Position *a* (cm)


**Liu, Figure 2**

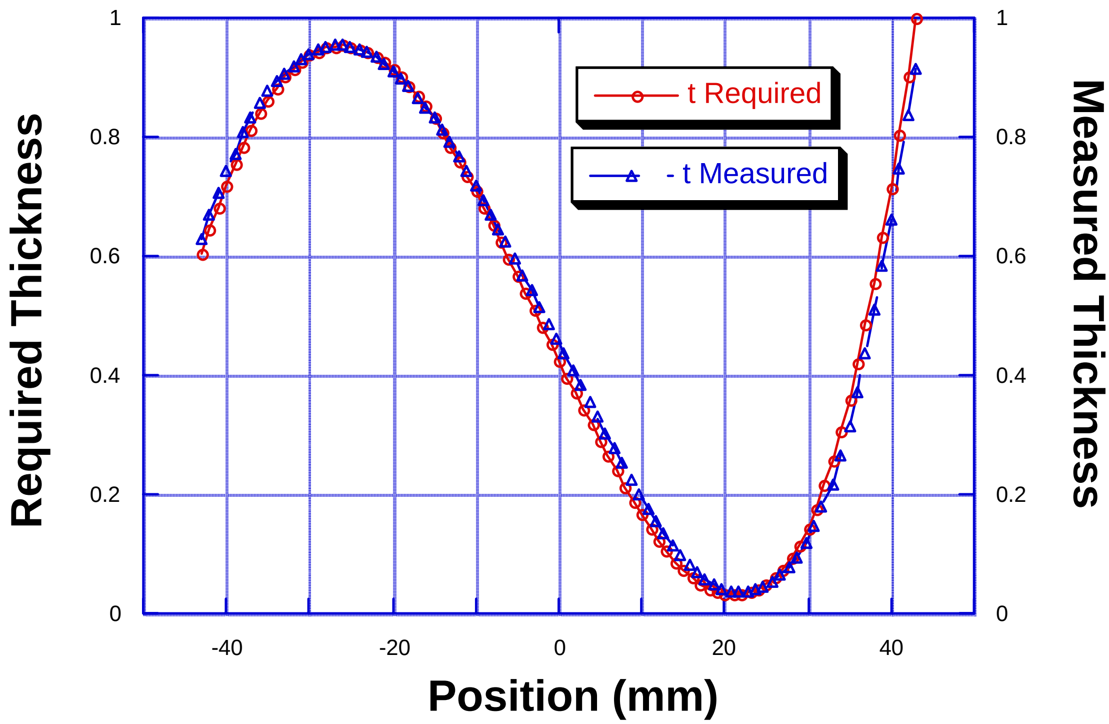


**Liu, Figure 3**



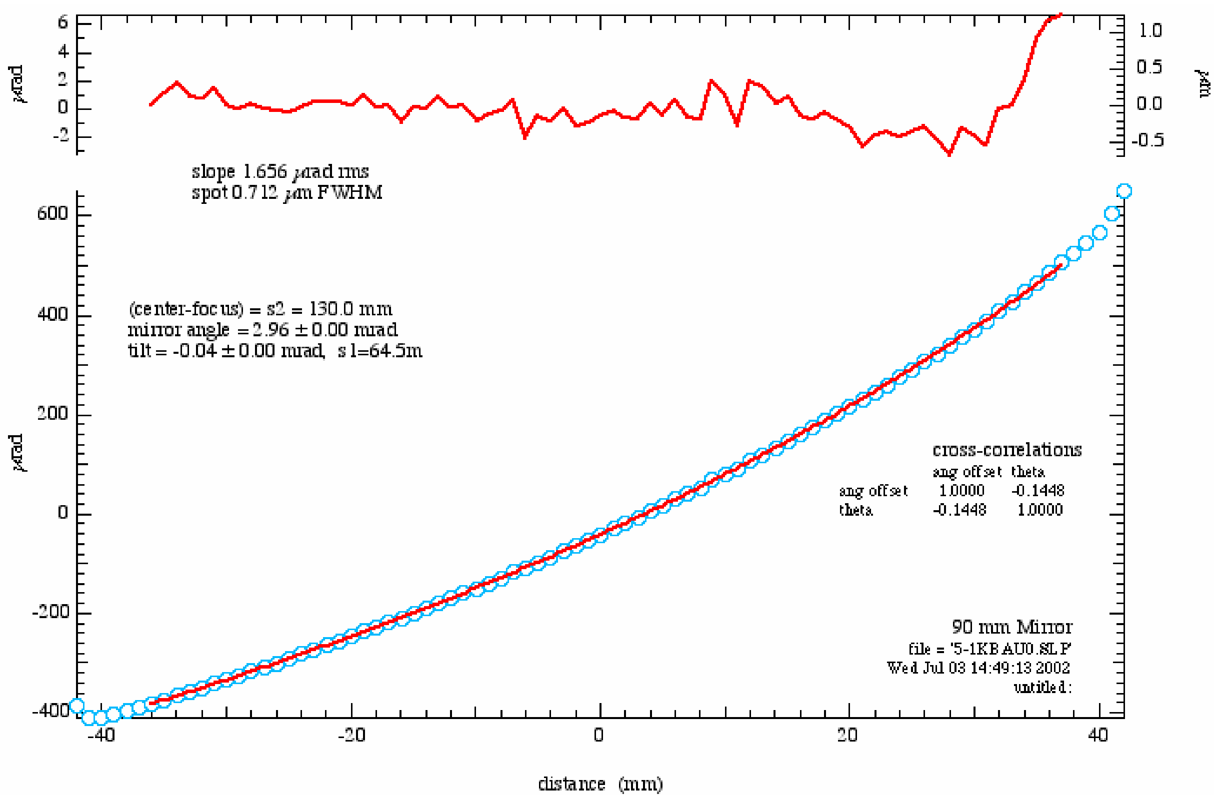
Thickness-Normalized
1
0.95
0.9
0.85
0.8
420
410
400
390
380
370
360
350
340
Thickness (0.1 nm)
-4
-2
0
2
4
Position (cm)


**Liu, Figure 4**



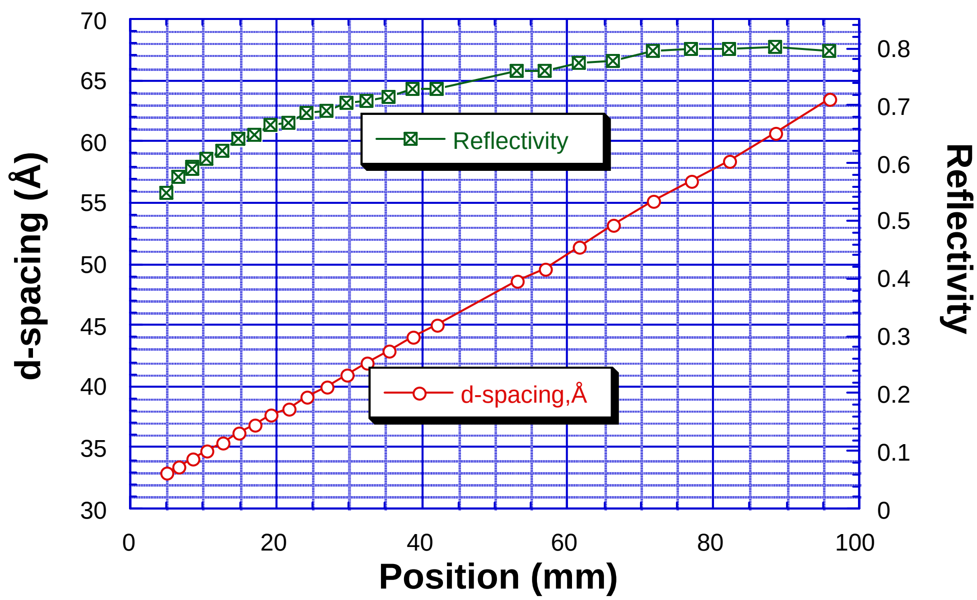
Required Thickness
Measured Thickness
Position (mm)
t Required
- t Measured
1
0.8
0.6
0.4
0.2
0
-40
-20
0
20
40


**Liu, Figure 5**



µrad
µm
slope 1.656 µrad rms
spot 0.712 µm FWHM
(center-focus) = s2 = 130.0 mm
mirror angle = 2.96 ± 0.00 mrad
tilt = -0.04 ± 0.00 mrad, s1=64.5m
µrad
cross-correlations
ang offset theta
ang offset 1.0000 -0.1448
theta -0.1448 1.0000
90 mm Mirror
file = '5-1KBAU0.SLP'
Wed Jul 03 14:49:13 2002
untitled:
distance (mm)


**Liu, Figure 6**



70
65
60
55
50
45
40
35
30
0
20
40
60
80
100
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0
d-spacing (Å)
Position (mm)
Reflectivity
Reflectivity
d-spacing,Å


**Liu, Figure 7**