Advanced multilayer Laue lens fabrication at NSLS-II

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ABSTRACT

In an ongoing effort to advance the state of the art in x-ray nanofocusing optics [1], multilayer Laue lens (MLL) [2,3] fabrication at NSLS-II has matured to include multi-gas reactive sputtering for stress and interfacial roughness reduction, which has recently led to a 70 micron thick single-growth MLL. Reactive sputtering was found to produce WSi_2/Si multilayers with an accumulated film stress significantly lower than Ar-only deposition with identical growth conditions. Significant effort has been focused on the achievement of highly-stable gas mixing and process gas pressure measurement for multilayer growth and the problems faced along with implemented solutions will be discussed in detail. Proper layer thickness and placement throughout the stack presents a major obstacle to the fabrication of high-quality nanofocusing MLLs. Initial metrology of extremely thick MLLs by stitching many scanning electron microscope images was found to be greatly simplified by inclusion of marker labels within the stack.

Keywords: thin film, multilayer, Laue lens, sputter, deposition, x-ray, optics

1. INTRODUCTION

An MLL consists of thousands of layers deposited using a depth gradient according to the zone-plate law, and are fabricated by thin film growth onto a flat substrate; most commonly, small silicon blanks. Sectioning and illumination through the stack in transmission mode allows for diffraction of the incoming radiation into a focus. Several practical methods exist for deposition of extremely thick multilayers; the work described herein utilizes sputtering. MLL fabrication has also been demonstrated utilizing pulsed laser deposition[4].

The NSLS-II deposition system[5] was designed specifically to tackle challenges inherent in the growth of MLLs, which may require combined lateral and depth gradients as well as accurate placement of the layers over tens or hundreds of micrometers of coating. While sputter deposition presents many advantages for fabrication of such optical elements, such as high frequency (layer-by-layer) growth rate stability, increased neutral energy and plasma bombardment during growth, and straightforward target production, the main disadvantage in using conventional, planar cathodes is that the targets erode over time, which produces a long-term growth rate decay that is significant for thick MLLs. The NSLS-II deposition system includes multiple cathodes which are activated in sequential order, and layer by layer, in order to spread the target erosion effect out over several sources and reduce the chance of plasma perturbation effects. Originally, two cathodes, one for each material (in this case WSi_2 and Si) were used. At present, four cathodes are used, with the ability to include four more as the total growth thickness increases. As the growth thickness has increased, an upper boundary of ~45µm due to film stress was reached where growth above this limit resulted in bulk growth defects, multilayer disintegration, and substrate cleaving. Reactive sputtering with a partial nitrogen atmosphere was found to greatly reduce accumulated film stress, and an MLL with total thickness of 70µm was grown without incident. While this is a significant breakthrough in terms of overall growth thickness, the growth rate of these reactively sputtered films is highly dependent on gas pressure and species concentration. Data suggests that this pressure instability may be caused by a pressure gauge drift due to ambient temperature fluctuation, which went unnoticed during multilayer depositions involving only Ar as the process gas. In order to reduce, or eliminate this instability, an isolation enclosure was designed and constructed around the gauge.

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2. STRESS REDUCTION BY REACTIVE SPUTTERING

Accumulated film stress within a sputter-deposited multilayer is generally highly compressive when grown at low pressures, which gradually inverts to stress-neutral and then tensile with high working gas pressures. Unfortunately, reflective multilayers grown at high pressures tend to exhibit poor optical performance due to interfacial roughness. The same adverse effect holds true for MLL optics; interfacial roughness must be kept to roughly 1/3 the zone width or below in order to keep efficiency from being negatively affected. For relatively thin multilayer systems such as are commonly used for reflective optics, compressive stress can cause optical surface figure alterations or delamination, however these issues are greatly magnified when multilayers many 10's of microns thick are produced. It was found previously (ref Headrick, et. al.) that at a pressure of about 6mTorr, WSi_2/Si multilayers sputtered with Ar exhibit an exaggerated increase in roughness. The accumulated film stress for multilayers grown at 6 mTorr is only slightly less compressive than those grown at much lower pressures, and a stress-neutral state is not achieved until working gas pressures are raised above 10 mTorr[6]. *In-situ* film stress measurements using a laser-based curvature monitor[7] were taken for several different MLL depositions and the compressive film stress ranges from ~770MPa for normal, Ar-only low pressure growths, to almost no compressive stress (20MPa) for a multilayer grown at 16mTorr. The data are shown in Fig. 1. However, scanning electron microscope (SEM) data clearly shows severe waviness and roughness within the layers. The incorporation of nitrogen during multilayer deposition has been shown to reduce stress and interfacial roughness in other material systems, such as Cr/Sc[8] and W/B_4C[9]. Following these efforts, we have found that the incorporation of nitrogen during the growth of WSi_2/Si multilayers also reduces stress, and there appears to be no added effect or, perhaps, even a reduction of interfacial roughness.

Structure Outmost zone/total thickness	Conditions	Stress (MPa)
10nm, 12.4μm tilted/partial	2.3mTorr 100% Ar	-742
5nm, 12.35μm tilted/partial	2.3mTorr 100% Ar	-917
4nm, 40μm flat/full	2.3mTorr 100% Ar	-651
3nm, 7μm wedged/full	16mTorr 100% Ar	-20.8
100nm, 6μm ML	4mTorr 95%Ar, 5% N2	-151

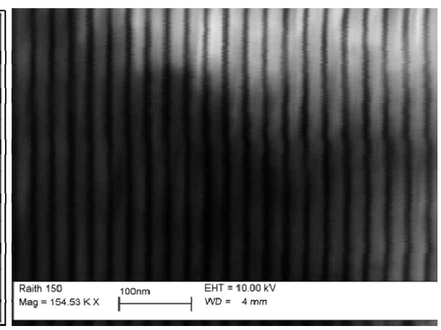

Figure 1. Accumulated film stress vs. structure, and evidence of waviness due to high pressure growth.

Deposition rates for sputtered materials are dependent on a large number of environmental parameters, including source-to-substrate distance, cathode power, aperture openings, process gas pressure, and gas species. In order to calibrate the growth rate of the WSi2 and Si materials according to nitrogen gas concentration, a series of multilayers were deposited whereby identical conditions are held for all environmental parameters except gas concentration as illustrated in Fig. 2.

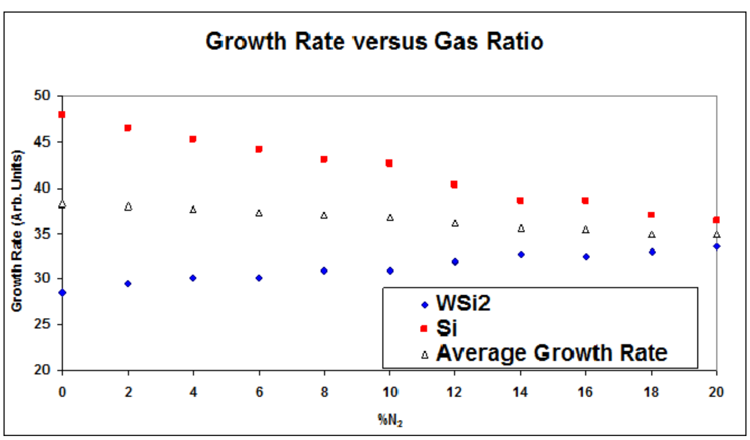

Figure 2. Growth rate of WSi_2 and Si for concentrations of nitrogen from 0% to 20%.

The deposition conditions used were 170 watts of cathode power, a source to substrate distance of approximately 90mm, and a working gas pressure of 4mTorr. Ten sets of multilayers were grown, with each set consisting of two separate depositions of 20 bi-layers each. The layer thickness of the Si is held constant, and the WSi_2 is doubled for one of the two multilayers in the pair. Reflectivity fitting of d-spacing with this layer thickness relationship allows for straightforward evaluation of individual layer growth rates. One interesting behavior to note is that while growth rates for the individual materials are sensitive to nitrogen concentration, the overall growth rate change is minimal, so the total deposition time for thick MLLs (where gamma is 0.5) is not adversely affected.

A 50mm diameter, 270µm thick silicon wafer was deposited with a periodic multilayer of WSi_2/Si where each layer is approximately 1nm, and curvature was measured following deposition of each layer as depicted in Fig. 3. Three different concentrations of nitrogen were used; 9%, 4%, and 0%. Following each multilayer growth, a 10 minute waiting period commenced in order for the next gas concentration to stabilize within the chamber. This waiting period is seen as data with no change in curvature. Although the data contain a high level of noise, it is clear that curvature increases more for the Ar-only growth when compared with the growths containing nitrogen. The steep increases for the first few data points at each multilayer section are likely attributed to thermal effects due to the beginning of plasma ignition.

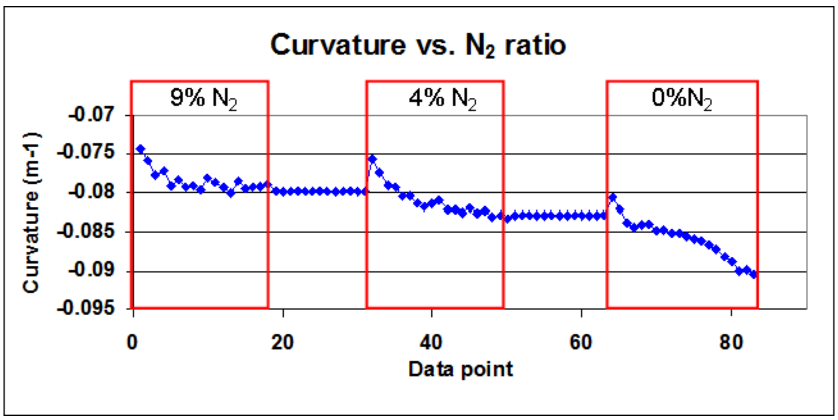

Figure 3. Curvature data vs. nitrogen concentration.

3. REACTIVE GAS PRESSURE STABILITY

The NSLS-II deposition system utilizes four cryopumps which are throttled during deposition to reduce pumping speed, and a two-stage process gas mixing and delivery system. The compressed gasses (Ar and N_2) are metered into a single pipe by two mass flow controllers (MFCs), where a PID feedback system maintains the pressure in the pipe at a constant 850 Torr, and the relative flow rate of each MFC dictates the gas concentration ratio. This gas supplies 9 MFCs, one for each cathode, that feed directly into the cathode bodies around the dark space shields[10]. The process gas pressure is held at a constant value within the chamber (for these experiments, 4 mTorr) by flow rate adjustment through the MFCs by another PID control system. The process gas pressure is measured by a precision capacitance manometer; namely, an MKS 627 Baratron, which incorporates a heating blanket to maintain the gauge at a constant temperature somewhat elevated above ambient. After the commissioning period was completed, multilayers grown with Ar only were universally found to exhibit high quality, low interface roughness, and sharp Bragg peaks consistent with the designed layer thickness values. Subsequent testing with reactive sputtering initially yielded sometimes unreliable results, as illustrated in Fig. 4.

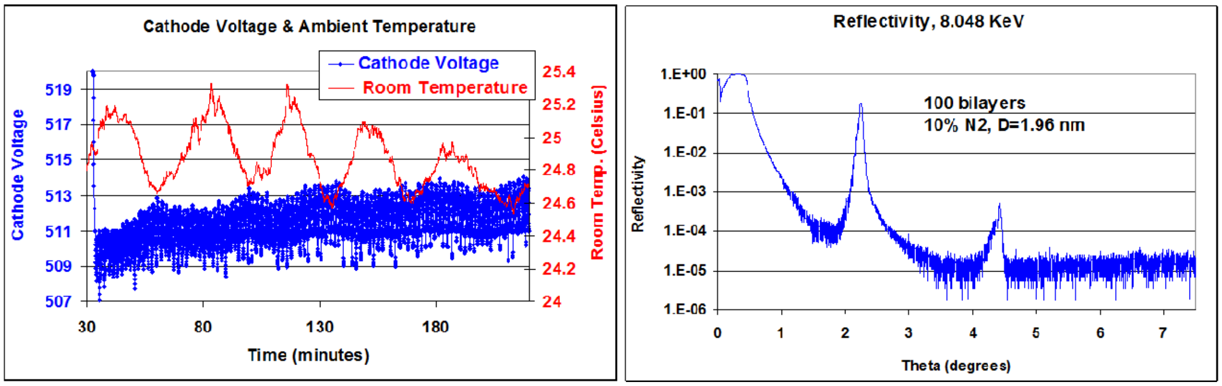

Figure 4. Applied cathode voltage fluctuations due to room temperature swings of ~0.5° C before addition of the gauge isolation enclosure, and the resulting x-ray reflectivity with clear evidence of growth rate instability.

Review of several growth data logs showed a slight voltage fluctuation on the cathode power supplies which appeared to track the ambient room temperature, with a lag of several minutes. Initially, the cathode power supply was suspected of being sensitive to temperature. By observing two capacitance manometer (CM) pressure gauges (both MKS 627) in the system, one the system control gauge, and a second as a witness gauge at a different location in the machine, a minor pressure instability was identified which also tracked the ambient temperature. Additionally, there appeared to be a significant amount of noise from the pressure readings due to vibration. An enclosure[11] for the control CM gauge was designed which consisted of a double-walled acrylic box with outer dimensions of 8 inches by 13 inches by 12 inches, and inner dimension of 10 inch by 5.75 inches by 10.25 inches, pictured below in Fig. 5.

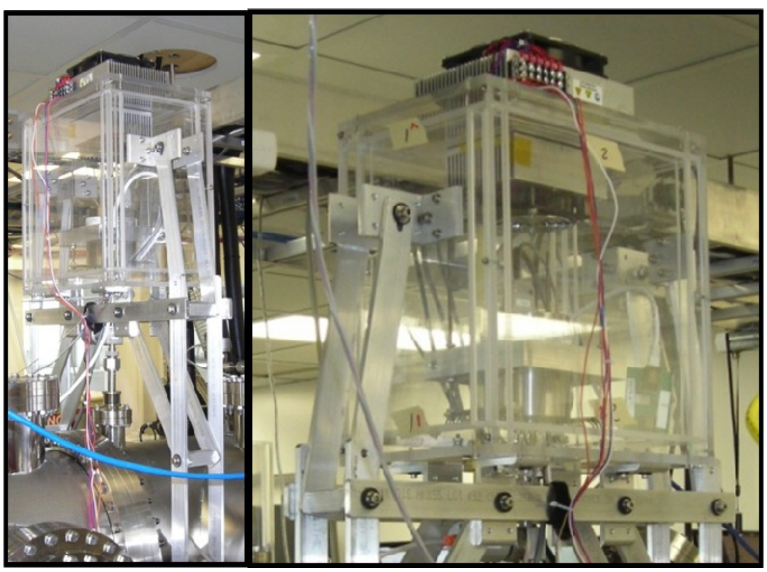

Figure 5. Double-walled acrylic temperature and vibration isolation enclosure for the capacitance manometer

In order to actively regulate the temperature within the enclosure, a thermoelectric heater was installed at the top, to which the power level is actively controlled by a feedback system which relies on a Pt RTD that is placed within the enclosure. In order to reduce vibration, a small edge-welded bellows connects the CM gauge to the vacuum chamber, and the gauge is held in place by four wire-wound vibration isolators. The temperature and pressure response of the capacitance manometer after the isolation system had been attached showed a marked improvement when compared to measurements taken prior to the isolation system installation. In terms of vibration, the noise in the measurements of pressure taken from the CM decreased by a factor of 2 to 3. In terms of temperature isolation, the enclosure maintains a set point temperature within a tenth of a degree Celsius (the resolution of the selected thermoelectric controller), which appears to produce satisfactory results as illustrated in Fig. 6. Multilayers produced with this system exhibit very sharp

Bragg peaks with no discernable instabilities, even with the room temperature fluctuating by almost 4 degrees Celsius. Recently, the deposition lab environmental control system was upgraded with multiple RTDs for temperature control and tuned to eliminate such large room temperature variations, and now consistently maintains the temperature within the lab to less than +/- 0.5 degrees Celsius.

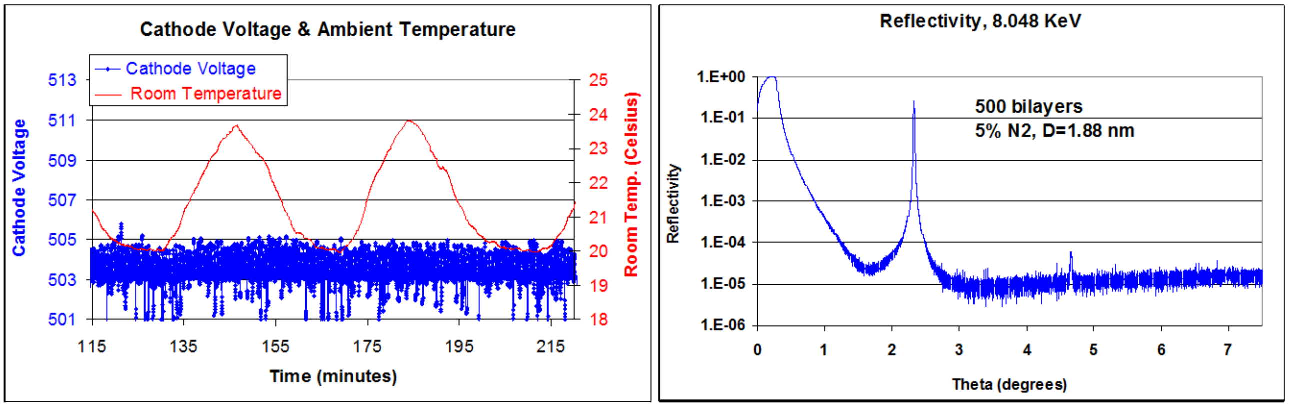

Figure 6. Applied cathode voltage is highly stable, even with room temperature swings of ~4° C after installation of the gauge isolation enclosure, and the resulting x-ray reflectivity with clear evidence of high growth rate stability.

4. MARKER LAYERS

Sputter deposition systems which utilize fixed, solid sources are very stable and repeatable during growth of relatively thin, reflective multilayers, however target erosion during growth of very thick MLL structures are effected by growth rate decay over time, which is accounted for by gradual changes in dwell time (exposure time). Conventional *in-situ* growth rate monitoring such as quartz crystal rate monitors (and many other types of instruments) provide neither sufficient accuracy nor stability to adequately measure growth rate; so, instead, deposition systems are designed to be as stable and reproducible as possible. Growth rate decay is monitored *ex-situ* by first sectioning, and then obtaining images of the MLL with a scanning electron microscope (SEM) and using the line profile data to iteratively modify the growth rate decay compensation table. The first few MLLs[12] were grown with a limited layer count and relatively small total deposition thickness such that a single SEM image provided enough pixel resolution accurately determine individual layer position. The smallest layer in the stack had layer thicknesses that were not difficult to image clearly while simultaneously keeping the field of view large enough to image the entire structure. As MLL deposition gradually increased in thickness, multiple sub-aperture images were required in order to provide adequate spatial resolution. A 13-micron thick MLL with 5nm outermost zones required just two sub-aperture images. A more recent 43-micron thick MLL series comprised of 4nm outermost zones and 6,510 layers analyzed in this way would have needed 45 sub-aperture images.

Marker layers[13] have been implemented within the MLL growth whereby the positions normally occupied by an invididual layer, or grouping of layers, are intentionally "consumed" by one material. After the marker layer is grown, normal MLL deposition continues unaffected. As the goal of the marker layer is to provide an artificially thick layer within the stack without disturbing the normal position of subsequent layers, the marker layer thickness is the summation of an odd number of layers. By imaging only these marker layers within the MLL, the sub-aperture stitching overlap error is eliminated as the gap between marker layers can be several hundred nanometers or more, which is far greater than the stage uncertainty in any modern SEM. As the marker layer thickness will be a multiple of the nearby layer thickness, lower magnifications are used to image the MLL. Several growths of the 43µm thick MLL structure were performed utilizing marker layers for layer placement metrology. As illustrated in Fig. 7, a SEM image reveals 12 marker layers within the stack which are each spaced by 512 MLL layers. The Moire pattern from the individual layers is clearly visible in regions seven and nine, and magnification of the upper region in the subset highlights the contrast between the marker layers and the rest of the structure. By using only the marker layers for MLL layer placement metrology, a set of three growths were able to produce successively higher quality lenses, with a final layer placement of approximately +/- 19nm.

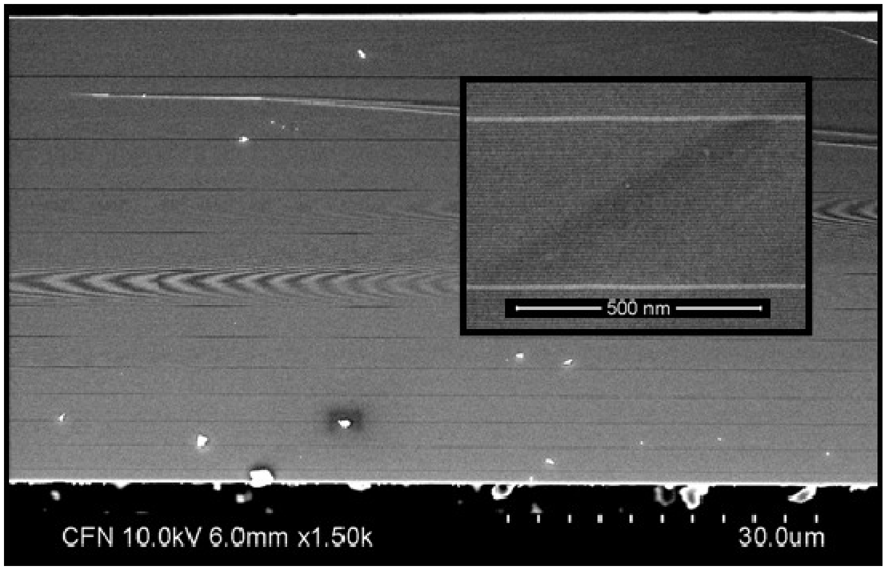

Figure 7. SEM image of a 43µm thick MLL with 12 marker layers inserted for layer placement metrology. The offset streak near the top is not a growth defect, but is due to polishing scratches.

5. CONCLUSIONS

While designed from the beginning for the highest level of stability possible, the NSLS-II multilayer Laue lens deposition system exhibited growth rate instability during reactive sputtering with nitrogen. This instability was eliminated with the design and installation of a highly stable temperature and vibration isolation enclosure for a precision capacitance manometer. Nitrogen reactive sputtering has demonstrated a significant decrease in accumulated film stress for MLL optics deposition, and it is anticipated that this technique will allow for MLL optics of greater than 100µm in the near future. Layer placement accuracy has consistently been a serious issue for the fabrication of diffraction-limited MLL, and the use of marker layers has greatly simplified this problem by providing an easy to implement solution.

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