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# X-ray reflectivity and mechanical stress in W/Si multilayers deposited on thin substrates of glass, epoxy replicated aluminum foil, and Si wafer.

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## ABSTRACT

Reflectivity at $\lambda$=0.154 nm and mechanical stress in the bulk thin films of tungsten and silicon and single d-spacing multilayers on their basis with d≈2.8nm deposited by the magnetron sputtering technique on flat thin substrates of Si wafer (~0.2mm), glass (~0.3mm), and epoxy gold replicated aluminum foil (~0.3mm) have been studied. The interfacial roughness of the multilayers has been calculated from the X-ray reflectivity curves as the following: on Si wafer $\sigma\approx$ 0.31nm, on glass $\sigma\approx$0.32nm, and on foil $\sigma\approx$0.34nm. There was not observed a significant dependence on the stress in the Si film with change in rf power, Ar gas pressure and biasing. For the W films an increase of dc power results in an increase of stress. A similar relationship is also evident for W films deposited by rf power, but this dependence is less pronounced. The influence of low temperature (up to 200 $^0$C) annealing on X-ray reflectivity and stress in the multilayers has been investigated. There was not found an appreciable changes in the absolute value of reflectivity or in d-spacing with annealing temperature. The stress in the coatings changes with annealing temperature from compressive to tensile. There was observed a temperature of annealing at which the stress is no longer present in the film. The absolute value of this temperature measured for W/Si multilayer is approximately 120 $^0$C.

**Keywords:** multilayer structure, X-rays, magnetron sputtering deposition, thin films, mechanical stress, thin substrates, epoxy gold replicated foil, thin glass, thermal annealing, X-ray reflectivity, interfacial roughness.

## 1. INTRODUCTION

Stresses in multilayer X-ray films are important for technological application and can affect thin film properties. Stresses can cause cracking and peeling of the coatings, deformation in flat or previously curved mirrors. The change in geometry can become troublesome when the substrate has a predetermined geometry which is necessary for the use in optics applications, i.e., focusing optics. Further, stress can have a negative affect on the performance of the multilayer by increasing the full width of the reflectivity peak, by reducing angular or spectral resolution and peak intensity.

Our interest in stresses in thin film multilayer structures is caused by the objective to develop X-ray supermirrors on thin (~0.3mm) substrates for use as reflectors in space telescopes in photon energy range from ~10 keV to 100 keV. Astrophysical telescopes require deposition onto thin surfaces which are either curved, formed before deposition of the multilayer, or flat surfaces subsequently bent into shape. This is because optimal optical elements consist of thin highly nested hyperboloid / paraboloid or conical shells. To achieve good performance, substrates must be found which are both smooth, and which have the appropriate mechanical properties to maintain integrity during the deposition process. In addition, the stresses induced on the substrate must be minimized so that distortion of the optic after deposition is reduced to an acceptable level, i.e., the telescope`s angular resolution is not compromised.

In this paper two substrates are considered. They are epoxy gold replicated aluminum foil (ERF) and thin (~0.3mm) DESAG AF45 and D263 glass. We have investigated Cu $K_\alpha$ reflectivity of bulk thin films and single d-spacing W/Si multilayers deposited on flat substrates of these materials and mechanical stress introduced in the multilayers. To minimize stress we varied the technological parameters of deposition and successfully to reduced stress by one order of magnitude without deterioration in X-ray performance. But stress in the supermirror deposited on glass is still high enough (~0.3 GPa) to result in a figure error of several degrees for a ~50 cm long mirror. To develop a technological procedure which would allow us to eliminate the mechanical stress in W/Si coatings, the influence of low temperature ( up to 200 °C ) annealing on stress and X-ray performance has been studied.

## 2. EXPERIMENTAL PROCEDURE

The bulk thin films and multilayer structures were deposited by the magnetron sputtering technique with dc- and rf-modes. The deposition parameters such as cathode power, gas pressure, biasing, etc. have been chosen to allow for high X-ray performance and low stress in the coatings. The X-ray reflectivity of the structures was measured by Huber - diffractometer at the wavelength of Cu $K_\alpha$ radiation ( $\lambda$=0.154nm ) with $\theta$-$2\theta$ scheme.

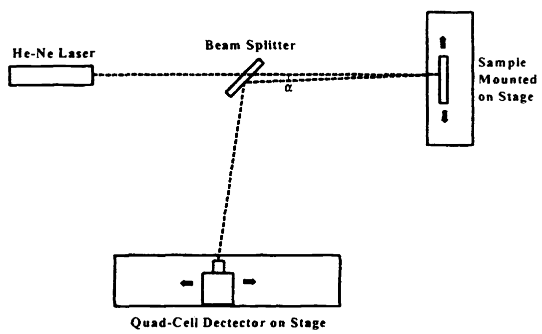
Stress in the coatings was estimated by measuring the change in curvature of the substrate caused by the coatings. For the case in which the total thickness of the films is less than that of the substrate the stress in the film is given by Stoney equation [1]:

$$\Sigma = \frac{E_s t_s^2}{6t(1-\nu_s)}\left(\frac{1}{R}-\frac{1}{R_o}\right) \tag{1}$$

where $E_s$ and $\nu_s$ are the substrate Young's modulus and Poisson ratio, $t_s$ and t are the substrate and film thicknesses, respectively, and $R_0$ and R are the initial and final radius of curvature of the sample. The thickness of the coatings investigated is significantly less than the substrates thickness and thus Stoney's equation can be applied.

The system used to measure the curvature of the samples is shown in Fig. 1. The system can measure the angle between the incident and reflected beam, $\alpha$, with 3 arcsecond resolution. The radius of curvature is determined from the change in the angle, $\alpha$, at intervals along the sample surface. The minimum radius of curvature that can be measured is 0.4 m.

Fig. 1: System for radius of curvature measurements.



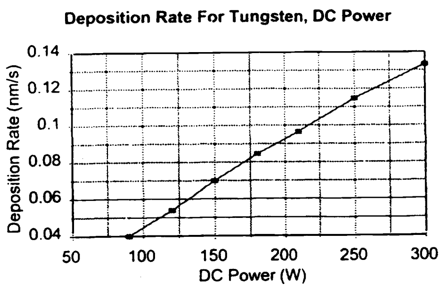

In the curvature measurement each sample was mounted on the sample holder with the use of double sided tape. The tape was in contact with a small fraction of the total surface area far from the region were the measurements were performed. This was done so that any change in curvature by the mounting procedure was minimized. Each sample was measured five times after removing and remounting. A procedure was used to ensure a repeatable mounting position so that the incident beam scanned across the same region of the sample each time. We considered the radius of curvature in one dimension only.

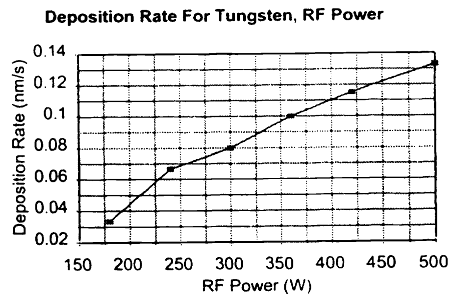
The samples were annealed in a vacuum oven and the temperature of the oven was monitored by the use of a thermocouple. The thermocouple was immersed in a small bath of indium in a container formed from aluminum which was placed on the bottom surface of the vacuum oven. The sample was then placed on an aluminum tray situated approximately 7.5 cm above the surface where the temperature was actually measured. This method of sample arrangement was used in the investigation of the multilayers deposited on Si-wafers. Since this does not allow for accurate measurement of the temperature of the sample we have considered a solution to increasing the certainty of the temperature measurements by incorporating a Wood's alloy with a relatively low melting point of about 70 $^{0}$C. A container of thin stainless steel was formed to make a bath with a large enough surface area for the sample rest on. A thermal couple gauge was also immersed in the bath of the alloy. This allows excellent thermal contact of the sample and an accurate measurement of temperature. The multilayer deposited on glass substrate has been studied using this method of the temperature value reading. We began the process of annealing with a temperature of 100 $^{0}$C and annealed the samples for approximately one hour, increasing the temperature in increments of 25 $^{0}$C after each step in procedure. The samples were then allowed to cool for approximately 30 minutes. After each step in annealing process the radius of curvature of the samples was again measured. The samples were also measured with an X-ray diffractometer at $\lambda$=0.154nm to investigate any possible effect of the annealing process on reflectivity.

## 3. RESULTS

### a) Thin bulk W and Si films.

We did deposition of 30 nm thick W and 50 nm and 100 nm thick Si bulk films on Si wafers using dc- and rf-modes for tungsten coating and rf-mode for silicon. The stress dependance on power applied to the targets has been studied. To keep the coating thickness constant we varied the deposition time. Dependance of the deposition rate on power applied is presented in Fig. 2.



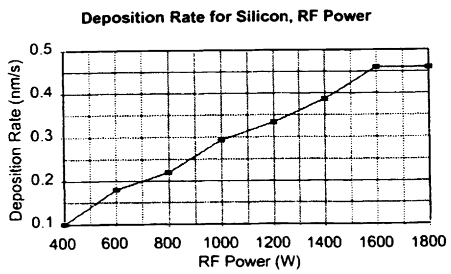
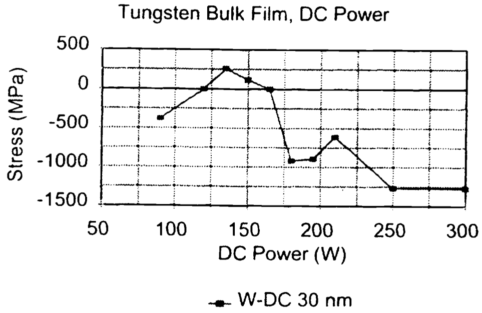
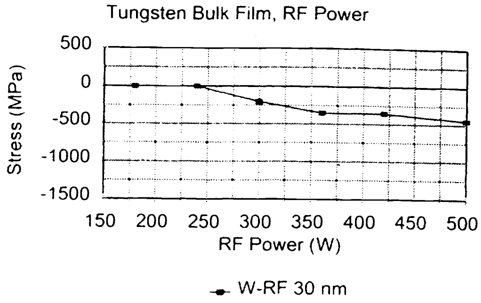




Fig 2 . Deposition Rate for Tungsten, both DC and RF Power, and for Silicon , RF Power.

Thickness of the coating was calculated from peak-valley angle position of the interference pattern at low angle diffraction by use the X-ray diffractometer. Results on stress in the films versus power setting are presented in Fig. 3. There was not a significant dependance on the stress in the Si films with changes in rf-power. For the W films, however, an increase of dc power resulted in an increase of film stress. A similar relationship is also evident for W films deposited by rf-power, but this dependence is less pronounced. Both Si and W films have a compressive stress. We did not observe stress reduction in the films by varying Ar-gas pressure from 1.5mTorr to 5mTorr and substrates biasing from 0V to -100V.



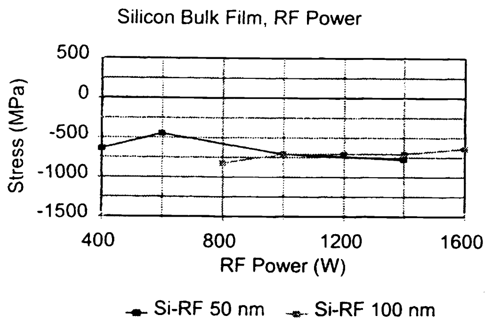




Fig. 3. Stress in a bulk film on a Si wafer versus power setting

## b) W/Si single d-spacing multilayers

We did deposition of single d-spacing W/Si multilayers with number of period N=100 on thin glass, epoxy gold replicated foil (ERF), and Si-wafer. All of these structures have been studied by X-ray diffractometer. Reflectivity of the multilayers is presented in Table 1. The values of d-spacing, $\gamma$, and $\sigma$ have been calculated from angle position and intensity of Bragg peaks in low angle reflectivity curve in assumption of sharp interfacial boundaries, bulk material density of the layers, and influence of roughness is described by Debye-Waller factor [2]. The multilayer deposited on Si-wafer shows the best performance, next is multilayer deposited on glass substrate, and the last one is on foils. The difference in the X-ray reflectivity for the multilayers considered is not so big and influence of initial substrate roughness is reduced with thickness of the layers.

Table 1. Reflectivity of W/Si multilayers deposited on different substrates at $\lambda$=0.154nm.

| Substrate | d-spacing, nm | $\gamma=h_w/d$ | Integral reflectivity at $\lambda$=0.154nm Ri=Rp·$\Delta\theta$, deg. | | | Interfacial roughness, $\sigma$ nm |
|---|---|---|---|---|---|---|
| | | | 1[st] order | 2[nd] order | 3[d] order | |
| Glass | 2.66 | .535 | 1.97E-2 | 4.7E-5 | 4.7E-6 | 0.32 |
| Foil | 2.63 | .53 | 1.50E-2 | 3.4E-5 | 2.6E-6 | 0.34 |
| Si-wafer | 2.63 | .545 | 1.88E-2 | 9.5E-5 | 9.2E-6 | 0.31 |

$h_w$ is the thickness of tungsten layer.

For the experiments on the influence of low temperature annealing on stress in the W/Si coatings, two Si-wafer and one glass substrates with different initial radii of curvature have been used. Si-wafers were initially convex and glass substrate was initially concave. The multilayers with d-spacing of ~2.8 nm, number of periods N=200, and $\gamma$~0.3 were deposited on these substrates. The annealing and the stress measurement procedure have been carried out as described previously. The deviation of the reflective angle when scanning the laser beam along the surface of the multilayer deposited on Si-wafer versus the temperature of annealing is presented in Fig. 4.

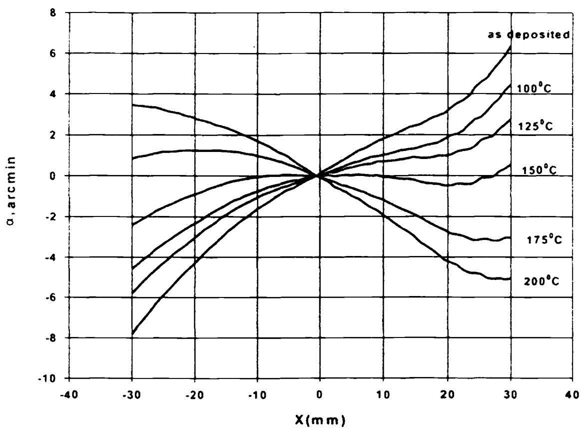



Fig. 4. Reflective angle deviation when scanning the laser beam along the surface of the sample XRO#12923-6 as versus of annealing temperature. X is coordinate of illuminated area on the sample surface.

In Fig. 4, a positive slope is convex, and a negative slope is concave. We see with increasing the temperature the shape of the sample changes from convex to concave. At the temperature of ~150 °C the reflective angle deviation became similar to that for un-coated sample.

Values of radii of curvature and X-ray performance of the multilayers tested are presented in Table 2. From the table we see that there is no appreciable changes in the absolute value of reflectivity or in d-spacing with annealing temperature.

Table 2. Radius of curvature and reflectivity at $\lambda$=0.154nm vs temperature of annealing for W/Si structures. Samples #12923-6,7 were deposited on 0.2mm thick Si-wafer and sample # 13097-2 on 0.3mm thick glass.

| T, °C | XRO # 12923-6 Si wafer | | | XRO# 12923-7 Si wafer | | | XRO# 13097-2 Glass | | |
|---|---|---|---|---|---|---|---|---|---|
| | r, m | $R_i$,deg. | d,nm | r, m | $R_i$,deg. | d,nm | r, m | $R_i$,deg. | d,nm |
| Substrate | -374.6∓10.6 | - | - | -25.84∓2.0 | - | - | 27.5±0.5 | - | - |
| As deposited | -23.0∓1.2 | 0.216 | 2.83 | -10.8∓0.1 | 0.021 | 2.83 | 30.6±0.7 | 0.020 | 2.83 |
| 100 | -40.3∓1.0 | 0.0216 | 2.83 | -13.1∓0.1 | 0.0208 | 2.83 | 28.9±0.2 | 0.020 | 2.83 |
| 125 | -61.5∓1.8 | 0.0219 | 2.83 | -14.7∓0.2 | 0.0210 | 2.83 | 26.9±0.2 | 0.021 | 2.83 |
| 150 | 508.0∓12.2 | 0.0220 | 2.82 | -25.5∓1.3 | 0.0207 | 2.84 | 25.9±0.1 | 0.020 | 2.83 |
| 175 | 49.0∓2.9 | 0.0208 | 2.84 | -77.8∓1.1 | 0.0209 | 2.83 | 23.0±0.1 | 0.020 | 2.83 |
| 200 | 21.5∓0.3 | 0.022 | 2.83 | -137.5∓6.7 | 0.0229 | 2.80 | 21.5±0.1 | 0.020 | 2.83 |

$R_I = R_p \cdot \Delta\theta$ is integral reflectivity, where $R_p$ and $\Delta\theta$ are peak reflectivity and FWHM of the reflectivity curve respectively.

The data of curvature radii presented in Tables 2 have been used in calculation of absolute values of stress in the coatings using Stoney equation (1). The stress dependance on annealing temperature is presented in Fig. 5. These graphs indicate that the stress in W/Si multilayer coatings change with annealing temperature from compressive to tensile. There is a temperature of annealing, which we denote as $T_c$, to be the temperature at which the stress is no longer present in the film. Values of $T_c$

observed for the multilayers deposited on 2 Si-wafers with different initial shape are close to each other and differ from $T_c$ for glass sample. The difference in values of $T_c$ and stress, especially at higher temperatures, for the samples on Si-wafers and glass is caused by the difference in the method of temperature value reading. As we described previously in annealing procedure used for the structure deposited on glass the sample was placed on the surface of liquid Wood's alloy were the thermocouple gauge was immersed. In experiments with the multilayers deposited on Si-wafers the samples were situated approximately 75 mm above the surface were the temperature was actually measured. We measured simultaneously the temperature reading in both cases and found that the difference is more than ten degrees and increase with the temperature.

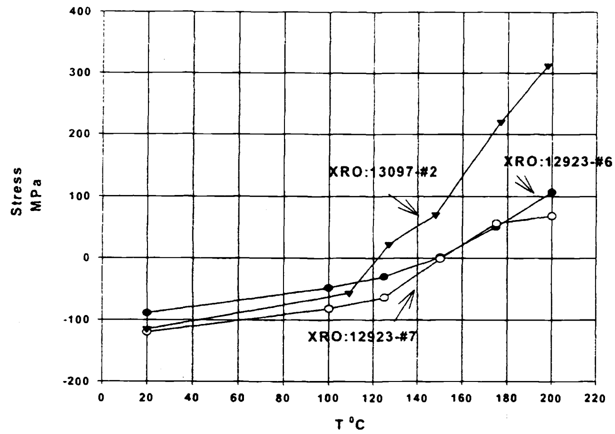



Fig. 5. Stress in W/Si multilayer with d=2.83 nm, $\gamma$=0.3, versus temperature of annealing

## 4. CONCLUSION

Reflectivity at $\lambda$=0.154 nm of W/Si multilayers with d$\approx$2.8 nm deposited by the magnetron sputtering technique on flat thin substrates of Si-wafer ( ~0.2mm ), glass ( ~0.3mm ), and epoxy gold replicated aluminum foil (~0.3mm ) has been studied. The best performance show the structures deposited on Si-wafers, a slightly less reflectivity was observed on glass samples, and lower one on foils. The interfacial roughness for these samples calculated from the X-ray reflectivity curves are the following: on Si-wafer $\sigma\approx$ 0.31nm, on glass $\sigma\approx$ 0.32nm, and on foil $\sigma\approx$ 0.34nm. The difference in the performance of the multilayers on all these substrates is not so big and all of them can be considered as substrates for X-ray supermirrors for photon energy range of up to ~ 100keV.

Stress in 30 nm thick tungsten and 50 nm and 100 nm thick silicon bulk films deposited on Si wafers using dc- and rf- modes for tungsten coating and rf-mode for silicon has been measured. There was not observed a significant dependence on the stress in the Si films with changes in rf power. For the W films an increase of dc power results in an increase of stress. A similar relationship is also evident for W films deposited by rf power, but this dependence is less pronounced.

The influence of low temperature ( up to 200 °C ) annealing on X-ray performance at $\lambda$ = 0.154nm and mechanical stress in the single d-spacing W/Si multilayers deposited on Si wafers and glass substrates has been investigated. There was not found an appreciable changes in the absolute value of reflectivity or in d-spacing with annealing temperature. The stress in the coatings changes with annealing from compressive to tensile. There was observed a temperature of annealing, which we denote as $T_c$ to be the temperature at which the stress is no longer present in the film. Value of $T_c$ observed for the multilayers deposited on substrates with different initial shape are close to each other. The absolute value of $T_c$ measured for W/Si multilayers with d~2.8nm and $\gamma$~ 0.3 is approximately 120 °C.

The result on observation of the annealing temperature $T_c$, at which the stress in W/Si coating is eliminated is the most interesting from the point of view of development of X-ray supermirrors on curved thin substrates. We plan to continue the research taking in consideration flat and curved multilayers with different values of d-spacing, material of layers and, also, depth graded structures-supermirrors.

## 5. ACKNOWLEDGEMENTS

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