T.3: Surface and interface studies on short period multilayer structures

Sanjay Kumar Rai
Synchrotron Utilization Section
E-mail: sanjayrai@rrcat.gov.in

Abstract

X-ray multilayer structure consists of alternating layers of high and low electron density materials. This arrangement of layers is very useful for developing artificial Bragg reflectors for wavelengths in x-ray region at which no natural crystals are available. X-ray multilayers with small periods around ~ 2nm are of great significance. This is because of their widespread use as mirrors, dispersive optical elements, normal incidence reflectors for (soft-) x-ray microscopy. Multilayers are also useful in x-ray astronomy, plasma diagnostics and for experiments with x-ray free electron lasers (XFEL). Worldwide, use of multilayer mirrors at synchrotron beamlines has increased significantly in recent times. Various beamline on these sources find multilayer based x-ray optics quite useful. Using multilayer optics in a beamline enhances the range of experiments with increased flux. Our center has undertaken development of short period multilayer, which can be used as monochromator for hard x-rays on Indus-2 beamline and normal incidence mirrors for soft x-rays on Indus-1/ Indus-2 source.

Usefulness of these structures is governed primarily by the practically achievable interface perfection. Compositionally sharp and topologically smooth interfaces with good optical contrast are required for high reflectivity. Atomic-scale surface or interface roughness causes scattering losses and can significantly lower the reflectivity. Details of optimization process and issues related with the growth of short period sputter deposited W/Si, W/B_4C and NbC /Si multilayer mirrors have been described in the report. Changes in the interface structure on reducing the multilayer period have been studied and reasons for these variations have been explained. Understanding of origin of stable shortest period possible by sputtering deposition has been established.

1.0 Introduction:

Worldwide, use of multilayer mirrors at synchrotron beamlines has increased significantly in recent times. Increased need of short period stable multilayer mirrors has kicked off research in developing newer multilayer combinations for various applications. Present day multilayers require ultra short period structure (<2nm) with large number of layer pairs and long term stability of interface under high heat load. Indian SR sources Indus-1 operating at 450 MeV emits radiation upto VUV/ soft x-ray region, and Indus-2 with 2.5 GeV energy emits radiation up to hard x-ray region. We have undertaken development of short period multilayer, which can be used as monochromator for hard x-rays and normal incidence mirrors for EUV/soft x-rays. Both these requirements need multilayer in periods range from 2-3 nm. At periods below 3nm it becomes difficult to get measured reflectivity close to theoretically possible values due to various issues related with growth. In practice, the structures of most short period multilayers are far from the assumed ideal structures. Diffusion and intermixing of the materials at their interfaces cause imperfection. These imperfections add to interface roughness generated during depositions in the layers. Influence of a 0.4 nm r.m.s. roughness on the reflectivity of the first Bragg peak of multilayers with a period of 4, 3 and 2 nm shows as drop of reflectivity to 91, 82 and 35% from ideal reflectivity with zero roughness. These calculations show that how much important it is to minimize interface roughness at shorter periods.

These issues suggest that a careful investigation of changes in interface structure with multilayer period reduction is necessary for developing short period multilayers of periods ~ 2nm.

1.1 Concern of short period multilayers

Interface roughness results from three main imperfections in the multilayer structures: roughness of the substrate surface that may propagate into the multilayer, variation of the layer thicknesses and imperfections at the interfaces due to growth mechanism used. The first two factors are extrinsic to the formation of the multilayers, while the last factor is an intrinsic parameter, which depends on the materials characteristics and reactions between the constituents of the multilayers during and after deposition. Imperfections at interfaces can be divided into two different categories: one is the composition gradient across the two materials resulting from interdiffusion of the constituents, and the other is structural roughness at the interfaces. Optimization of the multilayer performance thus includes choosing constituent materials that undergo minimum inter-layer diffusion and intermixing and that form smooth and uniform interfaces, choosing appropriate starting substrate and deposition method.

In real structures, it is often unclear which of the interfacial imperfections, compositional gradients or structural roughness, has a greater effect on the performance and characteristics of the multilayers. Studies to separate the effects of the intermixing and the structural roughness components on the reduction of the multilayer reflectivity are important because by understanding which effects limit the reflectance, fabrication of improved performance multilayers may be possible. Attempts to determine these effects have been reported by different groups. Auger depth profiling and cross-sectional TEM analysis have been used to study the concentration gradients across the interfaces, and specular and non-specular x-ray scattering have been used to quantify the interfacial roughness. Techniques like cross-sectional TEM, Auger, and depth resolved x-ray photoelectron spectroscopy give details with nm level resolution but this information is highly localized which may not represent the average behavior. In x-ray reflectivity measurement information is collected from area as large as 10 mm in length and 0.05 mm in width which represents average behavior of the structure. X-ray reflectivity has been used extensively in this report.

1.2 Influence of reflected neutrals

Other than issues discussed above there is one more problem which has not been addressed adequately in the past. That is, how the reflected neutrals influence the interface structure of multilayer at shorter periodsÅ In all sputtering deposition techniques it is known that large number of sputtering gas neutrals and ions get reflected from the target and hit the freshly growing film surface. This process of back scattering is a kinematical effect of ion bombardment on a target. If mass M of the incident ion is less than the target atom mass, law of momentum conservation allows the ion to be reflected from the target atom. The angular distribution of the reflected neutrals is also approximately cosine distribution same as for sputtered atoms. The most significant problem with the reflected neutrals is that they have 2-6 times the energy of the sputtered atoms and their number density also could be as high as 0.6-0.8 of sputtered atoms from the target. Results of simulation using TRIM code of SRIM 2008 to estimate the reflected neutrals when Ar ions of varying energy are hitting a W target are shown in the Fig.T.3.1. Figure T.3.1(a) shows the number density and energy value distribution of sputtered W atoms and as well as reflected Ar ions/neutrals when incident energy of the Ar ions is 500 eV. This figures shows that total numbers of reflected neutrals is high and comparable to numbers of sputtered atoms. Also energy of reflected neutrals can be very high compared to sputtered atoms. Figure T.3.1(b) shows changes in mean energy of sputtered atoms (red line) and reflected neutrals with the incident Ar ion energy.This graph shows that on increasing the Ar ion energy the energy of sputtered atoms do not change but mean energy of reflected neutrals increases

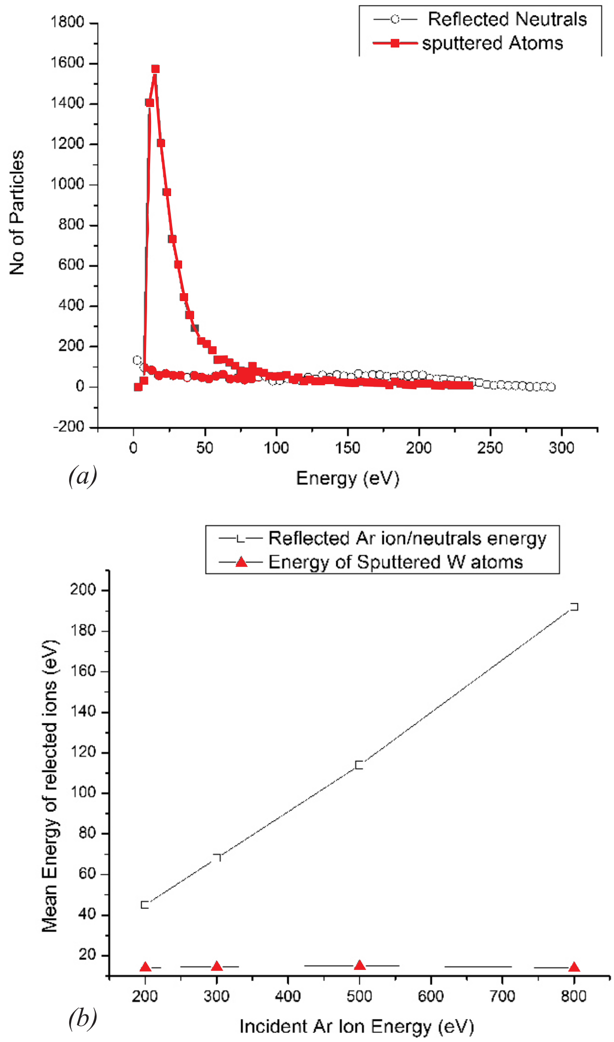

Fig.T.3.1: Top (a) number density and energy distribution of sputtered atoms (red line) and reflected ar ions/neutrals (black line) (b) (bottom). Changes in mean energy of sputtered W atoms (red triangle) and reflected neutrals with the incident Ar ion energy

significantly. This high energy is equivalent to that of ion/neutral beam hitting the growing surface as if ion assisted deposition is happening. Flux and energy of reflected neutrals was measured by Rossangel et al.. The paper concludes that bombardment of depositing film during sputtering should be considered as an unavoidable artifact of sputtering deposition technique. A review by Mattox covers the range of modifications which can happen in the film due to bombardment of energetic particles. These neutrals and ion hitting the film have sufficient energy to re-sputter the growing film, modify growth morphology, density, adhesion, interface mixing and can also generate residual stresses in the film. In case of multilayers, as we are depositing High Z and Low Z materials sequentially and the bombardment of neutrals seen by both interfaces is not same. It becomes important to understand the influence of these neutrals. At shorter periods these neutrals pose a challenge in fabricating a thin continuous layer itself.

This study has focused on to understand the role of reflected sputter gas neutrals on the growth and interface modification of short period multilayer mirrors. We have studied W/Si, W/C and W/B_4C combination by depositing multilayers of various periods starting from 9 nm to 1.5 nm. We have examined the influence of these reflected neutrals as how they affect the interface structure in these combinations. In this study we have demonstrated that these neutrals not only re-sputter the low Z layer but also produce intermixing during deposition which is more serious at shorter periods. This puts a limit on the lowest period which can be deposited without intermixing. In this study we have estimated the shortest period which can be deposited with a continuous layer for both W/Si and W/B_4C combinations. It is found that W/Si is more susceptible to damage due to reflected neutrals. Multilayers with continuous layers could show better thermal stability than intermixed multilayer mirrors. This aspect we examined by annealing W/B_4C multilayer of varying periods.

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2.0 Studies on W/Si multilayers

W/Si multilayers on float glass substrate were deposited using the in-house developed ion beam sputter deposition system. Base pressure in chamber before gas flow was 6 X 10^{-7} mbar and during the deposition it was maintained at ~ 4 X 10^{-4} mbar. We used commercially available 4" sputtering targets of 99.95% purity for W and 99.99 % purity for Si. The target to substrate distance during deposition was 35 cm. Roughness of each substrate was measured by x-ray reflectivity (XRR) technique on a reflectometer developed in house. The measured roughness of all substrates was ~0.4 nm.

Before multilayer deposition, we optimized deposition process parameters so that films with low interface roughness could be produced. For optimization we deposited various bilayers of 10nm thick Si on 10nm W, on a float glass substrate with different deposition parameters. It was found that films deposited at 1000 V beam voltage and a gas flow rate of 3 standard cubic centimeter per minute (SCCM) were best from the interface roughness point of view. The roughness of W layer was estimated to be 0.3 nm and of Si layer was estimated to be 0.4nm. The deposition rate for different materials was determined by depositing various thin films of W and bilayers of Si on W on float glass substrates and their thicknesses were measured by XRR. It was found that the deposition rate of W layer was ~1.3-1.4 nm/min and of Si layer was ~1.2.nm/min for all samples. Deposition rate was kept low as very thin layer were to be deposited. The thickness values estimated by XRR were used to calibrate the difference between actual deposition thickness and thickness shown by the quartz crystal monitor to accurately estimate the nominal thickness.

After the optimization, seven samples of different period thickness (d) were deposited. The nominal thickness of multilayer period was varied as 10.5, 7.8, 6.6, 4.0, 3.4, 2.9 and 2.4 nm marked as A-G.

To understand the changes in the interface structure of the samples at low period thickness detailed XRR fitting was carried out. For analysis of XRR measurements, Parrett recursive formalism was used to model reflectivity calculation. Roughness was taken into account using Nevot-Croce model. Out of various models considered it was found that best match with the experimental data for all samples was obtained with two-layer model. In two-layer model, only layers of W and Si were considered for all samples. There was no need of an interlayer in any of the samples. Considering, top Si layer to be partially oxidized helped to improve the fit quality. This is expected due to exposure of sample to air. Fig.T.3.2 shows the XRR experimental profiles (open circles) along with fitted curves (solid red lines) as a function of wave

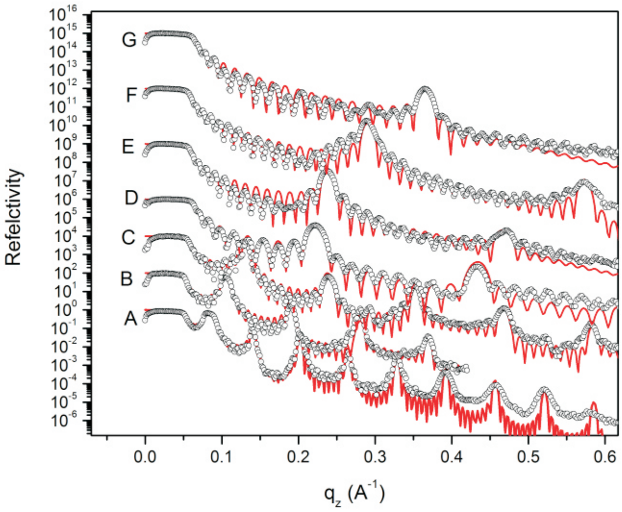

Fig.T.3.2:X-ray reflectivity curves both experimental (open circles) and fitted (solid lines) as a function of wave vector transfer 'q_z ($Å^{-1}$) for all samples (A-G) vector transfer q=4πsinθ/λ for all A-G samples. One can see that Kiessig oscillation in all samples are clearly visible and are matching well with the simulated profile. Simulations show that in samples with shorter period (e.g. F- G) even a thickness error of less than 0.05nm would spoil the regularity of Kiessig oscillation due to the uncertainty in total period thickness. The observation of well defined Kiessig oscillation indicates that bilayer periodicity is maintained in all samples for all layer pairs.

The most striking results of XRR analysis is that the estimated period thickness is less than the nominal thickness expected from optimization. The difference in the estimated thickness and the nominal thickness for W layer is negligible. But in case of Si layers, it is observed that the thickness is less than nominal value. The thickness for samples A-D is reduced by around 0.8nm, but for samples E-G difference is ~ -0.6 nm. From this analysis it is evident that only Si layer is getting lost during deposition similar loss of Si has been observed by Hasan *etal.* in samples made by sputtering. They attributed this loss to high-energy neutrals and ions getting reflected from the target during W deposition and hitting the Si layer. They have explained loss of Si as a two step process in which Si atoms are first transported through the growing W layer and then removed by collision with reflected high-energy particles bombarding the growing surface.

The second most interesting thing observed from reflectivity data analysis is that roughness (Table T.3.1) of the

Table T.3.1

S. N .	Esti. Period Thick ness (nm)	Thick ness (nm) and Density of W Layer (gm/cc^3)	Rough ness of W Layer (nm)	Thick ness (nm) and Density of Si Layer gm/cc^3	Rough ness of Si Layer (nm)
A	9.68	4.22/ 17.4	0.55	5.46/ 2.13	0.98
B	6.87	2.7/ 17.4	0.49	4.17/ 2.16	1.15
C	5.4	2.52/ 17.4	0.35	2.88/ 2.20	0.79
D	2.93	1.68/ 17.4	0.35	1.23/ 2.83	0.85
E	2.7	1.40/ 17.3	0.35	1.3/ 2.9	0.85
F	2.24	1.05/ 15.9	0.45	1.19/ 4.17	0.75
G	1.74	1.01/ 15.6	0.45	0.7/ 6.45	0.7

Si layer has increased from 0.4 nm observed in bilayers deposited to ~0.8-9 nm in multilayer and roughness of W layer is around 0.45 nm as observed during optimization. Both these observations suggest that as discussed earlier in section 1.2 reflected neutrals are playing a role in re-sputtering the Si layer. The Si layer roughness is also getting increased due to bombardment of high energy neutrals hitting Si layer during deposition of W. Whereas, W deposits smoother as neutrals hitting during Si layer deposition are not of high energy. To confirm this fact we again deposited few bilyers and trilayer of W/ Si and W/Si /W. XRR of all samples showed that in bilayer the roughness of Si was 0.4 nm and in trilayer when W layer was deposited on top of Si it was increasing to 0.9 nm. This observation confirms that hitting of reflected neutrals is modifying the interface and generating an asymmetry in the roughness of W on Si interface to Si on W interface.

We can see from Table T.3.1. that, density of 4.22nm W layer is 9.5% less then bulk W density in sample A. Similarly density of Si layer is 8.5 % less than bulk, which is expected as by sputtering method one does not get densities equal to bulk density due to porosities getting developed in the film during deposition. In case of sample D, where thickness of W and Si is 2.52 nm and 1.23 nm respectively, density of Si layer has increased to 2.83 g/cc from 2.13 g/cc. This indicates that some amount of W has diffused into Si layer. This can be explained with relative increase in roughness of the Si layer. The thickness of Si layer is only 1.23 nm and roughness is 0.85 nm, this high percentage of interface roughness compared to the layer thickness indicates that Si layer must be discontinuous at the interface. The discontinuity should enhance the diffusion of W into Si and increase the density of Si layer. In sample E, where thickness of W and Si is 1.4 nm and 1.3 nm respectively, the density of Si layer has further increased and density of W layer has also reduced suggesting that the now Si has also diffused into W layer. This confirms the observation made in re-sputtering analysis above that when W layer when goes below 1.4 nm it becomes partially discontinuous which promotes mixing of Si into W layer. Increase in the roughness of W layer when its thickness is 1.4 nm or below also confirms that film is getting discontinuous. Hence, diffusion of Si gets enhanced and density of W layer begins to fall. In following samples (F and G) as thickness of both W and Si arelow and these layers become discontinuous hence intermixing gets enhanced and density of Si layer keeps on increasing and reaches to 6.45g/cc and density of W layer keeps on reducing and reaches to 15.68g/cc in sample G. Analysis of re-sputtering and density data indicates that when thickness of W goes below 1.4 nm and thickness of Si layer goes below 1.3 nm diffusion of W and Si layers into each other starts and density contrast between two layers starts to drop at a rapid rate. This means that in W/Si multilayer the shortest period multilayer which can be deposited without intermixing should be more than 2.7 nm. If we have to reduce this period thickness some technique needs to be adopted to stop intermixing and make the interface smoother. Walton etal. had observed that shortest period multilayer of W/ B_4C which can be deposited without appreciable loss in reflectivity is 1.7 nm while in W/Si case it is 2.7 nm. General explanation for this observation is that difference must be due to the fact that B_4C being a compound is more resistant to chemical diffusion compared to Si. Si being more reactive it is possible to enhance the reaction and intermixing at the interface. But in this case the observed interface mixing is bombardment induced and from our studies the answer for sharper interface should be that either B_4C must be resistant to bombardment induced intermixing or energy of the reflected neutrals should be low in Walton's case of magnetron sputtering chamber. As we know that energy of reflected neutrals is of same order in both ion beam sputtering and magnetron sputtering. Hence, B_4C must be resistant to damage due to neutrals.

2.1 Comparison of damage resistance for different combinations

To compare the damage resistance of Si compared to B_4C and C which are popular choices of spacer layer for fabricating hard x-ray mirrors we did TRIM simulations in which all these layer were bombarded with 100 eV Ar ions. From Fig. T.3.3, it is clear that damage extends much deeper up to ~ 2.2 nm in case of Si and in case of both B_4C and C it extends up to 1.5 nm. This proves that indeed both B_4C and C have larger damage resistance from reflected neutrals

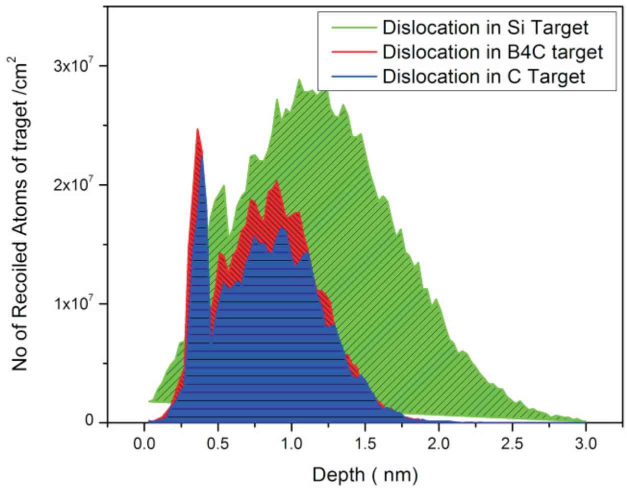

Fig.T.3.3: Damage profile produced due to hitting of 100 eV ions/neutrals on 3 nm layer of Si B_4C and C layer.

compared to Si. This indicates that using B_4C and C as spacer layers it should be possible to deposit multilayers with sharper interface and also with smaller periods. This is happening due to large lattice binding energy, surface binding energy and displacement energy of B_4C and C compared to Si. This seems a major reason why we are able to produce multilayers with sharper interface with C and B_4C. To confirm this observation we fabricated few bi layers of W/C and W/B_4C and tri layers of W/C/W and W/B_4C/W and analyzed the interface with XRR. The observations were following; the interface roughness of C/B_4C layer in bilayer was 0.5 nm and in tri layer increased to 0.55nm. This indicates that depositing W on top of C/B_4C does not change the interface roughness much. This confirms that reason for getting sharper interface and lower period thickness with B_4C spacer layer is its resistance to damage by reflected neutral.

3.0 Studies on W/B_4C multilayers

As explained in previous section that if W/ B_4C is deposited with magnetron sputtering it should be possible to get much lower interface roughness compared to W/Si. To check this possibility we decided to carry out a study on W/B_4C system.

The multilayer samples were deposited using magnetron sputtering system on Si (100) wafers, after ultrasonically cleaning first in acetone and then in methanol were used as substrate. Native silicon oxide layer was not removed from substrate. The base pressure in the process chamber was of the order of 10^{-8}mbar, and Ar pressure during the deposition was $5x10^{-3}$ mbar. The power of W target was varied from 80-130 W and rf power of B_4C target was 700 W. To deposit the shorter period W/ B_4C multilayers, lesser power was applied on W target to reduce the deposition rate. Eight samples marked as A-H were made.

In our samples nominal thickness of B_4C layer was continuously reduced from 2.6 to 0.8 nm and the nominal thickness W layer was kept around 1.7 nm in samples A-C and around 0.9 nm in samples D-H as given in Table T.3.2. Thickness of W was not reduced below 0.9 nm as there is a possibility that it may become discontinuous below this thickness. By systematically reducing the B_4C layer thickness it would be possible to find out minimum thickness of B_4C, at which it remains continuous.

All the XRR measurements were performed with a step size of 0.005° in theta axes, which is sufficient to observe any

Table T.3.2

S. N.	W layer Thickness /roughness (nm)	B_4C layer Thickness/ roughness (nm)	W density g/cc	B_4C density g/cc
A	1.60 /0.25	2.43 /0.3	16.4	2.2
B	1.72/0.29	2.05/0.29	16.4	2.2
C	1.69 /0.37	1.85 /0.37	16.4	2.2
D	1.0 /0.35	1.42 /0.35	16.4	2.2
E	0.97 /0.35	1.05 /0.35	16.4	2.2
F	0.85/0.45	0.95 /0.45	16.2	2.4
G	0.8 /0.42	0.72 /0.42	16.2	3.0
H	0.8 /0.5	0.52 /0.5	15.7	5.6

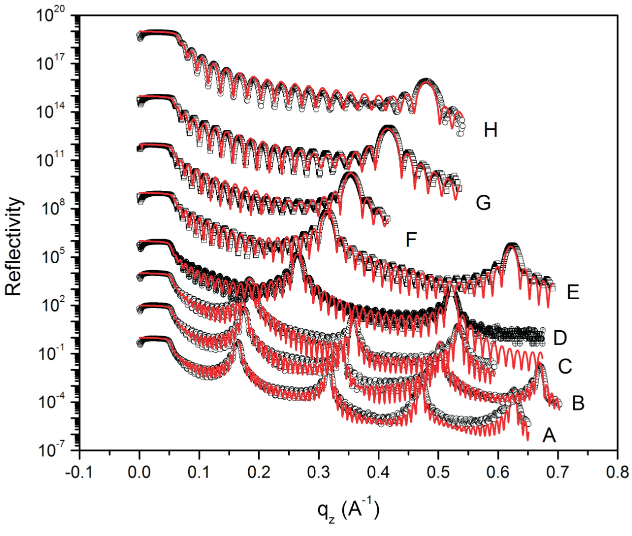

Fig.T.3.4: X-ray reflectivity curves both experimental (open circles) and fitted (solid lines) as a function of wave vector transfer 'q_z (Å^{-1}) for all the as deposited samples (A-H)

small changes in thin films. Specular reflectivity was measured with 100 micron slits in θ /2 θ geometry over the range of θ ≥ 0° to 3 to 5° depending upon the sample. Successive scans of same sample after removing and replacing on the diffractometer produced changes in peak position by less than 1 % and in peak reflectivity by less than 3 % of measured value. Fig.T.3.4 shows the XRR experimental profiles (open circles) along with fitted curves (solid red lines) as a function of wave vector transfer $q \geq 4\pi\sin\theta/\lambda$ for all the as deposited samples (A-H). One can see that Kiessig oscillation in all as deposited samples are clearly visible and are matching well with the simulated profile. Simulations show that in samples with shorter period (e.g. F- H) even a thickness error of less than 0.05nm would spoil the regularity of Kiessig oscillation due to the uncertainty in total period thickness. Estimated values of the average thickness, roughness and density of W and B_4C layer for all the as-deposited samples are listed in Table T.3.2. As mentioned earlier we wanted to keep W thickness around ~ 0.9 nm in samples D-H and around ~ 1.7 nm in samples A-C and reduce the B_4C thickness continuously. It observed that in as deposited samples estimated thicknesses of W and B_4C layers are close (within 0.2 nm) to the nominal values.This observation of estimated W and B_4C layer thickness emerging nearly same as nominal is differing with the observation made for W/Si multilayers. This confirms that due to higher damage resistance of B_4C layer there is no change in deposited film thickness. In the as deposited samples density of W layer was estimated to be 16.4 g/cc which is 15 % less than the bulk value and density of B_4C layer is 2.2 g/cc which is 12 % less than bulk. The reduction is expected as by sputtering method one does not get density equal to bulk density because of porosities getting developed due to extremely high quench rate in the film during growth. Density of W layer has remained constant in samples A to E whereas in sample F and G slight reduction to 16.2 g/cc is observed and in sample H density has reduced to 15.7 g/cc indicating that some intermixing of B_4C has happened in samples F,G and H. Density of B_4C layer has changed in as deposited layer for sample F to 2.4 g/cc (10 % more than samples A - E) and further increased in samples G and H to 36%, and 154% more respectively. This increase in density is signifying that W is getting mixed in B_4C layer in these samples. High density of B_4C layer in the as deposited sample H suggests severe intermixing. Looking at the variation in roughness of layers we find roughness of W layer has remained constant between 0.22-0.3 nm for all periods as shown in Table T.3.2. In case of B_4C layer roughness is showing a non linear increasing trend with thickness reduction. The ratio of magnitude of roughness to layer thickness is increasing very rapidly as the period is reduced and reaches 50 % of the film thickness for samples F, G & H. This high relative roughness ratio indicates that coverage of B_4C layer may not be complete in these samples. Tabulated density values of B_4C gives a clear picture of this variation in density with film thickness. It can be seen from

Table T.3.2 that when B_4C layer thickness goes below 1.0 nm its density begins to increase indicating presence of W in the B_4C layer. When, B_4C layer thickness goes below 0.7 nm, its density increases more rapidly suggesting increased intermixing and loss of density contrast along with fall in peak reflectivity. Simultaneous increase in roughness of B_4C layer along with increase in density of B_4C in the as deposited samples F-H confirms that W is intermixing and interface roughness is increasing. This increase in roughness must be due to enhancement in chemical roughness because of intermixing. One can draw following inference from these observations (1) when B_4C layer thickness is above 1.0 nm no mixing of W in B_4C happens, (2) when B_4C thickness goes below 1.0 nm some amount of W gets implanted in B_4C layer, (3) when B_4C layer thickness goes below 0.7 nm severe intermixing of W takes place and (4) increased intermixing is the cause of reduction in Bragg peak reflectivity of multilayers. This kind of intermixing can have influence on the long term stability and performance of the multilayer mirror.

To understand the degradation in the interface character of low thickness samples, simulation of damage produced through hitting of B_4C layer by reflected Ar ion/ neutral from the W target was carried out. It was found using TRIM that 30% of Ar ions hitting the W target can get reflected and 25% of the reflected ions can have very high energies nearing 100 eV or more. This estimate matches with reference.

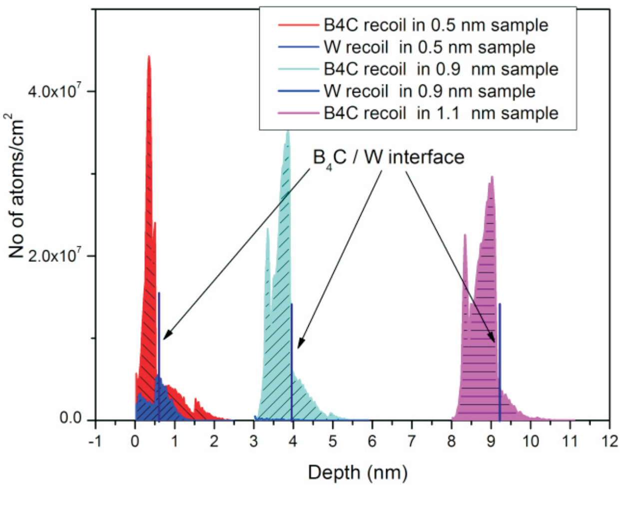

Fig.T.3.5: Recoil distribution of B_4C and W atoms when samples having top layer B_4C of 0.5, 0.9 and 1.1 nm thickness above 1.0 nm W layer is bombarded with 100 eV ions/neutrals. The recoil distribution graph has been shifted on X-axis (depth direction) for clarity.

Calculations were done to find out recoil distribution of W and B_4C due to hitting of 100 eV ion/neutrals. Simulations were done for three cases in first case we had 1.1 nm of B_4C layer on the top of 1.0 nm W layer and in other cases thickness of B_4C layer was reduced to 0.9 and 0.5 nm. All the simulations were done for hitting by just 20000 particles of 100 eV energy.

Fig.T.3.5 shows the recoil distribution of B_4C and W atoms when the top layer of B_4C of 0.5, 0.9 and 1.1 nm thickness is bombarded with 100 eV ions/neutrals. The recoil distribution graph has been shifted on X-axis (depth direction) for clarity. It can be clearly seen than when B_4C layer is 1.1 nm thick little amount of B_4C is recoiling towards W layer and no event of W recoil towards B_4C layer was recorded. However, when B_4C layer thickness was reduced to 0.9 nm more recoil of both W and B_4C was seen on each side of the interface and in case of B_4C being 0.5 nm large movements of both species was seen on either side. These simulations support the observations made above from XRR analysis that when thickness of B_4C layer is less than 1.0 nm more intermixing is expected and B_4C layer becomes nearly discontinuous at 0.5 nm thickness. This analysis again points out that the cause for making sharper interface at shorter periods is controlled by hitting of reflected neutrals and ions. From these observations we can conclude that when B_4C layer thickness goes below 1.0 nm intermixing of W starts in the B_4C layer. The intermixing is happening due to bombardment of high energy reflected ion/neutrals as confirmed by simulations. Lowest continues thickness of B_4C have been estimated to be 1.0 nm. By no other method than adopted in this study we could have deduced lowest continuous film thickness of the insulating B_4C layer. From these observations it can be concluded that multilayer with W around 0.8 nm and B_4C around 1.0 nm can be deposited with lowest interface roughness and intermixing. Hence the shortest period which can be deposited without any intermixing is 1.8nm.

To check the thermal stability of these multilayer samples some of the samples were annealed. The annealing treatments of multilayer samples were carried out as follows. The multilayer samples were placed in aluminum boat inside the quartz tube. The quartz tube is turbo molecularly pumped down to a base pressure of the order of 10^{-6} mbar using a turbo molecular pump. All the multilayer samples were heated at 500^0C for duration of 4 h. It was concluded that samples with period 2.0 nm (E) and above show good thermal stability with no or very little degradation and samples below this value show sensitivity to annealing and drop in Bragg peak

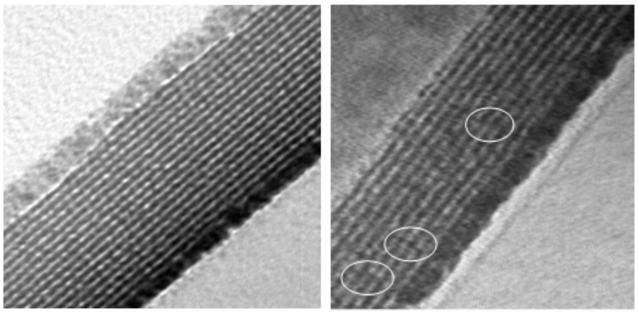

Fig.T.3.6: Cross sectional TEM of sample E (D= 2.02nm) and G(D=1.52nm)

reflectivity due to annealing. This indicates that as deposited samples with unmixed interface show good thermal stability also.

Examination of the interface structure was carried out by cross sectional TEM (CSTEM) of as deposited samples E (D≥ 2.02 nm) and G (D≥1.52 nm). The layer structure of the shorter period samples is shown in Fig. T.3.6. In this figure black layer represents W and white layer represents B_4C. It is visible that layer structure is maintained and layers in the multilayer are visible as separate layers in both samples. But contrast between W and B_4C layer is much better in sample D compared to sample G. This also confirms that intermixing is high in sample G. Multilayer layer period thickness estimated from the TEM image is 2.0 nm for sample E and 1.5 nm for sample G which matches with the thickness estimated by XRR. No swelling of W layer is seen as observed by Walton *et al*. A closer look at the image in sample G it can be clearly seen that at many places the W layers are connected across few B_4C layers (some of such locations are encircled in the figure). Another interesting observation is that in spite of intermixing overall multilayer periodicity is maintained. It can be concluded that W layer is continuous in both samples and B_4C layers is significantly discontinuous in sample G and B_4C.

Development of Large layer pair samples

After all the optimization and annealing studies we carried out deposition of large layer pair multilayer samples of W/B_4C. Since, it was concluded that the minimum thickness of W and B_4C which can be deposited as continuous layer were 0.9nm and 1 nm. To check the evolution of interface roughness with increasing bi-layer number, N we fabricated multilayer with period 2 nm and bi-layer numbers 20, 40, 100, 200 and 300. It was observed that intefaces were becoming smoother with increasing N and diffused component was reducing with increase in number of layer pairs. Reflectivity of first Bragg peak for sample with 100,200 ad 300 layer pair sample is shown in Fig.T.3.7. The 300 layer pair, 2 nm period sample has shown 1.2% energy resolution at 8 keV and 59% reflectivity. The achieved reflectivity and resolution are comparable with the best multilayers deposited by magnetron sputtering. The high reflectivity and high resolution proves that devloped multilayer mirrors are suitable to be used as a monochromater or grazing incidence mirror in hard x-ray region.

Major observations of our study on W/B_4C multilayers between 1.2 to 4 nm periods are the following. It is observed that when B_4C layer thickness goes below 1.0 nm intermixing of W starts in the B_4C layer. The intermixing is happening due to bombardment of high energy reflected ion/neutrals as confirmed by SRIM simulations. Lowest continues thickness of B_4C have been estimated to be 1.0 nm. From these observations it can be concluded that multilayer with W around 0.9 nm and B_4C around 1.0 nm can be deposited with lowest interface roughness and without intermixing. Our observations are in contrast with earlier work done by Walton *et al*. and Jankowski *et al*. We have observed increase in B_4C layer roughness with period reduction and no increase in W layer thickness which is opposite of observations by Walton et.al. The measured Bragg peak reflectivity after annealing of different samples show that samples of 2.0 nm period or above do not show much degradation in the reflectivity of first Bragg peak. Also, density of W and B_4C layers has remained unchanged after annealing in these samples indicating their stability. Present study reveals that 1.9 nm period is the smallest period which should shows sharp interface and good thermal stability for W/B_4C combinations. Our studies on large layer pair samples have confirmed these conclusions. This is very useful information for developing high heat load short period multilayer mirrors. We could fabricate 2 nm period 300 layer pair samples with best reflectivity and good resolution for hard x-ray applications.

Summary

On the basis of this study we can conclude that reflected neutrals play an important role in growth of multilayer mirrors. They influence the growth in two ways (1) re-sputter the low Z layer and make it rough (2) If thickness of low Z layer is less than 1.5 nm for Si layer and 1 nm for B_4C layer than reflected neutrals produces intermixing at the buried interface below low Z layer by creating a collision cascade. It this way these neutrals reduce the contrast between low Z and high Z layers. To further extend the period to shorter lengths it would be required to reduce the damage from reflected ions /neutrals of low Z layer. Simulations using SRIM have shown that using Xe in place of Ar would reduce the number of reflected ion/neutrals to 25% of what is expected with Ar with slightly reduced energies. But use of Xe would reduce the sputter yields of B_4C by half. The thermal stability shown by these multilayers and lack of chemical reaction between W and B_4C give an idea that depositing multilayer at higher temperatures than room temperature might help us in achieving even shorter periods with lower roughness.

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