



HAL
open science

Depth analysis of Al/ZrC interfaces using SIMS and x-ray reflectivity

Mohammed H. Modi, Mangalika Sinha, Aniruddha Bose, Amol Singh,
Philippe Jonnard

► To cite this version:

Mohammed H. Modi, Mangalika Sinha, Aniruddha Bose, Amol Singh, Philippe Jonnard. Depth analysis of Al/ZrC interfaces using SIMS and x-ray reflectivity. *Surface and Interface Analysis*, 2018, 50 (11), pp.1239 - 1242. 10.1002/sia.6443 . hal-01919286

HAL Id: hal-01919286

<https://hal.sorbonne-universite.fr/hal-01919286>

Submitted on 12 Nov 2018

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

Depth analysis of Al/ZrC interfaces using SIMS and x-ray reflectivity

Mohammed H. Modi¹, Mangalika Sinha¹, Aniruddha Bose², Amol Singh³, Philippe Jonnard⁴

¹ Soft X-ray Applications Lab, Raja Ramanna Centre for Advanced Technology, Indore 452013, India

² Superconducting Proton Linac Section, Raja Ramanna Centre for Advanced Technology, Indore 452013, India

³ National Synchrotron Radiation Research Center, Hsinchu 30076, Taiwan.

⁴ Sorbonne Université, Faculté des Sciences et Ingénierie, UMR CNRS, Laboratoire de Chimie Physique – Matière et Rayonnement, 4 place Jussieu, F-75252 Paris cedex 05, France

Corresponding Author: modimh@rrcat.gov.in

Abstract

The Al/ZrC/Al/W multilayer structure is suitable for waveguide applications in the hard x-ray range in order to confine the wave field in a nanometer-thick layer. Intermixing of Al at the interfaces is a serious problem to achieve the expected performances from an experimentally grown multilayer. In the present study, our aim is to investigate the effect of a capping layer on the ZrC/Al interfaces in an C/Al/ZrC/Al/W waveguide structure. We use time of flight secondary ion mass spectrometry (ToF-SIMS) and soft x-ray reflectivity (SXR) to study the x-ray waveguide structure. Structural parameters of the stack, density, thickness and roughness of the layers, are determined through fitting of SXR data. SIMS results indicate that the Al diffusion towards the top of the stack is responsible of the formation of wide and asymmetric interfaces in the waveguide structure.

Introduction

X-ray waveguides are a modern concept to generate nano-size x-ray beams required for the study of novel properties of nano-materials. X-ray waveguides can also be used to enhance the spatial coherence of x-ray beams and therefore can be very useful in x-ray holography and coherent diffractive imaging experiments. Waveguides are essentially non-dispersive optical devices suited to a wide range of photon energies and bandpass. The limit of full coherence can be reached with proper design and with a suitable choice of the guiding and cladding materials. X-ray waveguide is analogous to optical fibre where the beam is guided through a core sandwiched between two cladding materials.

In x-ray waveguides, the electric field of the incident photon beam is confined inside a layer through the generation of x-ray standing waves. The guiding layer is generally made of a light material surrounded by two layers made of heavy materials. In the literature very few attempts have been made to understand the role of materials combination, film roughness, interface width, etc. on the performances of waveguides. Simulation shows that the Al/ZrC/Al/W structure can serve as a good combination for waveguide applications in hard x-ray region. Indeed, Figure 1 shows the depth and angular distributions of the electric field of 10.25 keV x-

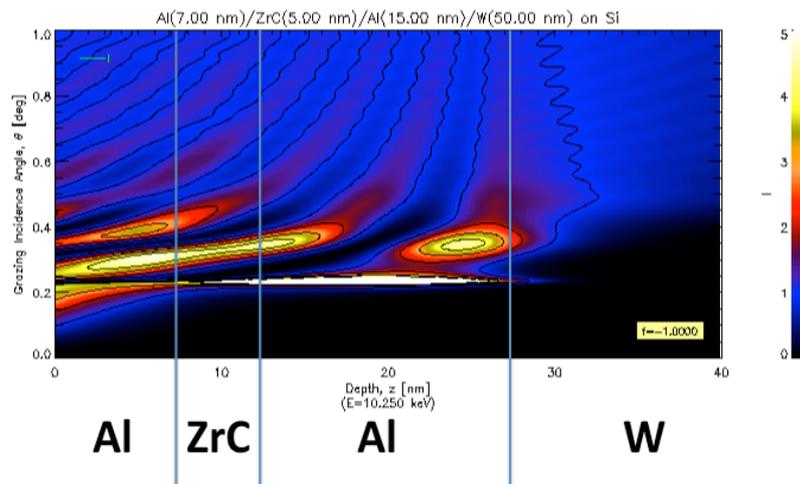


Figure 1: Calculated angular and depth distributions of the electric field inside an Al/ZrC/Al/W waveguide structure excited with a 10.25keV incident x-ray beam. The lateral color bar on the right is proportional to the intensity of the electric field.

rays inside this structure. It is evident that the x-rays can be confined in the Al guiding layer

when the glancing angle is close to 0.23° . In order to experimentally realize such high quality confinement one needs to make devices with ultra-smooth interfaces and no interdiffusion layer. The main challenge in fabrication of such devices is to prevent intermixing between the guiding and cladding layers.

Grazing incidence x-ray reflectivity (GIXR) using Cu K_α (0.154 nm) radiation is a powerful tool to probe thickness, roughness and density of thin films. However this technique has severe limitation to probe structures where there is a poor refractive index contrast between layers. Because of the limited resolution of momentum transfer vector for the Cu K_α radiation, it is very difficult to analyse films of very large thicknesses. In the soft x-ray region, near absorption edges, energy-dependent atomic scattering factors have a resonant behaviour¹ and offer an opportunity to enhance the optical index contrast among the thin films. This can be achieved by tuning the energy of x-rays to the absorption edges of specific elements. Soft x-ray reflectivity (SXR) has been used for the characterization of organic and low contrast thin films². Al/ZrC/Al/W structure is found difficult to get analysed by GIXR because of the large thickness of the overall structure and the poor refractive index contrast at the interfaces owing to intermixing. Earlier studies^{3,4} carried out using SXR suggested a large intermixing at the Al/ZrC interfaces and an oxidation on top of the Al layer.

In order to make the Al/ZrC system useful for waveguide applications it is required to protect the top Al layer with a non-reacting capping layer and study the resultant interface quality. Thus it is necessary to analyse the nature of the interfaces formed following the intermixing of adjacent materials in order to improve the interface quality for actual devices. In the present study we have incorporated a protective layer of carbon on top of the Al/ZrC/Al/W structure. The top carbon layer is designed to prevent the oxygen interdiffusion from the top of the stack. We have deposited a 5-layer structure C/Al/ZrC/Al/W on a Si substrate. To understand the nature of interface diffusion we have used time of flight secondary ion mass spectroscopy (ToF-SIMS). To obtain reliable structural parameters, SXR measurements are carried out at 5, 6 and 7 nm incident wavelengths at the Reflectivity beamline of Indus-1 synchrotron radiation facility and best fit is obtained using the same parameters for all three wavelengths.

Experimental Procedure

An x-ray waveguide composed of 5 layers, C (5 nm) / Al (7 nm) / ZrC (5 nm) / Al (15 nm) / W (50 nm), Figure 2, has been deposited onto a silicon substrate.

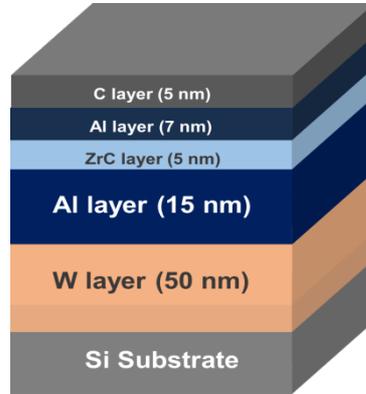


Figure 2: Schematic diagram of the five-layer sample C/Al/ZrC/Al/W deposited onto a silicon substrate.

(15 nm) / W (50 nm), Figure 2, has been deposited on a polished Si (100) wafer using ion beam sputtering. Prior to deposition, a base pressure of 3×10^{-5} Pa was achieved. Deposition has been carried out under argon ambient at constant pressure of 6×10^{-2} Pa. Commercially available sputtering targets of ZrC, Al and W (of 99.99% purity) have been used. Optimized parameters for better *rms* roughness, 1 kV beam voltage and a gas flow rate of 3 standard cubic centimetres per minute, were used.

SXR measurements have been carried out using Reflectivity beamline (BL-4)⁵ at Indus-1 synchrotron radiation facility. A toroidal grating monochromator delivered monochromatic photons in the 4-100 nm wavelength region with high flux and moderate spectral resolution ($\lambda/\Delta\lambda \sim 200-450$). The beamline employs toroidal mirrors for pre-and post-focussing. Different harmonic suppression edge filters are installed in the beamline to reduce higher harmonic contamination⁶. The experimental station is a high vacuum reflectometer consisting of a two-circle goniometer to accomplish θ - 2θ scans. A soft x-ray silicon photodiode detector (International Radiation Detector Inc.) is used for measuring the reflected intensity. SXR data have been analysed using the Parratt formalism⁷. The effect of surface roughness has been taken into account using the Nevot-Croce model⁸.

ToF-SIMS measurements were performed using Cs^+ ion gun operating at 1 keV (TOFSIMS-5, Iontof) to sputter the sample. However, the analysis was carried using Bi^+ ions

operating at 30 keV, 4.7 pA. The analysis area was $100\ \mu\text{m} \times 100\ \mu\text{m}$ inside the sputter crater of $300\ \mu\text{m} \times 300\ \mu\text{m}$.

Results and Discussions

Figure 3 shows the measured and fitted soft x-ray reflectivity patterns of the waveguide structure obtained at the three incident wavelengths. Structural parameters, thickness, roughness and delta (δ) and beta (β), *i.e.* the real and imaginary parts of the optical index respectively of the

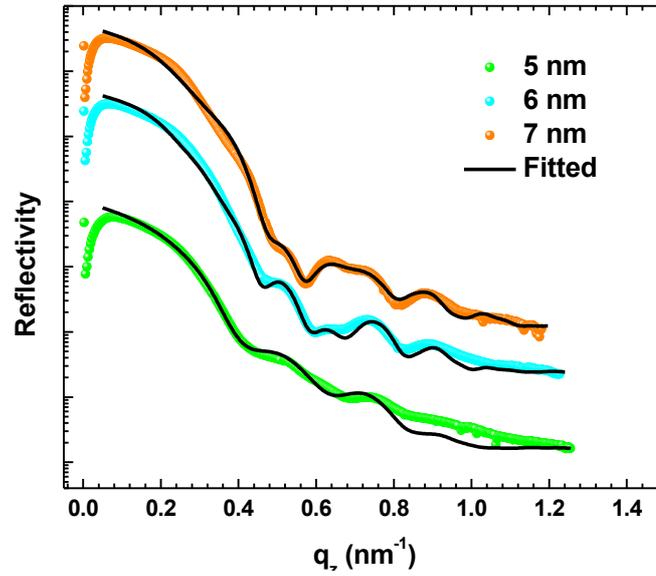


Figure 3: Measured (dots) and fitted (solid black line) angle-dependent soft x-ray reflectivity patterns of the C/Al/ZrC/Al/W waveguide system for the incident wavelengths of 5, 6 and 7 nm. Curves are vertically shifted for clarity.

different layers were considered as the fitting parameters. The reference values of delta (δ) and beta (β) were obtained from the CXRO website⁹. The aimed thickness values were taken as the initial parameters for the fitting of the SXR data. The best fit of the SXR data was obtained using a 5-layer model (C/Al/ZrC/Al/W) and a native oxide layer of the silicon substrate. The obtained

Layer	Thickness (nm)	Roughness σ (nm)
C	7.4	1.9
Al	9.95	1.8
ZrC	7.53	1.8
Al	18.3	2.1
W	55.7	2.4
SiO ₂	4.5	0.9
Si sub	-	0.4

Table 1: Structural parameters of the different layers in the analysis of the soft x-ray reflectivity data of the C/Al/ZrC/Al/W waveguide system.

structural parameters of the different layers are tabulated in the Table 1. The same parameters were valid for three wavelengths (5, 6 and 7 nm) where reflectivity measurements were performed.

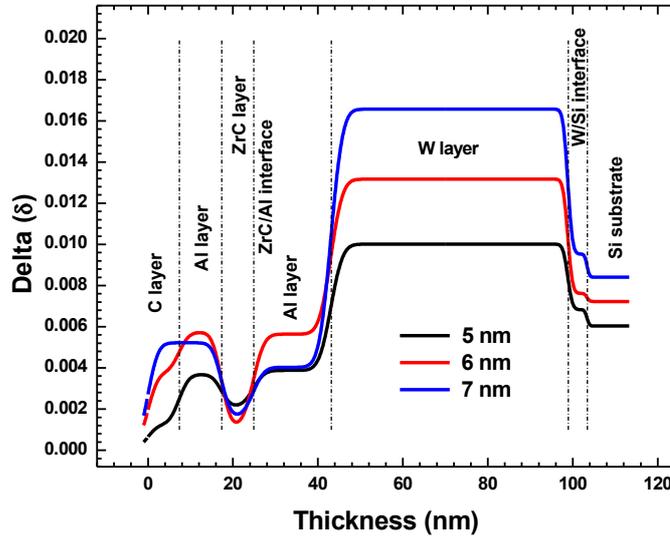


Figure 4: In-depth profile of the real part of the optical index obtained from the fit of the SXR data for the incident wavelengths of 5, 6 and 7 nm.

The in-depth profiles of the real part of the optical index in the C/Al/ZrC/Al/W waveguide system as obtained by fitting the SXR data are shown in the Figure 4. It is evident that the 50 nm thick W layer is almost uniform except near the Al-on-W and the W-on-substrate interfaces. Near the W-on-Si interface, the presence of a native oxide layer was assumed. However, it is found that the obtained delta values at this interface are comparatively higher than that of bulk SiO₂, indicating the incorporation of W into the native oxide layer of substrate. Near the ZrC-on-Al and Al-on-ZrC interfaces, a gradual increase in the delta value is observed instead of a sharp jump, indicating some intermixing of between the ZrC and Al layers.

The SIMS depth profiles of the Al, C, Zr, ZrC and W positive ions are shown in the Figure 5. The sputtering time scale is not calibrated to get the direct information about the thickness value. The regions corresponding to the C, Al, ZrC and W layers as well as the substrate are marked by the vertical lines. The carbon ion curve suggests that carbon is diffusing towards the first Al layer. The presence of carbon in the ZrC layer can be due to the presence of the unreacted carbon atoms in the ZrC layer itself. The Al profile suggests that aluminum

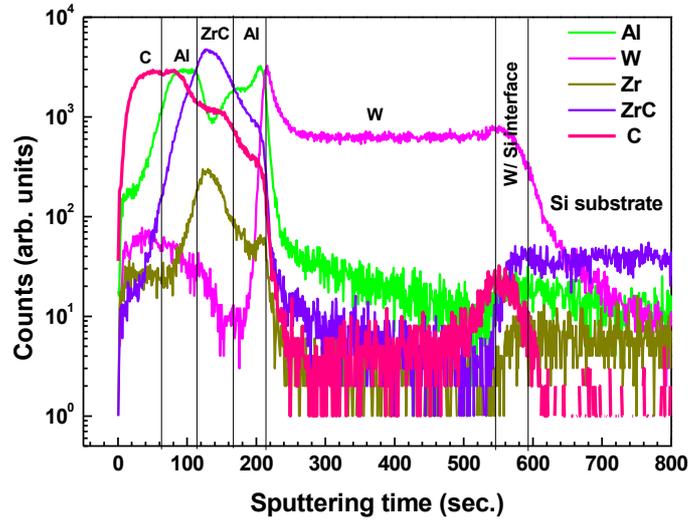


Figure 5: Time of flight secondary ion mass spectrometry profiles of the C/Al/ZrC/Al/W waveguide system.

diffuses towards the top surface. Therefore this leaves behind an empty space where the material from the top layer can diffuse (inside the bottom Al layer) and causes an asymmetry in the Al layer profile. The ZrC profile has an extended diffusion path towards the buried Al layer whereas it has short diffusion path into the top Al layer. This argument supports the statement of Al preferring the diffusion towards the top of its layer. The W layer profile is uniform except near the W-on-Si interface, showing that tungsten diffuses towards the Si substrate. Consequently, large interface widths in C/Al/ZrC/Al system are observed, in agreement with the large interfacial roughnesses, around 2 nm, deduced from the SXR data.

Conclusions

In the present study, SIMS and SXR techniques are used to analyse the interface profile and structural parameters of a C/Al/ZrC/Al/W waveguide system. The analysis of the SXR data provides information about the thicknesses of the different layers of the stack as C (7.4 nm) / Al (9.95 nm) / ZrC (7.5 nm) / Al(18.2 nm) / W(55.7 nm). The roughness of the Al-on-ZrC, ZrC-on-Al and Al-on-W interfaces are found in 1.8 - 2.4 nm range. The SIMS data suggest that aluminum diffuses towards the top layer and leaves empty space for diffusion from the layer deposited on top of Al layer. This phenomena leads to an asymmetry of the interfaces.

Acknowledgements

Authors are thankful to Dr. Sanjay Rai and Mr. Rajnish Dhawan for providing samples and to Mr. R. K Gupta for help in soft x-ray reflectivity measurements.

References

- ¹ Resonant Anomalous X-ray Scattering: Theory and Application, edited by G. Materlik, C. J. Sparks, and K. Fischer (North-Holland, Amsterdam,1994).
- ² Cheng Wang, T. Araki, and H. Ade, *Appl. Phys. Lett.* **87**, 214109 (2005).
- ³ Amol Singh, M.H. Modi, R. Dhawan, P. Jonnard, K. Le Guen, and J.-M. André, *AIP Conference Proceedings*, **1832**, 080093, (2017)
- ⁴ Amol Singh, M.H. Modi, R. Dhawan, P. Jonnard, K. Le Guen, and J.-M. André, *AIP Conference Proceedings*, **1832**, 080093, (2017)
- ⁵ R. V. Nandedkar, K. J. S. Sawhney, G. S. Lodha, A. Verma, V. K. Raghuvanshi, A. K. Sinha, M. H. Modi, and M. Nayak, *Curr. Sci.* **82** 298 (2002).
- ⁶ Mohammed H. Modi, R. K. Gupta, Amol Singh, and G. S. Lodha,2012 *Appl. Opt.*. **51** 3552.
- ⁷ L. G. Parratt, *Phys. Rev.* **95** 359 (1954).
- ⁸ L. Nevot and P. Croce, *Rev. Phys. Appl.* **15** 761(1980).
- ⁹ http://henke.lbl.gov/optical_constants/asf.html