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Auspices Statement

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Developing multi-layer mirror technology near 45 nm using Sc/Si interfaces

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Abstract: Given the existing X-ray laser sources near 45 nm it would be useful to produce efficient X-ray optics in the 35 to 50 nm wavelength range that could be utilized in new applications. In this work we are developing the process to stabilize the interfaces of nanolaminate structures using materials such as Sc and Si. These materials will enable us to develop new multi-layer mirror technology that can be used in the wavelength range near 45 nm. To obtain this objective, the interfacial structure and reaction kinetics must first be well understood and then controlled for design applications. In this work we fabricate several Sc/Si multi-layer mirrors with and without a B_4C *barrier layer. The structure and reflectivity of the mirrors are analyzed.*

1. Introduction

Since the development of the tabletop X-ray laser [1] there has been a need for high reflectivity coatings that work in the 35 to 50 nm wavelength range to facilitate the development of applications such as interferometry and other optical techniques in this shorter wavelength region. Multi-layers made of scandium (Sc) and silicon (Si) have shown promise for producing high reflectivity mirrors in this wavelength range.[2] Despite initial success, the Sc and Si layers tend to inter-diffuse, especially at temperatures above 150 to 200 degrees Celsius. To prevent inter-diffusion, tungsten (W) barriers have been used between the Sc and Si layers. However W is highly absorbing and this significantly reduces the reflectivity of the multi-layer mirror. In this work we pursue the use of a boron carbide (B_4C) diffusion barrier since it has much lower absorption than W and has the potential for creating smoother layers.

2. Interface stability

The use of nanolaminates, i.e. multilayered structures, is well established in applications that range from optical coatings to hard surfaces for cutting tools. In all applications, there is an underlying materials science issue that limits utility – that is, interface stability. Nano-laminates are artificial as synthesized from vapor deposition techniques.[3] An artificial short-range order is introduced that's meta-stable in origin. Nano-laminates will transform to ground states with sufficient energetic activation. For example, the low-temperature anneal treatments of nickel layers alternating with chrome-molybdenum layers initiates inter-diffusion and reveals the kinetics that are useful to predict long-term stability of alloys for a potential application as a Yucca Mountain container.[4] In this work we explore the nature of interface stability in a nano-laminate system that exhibits the tendency for fast diffusion. A case study is found in refractory, rare-earth, or transition metals layered with silicon. A picture that illustrates this issue is seen for a multi-layer that consists of layers A and B with thickness l_a and l_b, respectively. In all cases, the repeat periodicity (also called the composition wavelength) of the multi-layer (l_{ab}) is less than the additive sum of its component layer thickness when deposited separately, i.e. $l_{ab} < l_a + l_b$. Some mixing, inter-diffusion, and/or relaxation takes place in the laminated structure. This is the generic nature of the instability. We examine the change in the interface structure that occurs during the deposition process and that which occurs afterwards.

Of particular interest in the silicon (Si) nano-laminate family with regards to optimizing reflectivity are the material systems with molybdenum (Mo) and scandium (Sc). Mo/Si multi-layers have become prominent as a manufactured material of choice for deep extreme ultraviolet (EUV) lithography near 13 nm wavelength. Sc/Si offers a potential application for effective manipulation of compact-discharge and laser-driven tabletop X-ray laser (XRL) beams that currently produce output at 46.9 nm.[5-7] As this Sc/Si system represents a fast diffusion couple, interface barriers that consist of only a few mono-layers of B_4C can be included to try to stabilize the structure. These experiments explore a regime of wavelength (as the Henke tables for optical constants cut off at 40 nm) and target design that represent a new challenge to our understanding of interface stabilization. Furthering our basic understanding in evaluating the interfacial structure of intrinsically metastable structures can only broaden and deepen our understanding for application of interest to Laboratory Programs. Sc/Si may provide a reflecting surface suitable for use in back light diagnostic tools for LLNL laser facilities. Also, there is always room for new technical improvements that can enhance the reflectivity of Mo/Si for the deep EUV or soft X-ray regime.

3. Fabrication of Sc/Si multi-layers mirrors

We fabricated two types of multi-layers mirrors to assess the effect of using a barrier layer to reduce the effect of the inter-diffusion. Target materials and substrates that were in hand were used in an existing multi-layer deposition system of the Chemistry and Material Science (CMS) Coatings Capability. Planar magnetron sputtering was used to deposit the layers. To do the deposition the Sc and Si targets are held at a negative potential and the Ar+ ions hit the target surface and sputter off neutral target material that diffuses to the substrate surface. Our deposition system is shown in Fig. 1. The substrate is rotated to the sputtering targets of Si, B_4C, and Sc. An ion mill was used initially to clean the substrate surface. Our targets used various Si substrates with the final samples being produced on super-polished fused silica substrates with surface roughness of approximately 0.1 nm. The three magnetron sources were operated in DC mode and were shielded from each other to prevent cross contamination. The deposition rates and total accumulation on the substrates were measured with quartz crystal monitors.

The multi-layers were produced with 20 layer pairs. First we produced a baseline multi-layer without any barrier layer. For sample No. 319 we used layer dimensions: 9.0 nm Sc and 14.9 nm Si for a total layer thickness of 23.9 nm. To compare with this we used the B_4C barrier to produce sample No. 320 with layer dimensions: 8.3 nm Sc, 0.7 nm B_4C, 14.8 nm Si, and 0.7 nm B_4C for a total layer thickness of 24.5 nm. The profilometry measurements of the quartz substrates indicate a total thickness of 0.47 microns for sample No. 319 and 0.49 microns for No. 320. These multi-layers start with a Si layer and end with a Si layer. A 4 to 5 nm thick cap layer of Si is deposited on top of the multi-layer stack to minimize oxidation. A diagram of the multi-layer with the barrier layer is shown in Fig. 2.

4. Characterization of the multi-layers

To further characterize the Sc/Si multi-layers several methods were used. To use the atomic force microscope (AFM) a sample was cleaved in cross section to reveal the layers. In contact mode, the AFM [8] functions by moving a probe tip across the sample surface and mapping its features. The tip is attached to a cantilever whose deflection is used to map the height of the surface. The deflection is measured by reflecting a laser off the cantilever and measuring the deflection of the laser on a photo-detector. The AFM has 2 nm resolution in the plane of the sample and 10 pm resolution in mapping the height of the surface. To avoid the tip of the probe running off the edge of the multi-layer sample, a layer of enamel was bonded on to the top of the multi-layer. Fig. 2 shows a diagram of the Sc/Si multi-layer sample No. 320 with the B_4C barrier layer. Fig. 3 shows the contact mode deflection image taken of sample No. 320 using the AFM. The image demonstrates the uniformity of the layers and displays the grain boundary structures associated with the fracture. It takes much less time to produce the AFM images than comparable images using a TEM. However the resolution is more limited with the AFM.

Ideally we would like to measure the normal incidence reflectivity of the different multi-layer mirrors to compare the effectiveness of using the barrier layer. The traditional facility used to measure multi-layer reflectivity is the ALS beam-line at Lawrence Berkeley Laboratory. However this only works for wavelengths shorter than 40 nm. In order to use this beam-line with our samples we had to do measurements at 45-degree angle of incidence. Samples Nos. 319 and 320 were measured. Fig. 4 shows the reflectivity versus wavelength. Sample No. 319, made just with Sc and Si, had peak reflectivity of 25.6 and 25.9% in two different spots, at a wavelength of 22.4 nm. Sample No. 320 with the B_4C barrier layer had peak reflectivity of 25.3, 24.8, and 25.8% in 3 spots, at wavelengths of 23.9, 24.0, and 23.4 nm. So the reflectivity is nominally the same, but the multi-layer with the B_4C barrier layer is shifted up in period by somewhat less than the nominal thickness of the B_4C layers added. The use of nanometer-thin, boron-carbide barrier layers is shown to stabilize the interface structure without adversely affecting the measured reflectivity in comparison to the Sc/Si mirror.

Fig. 5 shows the predicted reflectivity for the two samples at normal incidence. The peak reflectivity is predicted near 37 nm while we were targeting 45 nm. Now the analysis of the LBL measurements indicated a period of 18.1 and 19.5 nm for the two samples as compared with the 23.9 and 24.5 nm thickness measured during the production process. We believe this discrepancy may be due to the reflectivity measurements being done near the edge of the samples where the deposition thickness may be thinner.

We have made a new set of Sc/Si multi-layer optics coated on one inch diameter super-polished fused silica substrates for potential experiments using the 46.9 nm laser sources. Two sets of mirrors were made with and without the barrier layers. For experiment No. 723, the Sc layers are 9.1 nm, the Boron-Carbide layers are 0.7 nm, and the Si layers are 15.5 nm thick for a total thickness of 26.0 nm. For experiment No. 724, the Sc layers are 9.4 nm, the Boron-Carbide layers are omitted, and the Si layers are 17.0 nm thick. The repeat period of the multi-layer is 26.4 nm. Both sets of mirrors have larger periods than the earlier samples to better optimize them for 46.9 nm. These mirrors are still being evaluated for both the absolute value of the peak reflectivity and the wavelength at which the peak occurs.

5. Conclusions

In the course of this work we did accomplish many of our initial goals. We developed an analytic model to assess the meta-stability of the multi-layer by computing the inter-diffusion rate for multi-layers composed of *dissimilar crystalline structures* using the artificial concentration wave method. A trial application was made for the Ni/CrMo system and that result was presented at the 132^{nd} Annual Meeting of The Metallurgical Society in San Diego, California on March 2-6, 2003.

We did synthesize a number of Si-based nano-laminates using a computer-controlled, planar-magnetron sputter-deposition system. We made several samples of Sc/Si multi-layer mirrors that were tested and new iterations of these mirrors that are still being evaluated.

The reflectivity of the Sc/B_4C/Si and Sc/Si nanolaminates were measured at grazing incidence using 8 keV radiation and at 45^o using the ALS beam-line at LBL. The structure of the Sc/B_4C/Si nano-laminate was imaged in cross-section using atomic force microscopy.

The use of nanometer-thin, boron-carbide barrier layers is shown to stabilize the interface structure without adversely affecting the measured reflectivity in comparison to the Sc/Si mirror. Finally, a set of prototype multi-layer optics was prepared for a collaborative evaluation at normal incidence reflectivity at 46.9 nm.

Acknowledgements

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Fig. 1. Magnetron sputtering deposition system used to fabricate Sc/Si multi-layer mirrors with and without B_4C barrier layer.

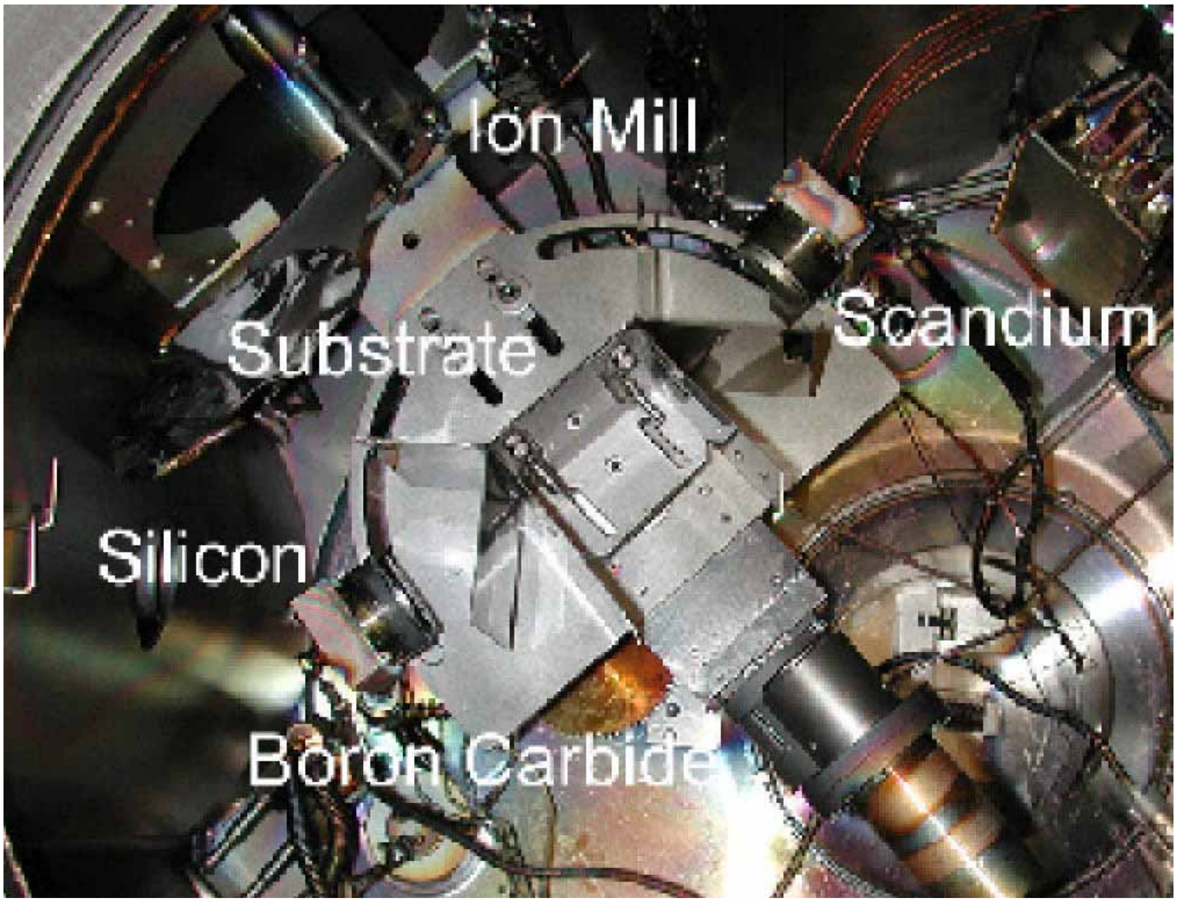

Fig. 2. Diagram of Sc/Si multi-layer mirrors with B_4C barrier layer.

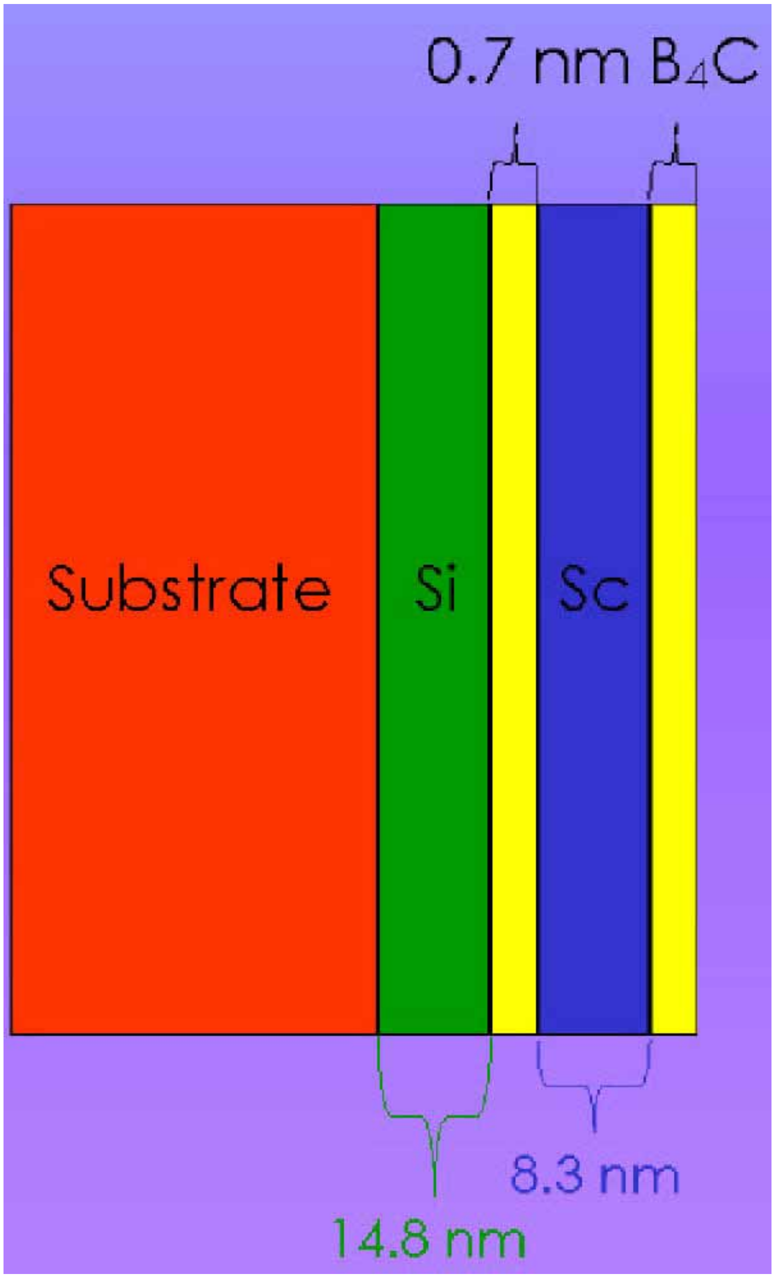

Fig. 3. Contact mode deflection image taken of sample No. 320 with the B_4C barrier layers using the AFM.

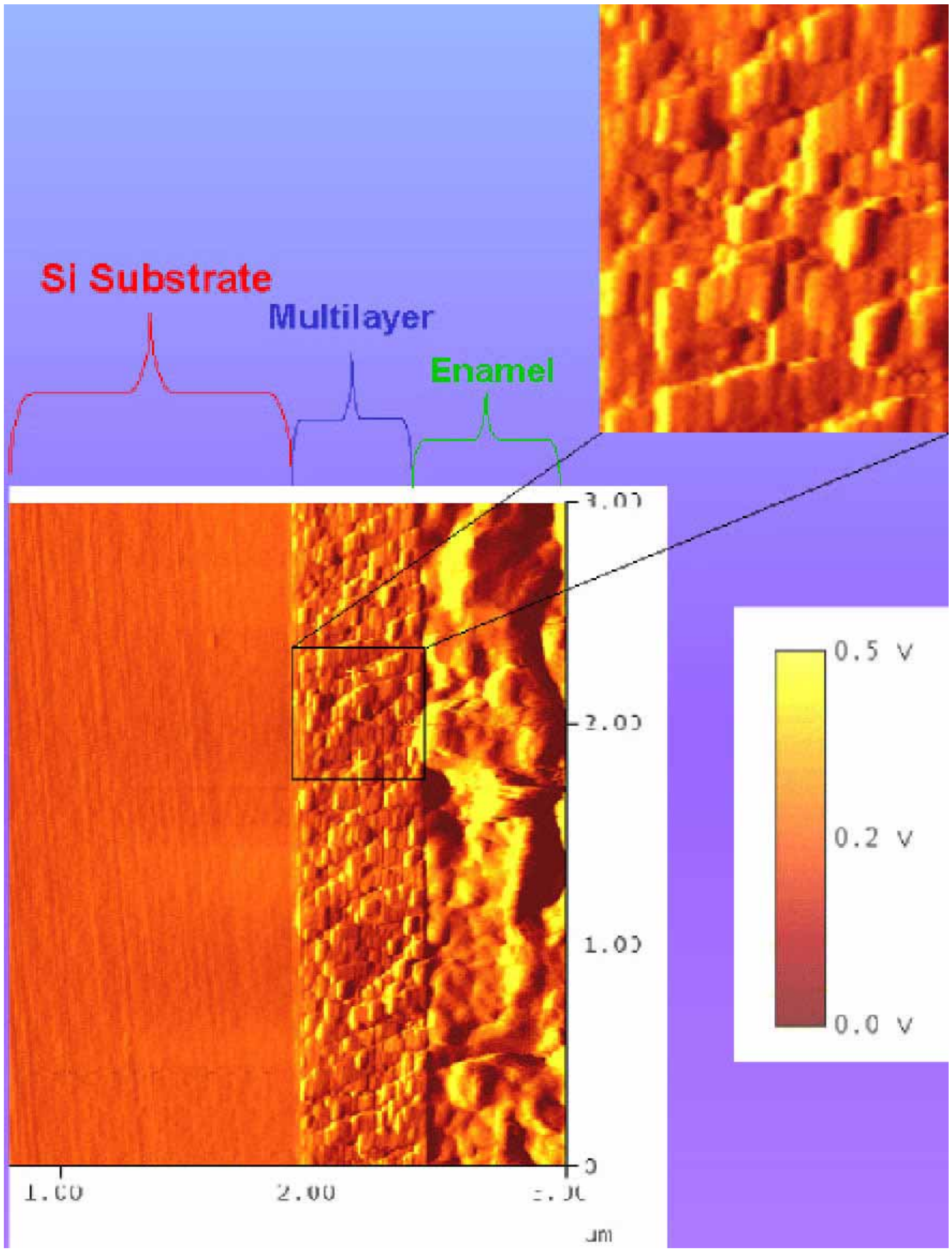

Fig. 4. Experimental reflectivity from the ALS (45º incidence) for samples Nos. 319 and 320. These are the Sc/Si multi-layer mirrors without and with the B_4C barrier layers.

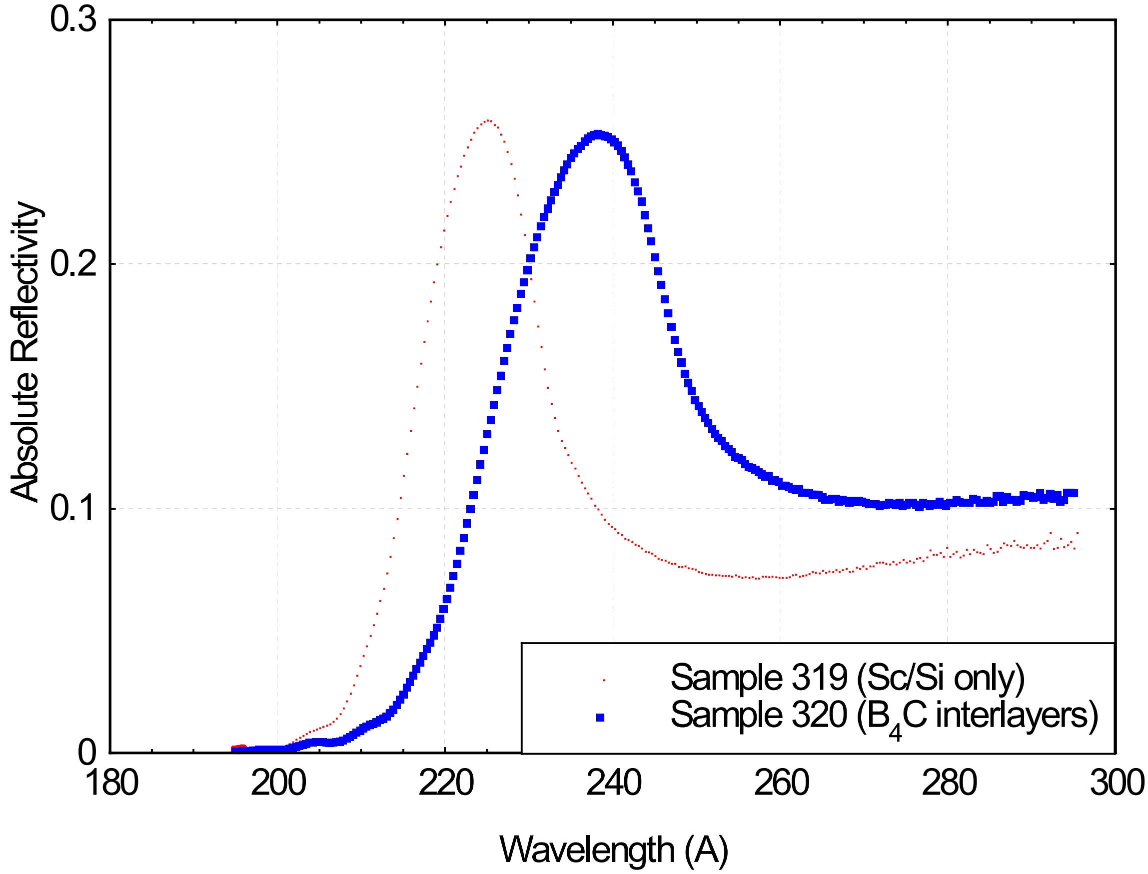

Fig. 5. Predicted reflectivity at normal incidence for samples Nos. 319 and 320 based on the fitted parameters. These are the Sc/Si multi-layer mirrors without and with the B_4C barrier layers.