Thin Solid Films 661 (2018) 65–70

Contents lists available at ScienceDirect

Thin Solid Films

journal homepage: www.elsevier.com/locate/tsf

Time-of-flight secondary ion mass spectrometry study on Be/Al-based multilayer interferential structures

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ARTICLE INFO

Keywords:
Be/Al-based multilayers
Magnetron sputtering
Depth profiling
Time-of-flight secondary ion mass spectrometry
Barrier layer
X-ray interferential mirror

ABSTRACT

Time-of-flight secondary ion mass spectrometry has been used for depth profiling of 530 nm-thick Be/Al-based multilayer interferential structures fabricated by magnetron sputtering for X-ray radiation at the wavelength of 17.1–17.5 nm. The introduction of ultra-thin (< 1 nm) Si barrier layers inside each period of ca. 8.9 nm decreased the thickness of interfaces between layers, increased the modulation factor of sputter depth profiles for both main elements and the relative intensity of Al_2^+/Be_2^+ secondary ions. For all structures with Si barriers, irrespective of the succession of the deposited layers, the improvement of the reflectance as compared with no-barrier Be/Al structure was revealed. Si/Al/Si/Be structure can be considered as the most prospective, but the realization of its potential requires further optimization of the width of Si barriers and the corresponding Si concentration in the multilayer structures.

1. Introduction

An introduction of ultra-thin barrier layers (BLs) between the layers of main elements in multilayer interferential structures (MIS) employed as X-ray mirrors has become commonly held manufacturing procedure (see, e.g., [1] and references cited therein). It was established [1] that BLs improve the reflectance (*R*-factor) of mirrors. However, the mechanism of this improvement can be multifarious because barrier layers are able: (i) to prevent the penetration of heavier element inwards lighter one in the course of MIS manufacturing and working [2], (ii) to decrease reactive diffusion and chemical interlayer interaction of main elements [3, 4], and (iii) to smooth out the interfaces between layers and to suppress their superfluous crystallization [5, 6]. The thickness of BLs should be as little as possible (usually, lower than 1 nm) because they must not cause significant extra-absorption of X-ray radiation. Changing of the mirror's reflectance and its spectrum dependencies, as well as the time stability of these characteristics under elevated working temperatures, serves as criterions for efficiency of BLs.

Generally, an influence of barrier layers on MIS characteristics is studied by means of small-angle X-ray scattering (SAXS) in combination with numerical methods for simulation and reconstruction of element depth profiles. For several years now, secondary ion mass spectrometry and X-ray photoelectron spectroscopy have become to be employed for such investigation due to significant improvement of sputter depth resolution of these techniques [3, 7–11]. Using time-of-flight secondary ion mass spectrometry (TOF-SIMS) and high-resolution transmission electron microscopy, we studied [11] the role of sub-nanosized (0.5 nm) carbon barrier layers in the fabrication of highly reflective La/B_4C-based mirrors used for near-normal incidence of X-ray radiation at $\lambda = 6.7$ nm. It has been shown that deposition of carbon BL on any boundary between layers increased the modulation of sputter depth profiles of main elements due to decreasing of the width of interface regions, and as a result, heightened the leap of electron density in these layers. Reactive diffusion supplemented and intensified by chemical interaction (for details, see [12] and references cited therein), was considered as the main mechanism for qualitative explanation of the

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https://doi.org/10.1016/j.tsf.2018.07.013
Received 26 January 2018; Received in revised form 12 July 2018; Accepted 12 July 2018
Available online 20 July 2018

obtained results, which have been in a good agreement with the direct reflectance measurements of La/B_4C-based mirrors and its simulation using SAXS data [13, 14]. It should be noted that the reactive diffusion is a nonequilibrium process of diffusion of atoms in solid structures consisting of several elements/components. The rate of this diffusion is determined not only by an initial concentration gradient of these elements/components, but also by chemical interaction between them [12].

In this work, we have used TOF-SIMS as a means of studying Be/Al-based mirrors with Si barrier layers. These mirrors have good prospects for solar astronomy of above 17 nm wavelength. The choose of barrier materials for MIS manufacturing is mainly determined by their coefficient of X-ray absorption in the given range of radiation. For the wavelengths nearby 17 nm carbon atoms possess higher absorption ability than silicon. And vice-versa, at $\lambda = 6.7$ nm, where La/B_4C mirrors are employed [11], carbon atoms have better X-ray transparency than silicon ones.

Recently, the *R*-factor of Be/Al-based mirrors with different location of silicon BLs between the layers of main elements has been measured in [15]. Although the theoretical reflectance of this MIS should be 78% for near-normal incidence of X-ray radiation at $\lambda = 17.14$ nm, only the maximum of 46% had been achieved in case of no-barrier Be/Al mirrors. At the same time, it was revealed that the mirror's reflectance increased up to 61% after the introduction of Si barrier with a thickness lower than 1 nm on the top of Be layer in each period. The results of small-angle X-ray scattering showed lowering in the thickness of interface regions in Si-containing structures, mainly due to the decreasing of root-mean-square roughness, from 1.3 nm to 0.6 nm. The present work purposes to carry out TOF-SIMS depth profiling of Be/Al-based multilayer interferential structures with Si barrier layers deposited on different interfaces between layers and on the basis of these results to understand an influence of silicon BLs on the characteristics of mirrors.

2. Experimental details

The Be/Al-based multilayer interferential structures were fabricated by magnetron sputtering on Si substrate in a specially certificated laboratory at IPM RAS since beryllium is a very toxic metal. The detailed information about the manufacturing process can be found elsewhere [15, 16].

The structures represent of ca. 530 nm-thick stacks consisting of 60 periods: Al/Be in case of T1 (no-barrier MIS), Al/Si/Be for T2, Be/Si/Al for T3, and, finally, Si/Al/Si/Be for T4 structure (the succession of layers is indicated from Si substrate upward the structures). It should be noted that for T4 structure the last Be and Si layers (on the surface of this structure) were not deposited. A schematic view of T1-T4 structures is shown in Fig. 1, and the data on the width of their periods and layers' thicknesses are presented in Table 1. The values of the layers' thicknesses were estimated from the SAXS data with an uncertainty of ± 0.05 nm [15]. The Si barriers are ultra-thin layers with a nominal thickness ranged within 0.6–0.8 nm. This technological parameter was estimated using the deposition and shutting rates of the magnetron sputtering processing. For all structures, the substrates are 0.4 mm-thick commercial Si (100) wafers with a mean-square surface roughness of 0.3 nm.

Sputter depth profiling of the structures was carried out at IPM RAS using a time-of-flight secondary ion mass spectrometer TOF·SIMS-5 by ION-TOF (Műnster, Germany). The instrument operates in dual-beam mode employing O_2^+ ions for sputtering with a 45° incidence angle. A usage of oxygen ion-beam bombardment is universally recognized analytical approach in SIMS technique. It stimulates the ion yield of electropositive elements like Al and Be in our experiments. An energy/current of the sputter ion-beam was chosen to be 2 keV/600 nA for the complete depth profiling (from the surface of structures down to Si substrate) and 1 keV/250 nA for the detailed depth profiling of only 10 near-surface periods. The sputter ion-beam was scanned over an area of

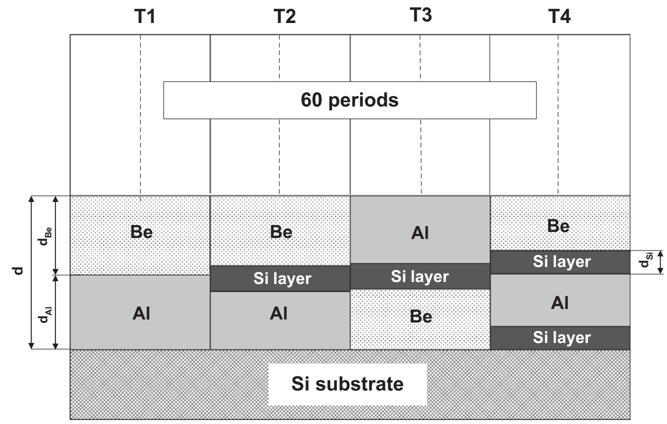

Fig. 1. Schematic view of Be/Al-based multilayer structures under study, not to scale. Layer thickness and width of periods are: $d \sim$ (8.75–8.9) nm, $d_{Si} \sim$ (0.6–0.8) nm, $d_{Be}/d \sim$ 0.39–0.55.

Table 1
Be/Al- based multilayer interferential structures.

Structure	Layers	Thickness (nm)				d_{Be}/d
		d	d_{Al}	d_{Be}	d_{Si}	
T1	Al/Be	8.8	4.9	3.9	–	0.44
T2	Al/Si/Be	8.75	3.25	4.8	0.7	0.55
T3	Be/Si/Al	8.9	3.65	4.45	0.8	0.5
T4	Si/Al/Si/Be	8.9	4.05	3.45	0.8/0.6	0.39

200 × 200 μm^2 in both depth profiling modes. The pulsed probing ion-beam was 25 keV/1 pA Bi^+ with a 45° incidence angle. The analyzed region was 40 × 40 μm^2, that is 4% in square around the center of sputter crater.

Depth profiles were collected in three different points for each MIS studied. Since the surface inhomogeneity of the structures is not > 1% in area of 50 × 50 mm^2, which is an important technological requirement for X-ray interferential mirrors operated with wide beams, the results obtained in the different points were very similar.

We also tested Cs^+ ions as a sputter-ion beam. However, in that case more intense and rapid development of surface roughness on the bottom of crater, as compare with oxygen ion-beam bombardment, resulted in the degradation of depth profiles of both main elements. In our opinion, considerable difference in mass between Be, Al, and Si, the elements of MIS, and Cs^+ sputter ions can stimulate ballistic knock-in process and intermixing of MIS elements into the bottom of crater causing excessive surface roughness. At the same time, for MIS with at least one heavy element like Mo/Si or La/B_4C, Cs^+ ions provide better sputter depth profiling as compared with O_2^+ ions [17].

3. Results and discussion

Complete depth profiles of Al_2^+, Be_2^+ and Si^+ secondary ions measured for no-barrier T1 structure is shown in Fig. 2. As the characteristic ions for main elements, we selected dimers of Al and Be since the atomic ions were very intense and their registration resulted in the saturation of a detector. Si^+ secondary ions shown in this figure are impurities originated from substrate material or introduced into MIS during its fabrication. Their intensity is on the level of noise, i.e. approximately equal or even lower than 10 cps over the total depth of the structure.

In Fig. 2 and in all following figures, the time of sputtering was recalculated into the depth of sputtering assuming constant average sputter rate. This assumption is valid for the total depth of structures. At the same time, inside each period the dependence between the time and