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# Reflective multilayer optics for 6.7 nm wavelength radiation sources and next generation lithography

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ABSTRACT

Reported is a computational and chemical analysis of near normal incidence reflective multilayer optics for 6.7 nm wavelength applications in e.g. the Free Electron Laser FLASH and next generations of EUV lithography. We model that combinations of B or $B_4C$ with La offer a reflectivity of ~70%. The small reflectivity bandwidth poses problems in applications, but it can be significantly improved by replacing La with Th or U. Grazing incidence X-ray reflectometry, cross-section TEM, and in-depth XPS analysis of B/La and $B_4C$/La multilayers reveal chemical reactivity at the interfaces. Significant $LaB_x$ interlayer formation is observed in especially B/La multilayers, stressing the relevance of interface passivation. We propose nitridation of the interfaces, which mitigates interlayer formation and simultaneously increases optical contrast.

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We present a computational and experimental survey of multilayer optics to maximize reflectivity around the 6.7 nm range at near normal angle of incidence (AOI). New short wavelength radiation sources, e.g. Free Electron Lasers (X-FELs) and next generations of Extreme UV lithography call for high reflectivity optics, in most cases preferably at near normal AOI [1].

Fig. 1 shows some reported reflectivity values ($R$) in the 2 to 20 nm wavelength and 0° to 5° AOI range [2]. We will here concentrate on optics for $\lambda = 6.7$ nm, the position of the boron K absorption edge, where B and $B_4C$ can act as low absorbing spacer layers. Several studies have shown that, compared to $B_4C$/Mo, $B_4C$/La multilayers offer significantly better reflectivity at relatively grazing AOI [3–5], with applications in e.g. soft X-ray spectroscopy, fluorescence analysis and imaging.

Fig. 2 shows the calculated reflectivity curve at 1.5° off normal AOI for the best candidate quarter-wave multilayers, assuming a 0.3 nm interface diffuseness which is conventional fur Mo/Si multilayer optics in EUV lithography [6]. Somewhat inferior candidates include B and $B_4C$ combinations with e.g. CsI and $LaF_3$. The number of periods is 200, the effective maximum that contributes to reflectivity. IMD simulations [7] show that the reflectivity bandwidth is only ~0.06 nm for the $B_4C$/La multilayer, which is ~11% of the Si/Mo multilayer reflectivity bandwidth for 13.5 nm EUV. When B or $B_4C$ is combined with Th or U, simulations indicate significantly larger bandwidths of 0.09 and 0.17 nm respectively, due to the higher optical contrast at the interfaces. AFM and TEM studies on Al/U and Si/U multilayers have revealed that these systems yield an interface diffuseness of ~0.5 nm [8]. Radioactivity hazard of Th or U is calculated to be of no significance in mirror applications [9,10]. Most multilayer deposition setups do however not accommodate experimental research with these metals and we will therefore here consider only experimental B/La and $B_4C$/La multilayers.

A reflectivity of 53% at 65.5° off normal AOI with 50 periods [3] and 39.3% at 75.0° off normal AOI with 150 periods [11] has been reported at $\lambda = 6.77$ nm. We can model these results with a $B_4C$/La bilayer structure with interface diffuseness up to 0.75 nm, which might be attributed to polycrystallinities. We will show that the high chemical activity of La with B and C results in significant interlayer formation, explaining the experimental data.

The presence of nano-size crystallites [12] was investigated with grazing incidence X-ray reflectometry (GIXR) spectrum at $\lambda = 0.154$ nm (Cu-$K_\alpha$) for two B/La and a $B_4C$/La multilayer that were produced with our multilayer deposition facilities [13], as shown in Fig. 3. The inset in Fig. 3 shows the cross-section transmission electron microscopy (CS-TEM) image of the $B_4C$/La multilayer.

The GIXR peaks at 27.3°, 56.3°, and 77.2° in Fig. 3 can originate from crystalline La <100>, La <200>, and La <210> planes respectively. The relative shifts of up to 5% for especially the non $B_4C$-containing multilayers would however indicate considerable lattice stress or distortions due to formation of broad $LaB_6$ interlayers at the reactive interfaces. The absences of 2nd order peaks in the spectra suggest an amorphous structure for all three multilayers with a general nearest neighbor distance only. Also CS-TEM reveals no crystallinity in the multilayers, but we can observe an increasing lateral waviness of the layers from bottom to top. This is likely caused by relaxation of the lattice stress due to the significant lattice parameter differences

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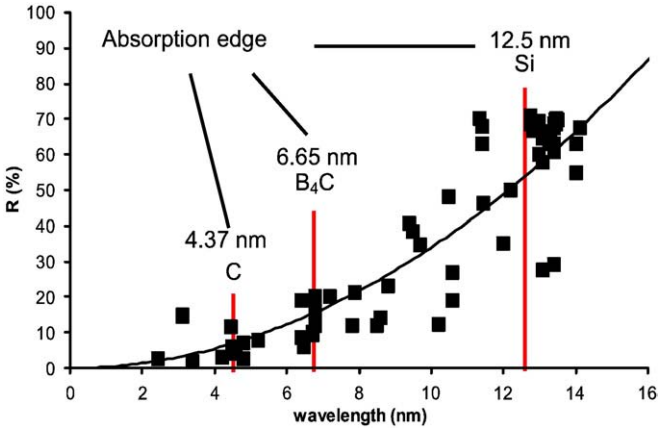


**Fig. 1.** Reported *R* for 0° to 5° off normal AOI. C, $B_4C$ and Si absorption edges are indicated.

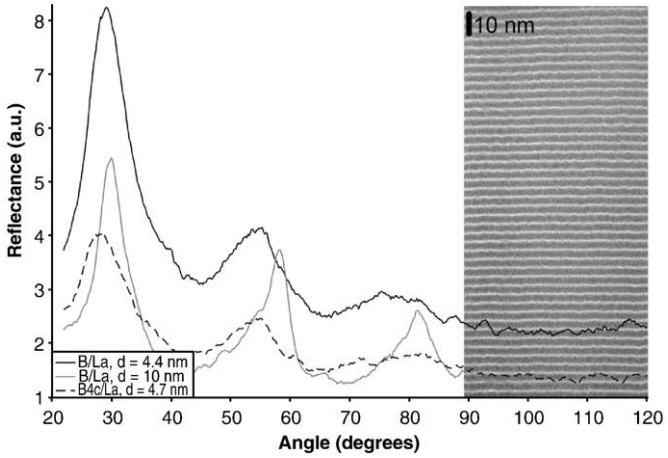


**Fig. 3.** GIXR at $\lambda = 0.154$ nm (Cu-$K_\alpha$) of three multilayers. The inset shows the CS-TEM image of the $B_4C$/La multilayer.

[14]. We do observe La crystallization of 7 nm thick La layers in CS-TEM. The $B_4C$-on-La interfaces appear to be more diffuse than La-on-$B_4C$ interfaces, attributable to the lower surface free energy of La, compared to B and $B_4C$ [15–17].

GIXR measurements of the $B_4C$/La multilayer suggest a ~0.67 nm interface diffuseness [15,18,19] or up to 1.5 and 0.5 nm thick kinetically favorable $LaB_6$ and $LaC_2$ interlayers at the $B_4C$-on-La and La-on-$B_4C$ interface resp. In-depth X-ray photoelectron spectroscopy (XPS) analysis of Mo/Si multilayers with $B_4C$ diffusion barriers reveal a B-rich stoichiometry, while C appears to be more diffused and weakly bounded [20]. The experimental results show reactive interfaces when $B_4C$ is used in multilayer applications, and stable metal borides and carbides appear indeed to be favored over $B_4C$. Metal boride formation at the cost of $B_4C$ has also been observed by P. Mogilevsky et al. [21].

With in-depth XPS analysis by 0.5 kV $Ar^+$ sputtering, we observe considerable intermixing of B and $B_4C$ with La, as can be seen in Fig. 4 for the B/La and $B_4C$/La multilayer with a *d*-spacing of 4.4 and 4.7 nm respectively. The multilayer profiles shown in Fig. 4 can be reconstructed using the mixing-roughness-information depth (MRI) model [22–25] with a calculated atomic mixing ($g_w$) of 2 nm and an information depth ($g_\lambda$) of 3 nm in linear approximation. We model a bilayered structure with 1.2 nm thick B and La layers for the B/La, and 2.8 nm thick $B_4C$ and 1.6 nm thick La layers for the $B_4C$/La multilayer. The reconstruction does not suggest significant interface gradients, although we observe a second La4d doublet at ~3.5 eV lower binding energy. Although XPS literature on La is very limited, this doublet might be attributed to $LaB_6$ and $LaC_2$ that are diffusely distributed throughout the multilayers. Taking also GIXR, CS-TEM, angular resolved XPS measurements [26,27], formation enthalpy ($\Delta H^{for}$) and optical constants as shown in Table 1 into account, we model an interface gradient that can be attributed to formation of $LaB_6$ and $LaC_2$, predominantly at the La layer front. This suggests atomic deposition of B and C [28], since incorporation of the dissociation of $B_4C$ in the reaction via $7La + 6B_4C \rightarrow 4LaB_6 + 3LaC_2$ practically cancels out any energy gain. La deposition onto an already recombined $B_4C$ substrate

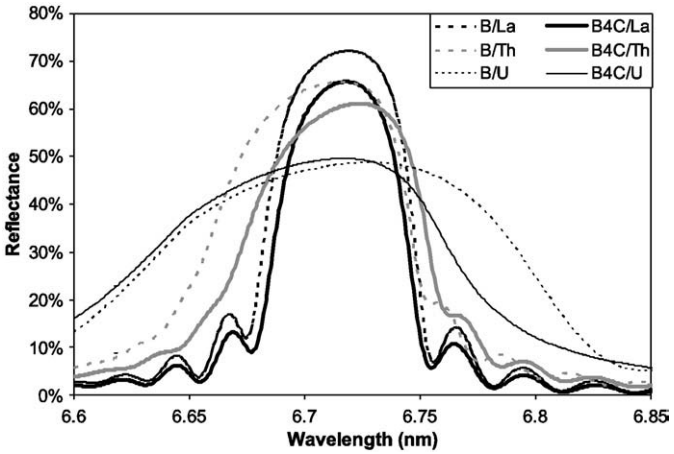


**Fig. 2.** Calculated reflectivity curves of the best performing 200 period multilayers at 1.5° off normal AOI, assuming 0.3 nm interface diffusiveness.

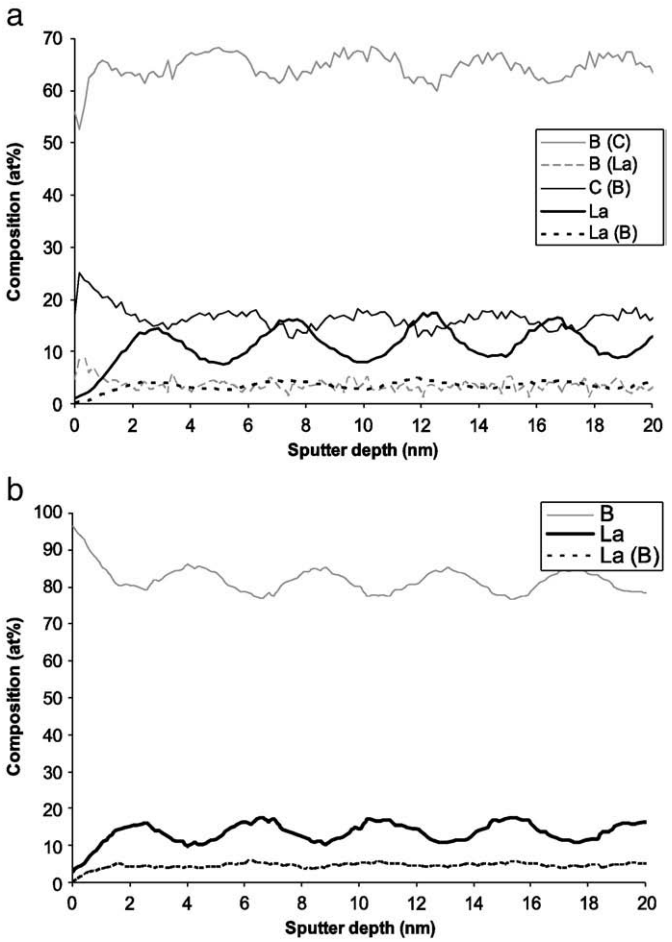


**Fig. 4.** In-depth XPS analysis of a B/La (a) and $B_4C$/La (b) multilayer, with 4.4 and 4.7 nm *d*-spacing respectively. In brackets are the species to which they are likely bonded.

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