

Reflectance of La/(C/B/C) Reflective Multilayers

Ryuki NAKANO¹, Tadashi HATANO^{2,3}, and Takeo EJIMA^{2,3,*}

¹Dept. Appl. Physics, ²SRIS, and ³IMRAM, Tohoku University
2-1-1 Katahira, Aoba-ku, Sendai, Miyagi 980-8577, Japan

1 Introduction

The beyond extreme ultraviolet (BEUV) wavelength region around 6.x nm has been proposed for next-generation semiconductor lithography systems [1]. Because BEUV light is strongly absorbed by matter, projection optics require reflective multilayer mirrors. Such mirrors are typically fabricated by periodically stacking two materials with low absorption and a large refractive-index contrast at the operating wavelength. Since the multilayer period is approximately half the operating wavelength, interfacial abruptness becomes increasingly critical as the wavelength decreases.

We have investigated La/B₄C multilayers as reflective multilayers operating near 6.x nm. La/B₄C was selected because it was expected to provide high reflectance in the BEUV region, and because B₄C has a markedly low standard enthalpy of formation [2], while metallic La is thermodynamically stable in its elemental state [3]. Abrupt interfaces were therefore anticipated. However, our previous investigation of ion-sputtered La/B₄C multilayers indicated that, under the deposition conditions examined, B₄C was likely decomposed at the interface with La. Consequently, the La layers were likely transformed into La–B compounds, while the B₄C layers were converted into B_xC_y ($x < y$) [4].

For magnetron-sputtered La/B₄C multilayers, inserting a C layer between La and B₄C has been reported to prevent direct contact and act as a diffusion barrier [5]. In that study, a broad intermixed layer formed at the La-on- B₄C interface, but C-layer insertion suppressed the intermixed-layer width to within the C-layer thickness of 0.25–0.3 nm, leading to La/C/B₄C multilayers with a reflectance of 0.58. Based on these findings, we attempted to fabricate ion-sputtered multilayers with C interlayers inserted between the La and B layers.

2 Design, Fabrication, and Evaluations

The constituent materials for the reflective multilayer at the design wavelength were selected on the basis of a complex-plane plot of the Fresnel coefficients (Fig. 1). The selection criteria were as follows: (1) the Fresnel coefficients should be close to the real axis, corresponding to low absorption in both materials, and (2) the two materials should be well separated in the complex plane, corresponding to a large refractive-index contrast between them. As shown in Fig. 1, La and B satisfied these criteria. We further considered that the insertion of an interlayer that does not significantly reduce the reflectance could suppress the reactivity of the La layer while maintaining high reflectance. Based on the results described above, C was selected as the interlayer material because its reaction

with La was relatively limited. Therefore, the interfaces of the La layers were covered with C layers. The designed multilayer structure was [La 1.72 nm/(C/B/C) 1.72 nm]×150.

The samples were deposited using an ion-beam sputtering system (TOYAMA MST-4). La, B, and C targets with purities of 99.9%, 99%, and 99.9%, respectively, were used. During deposition, the Ar⁺ acceleration voltage was 1 kV, and the vacuum pressure was 2.3×10^{-3} Pa. The sputtering rates were 2.271 nm/min for La, 0.3352 nm/min for C, and 0.1401 nm/min for B. The fabricated samples were La/(C/B/C) multilayers with 150 periods deposited on Si wafers. The thickness of the La layer was set equal to that of the C/B/C layer, and the thickness ratio of the C, B, and C layers within the C/B/C stack was 1:37:1. The multilayer period was determined by X-ray reflectivity. The obtained periods were 3.45 nm for sample A, 3.56 nm for sample B, 3.78 nm for sample C, and 3.83 nm for sample D. Reflectance measurements were performed using the reflectometer installed at BL-12A. The S path was used for the measurements, with incident and exit slit widths of 0.2 mm and 0.1 mm, respectively.

3 Results and Discussions

Figure 2(a) shows the angular dependence of the reflectance measured for each sample at a fixed incident wavelength of 6.80 nm. The reflectance peak position depended on the periodic length of the multilayer: as the periodic length increased, the peak shifted toward higher angles and became narrower. Reflectance calculations were then performed by varying the periodic length and interfacial roughness to reproduce the measured peak positions and heights of measured reflectance (Fig. 2(b)) [6]. In the calculations, the thicknesses of the La and C/B/C layers were set to be equal for all samples, while the incident wavelength and the number of periods were fixed at the experimental values. The optical constants included in IMD were used. The interfacial roughness were varied to reproduce the peak height, assuming the same roughness value for all interfaces. An error-function profile was used to describe the interfacial roughness.

Comparison between the measured and calculated results showed that the periodic lengths obtained from the reflectance calculations were 0.02–0.03 nm larger than those evaluated by X-ray reflectivity, and the interfacial roughness was estimated to be 1.02–1.04 nm. This roughness exceeded the thickness of the C layer and was comparable to that of the B layer. In the present calculations, the interfaces were modeled by assuming a continuous variation in the refractive index over a region comparable to, or larger than, the individual layer thickness.

The obtained values therefore suggest that uniform bulk-like regions were not sufficiently formed within the individual layers and that the layers may largely behave as intermixed regions with the adjacent materials. For comparison, Chkhalo et al. reported that, in La/B₄C multilayers, the intermixed-layer width was approximately 0.75 nm at the La-on-B₄C interface and approximately 0.35 nm at the opposite B₄C-on-La interface [5]. These values indicate that interfacial reaction or intermixing is particularly likely to occur at the La-on-B₄C interface, where La is deposited onto a B₄C layer.

On the other hand, the measured reflectance peak widths were more than twice the calculated values. This result suggests that the deposition rates of the individual layers changed during film growth, resulting in a multilayer with a nonuniform period. Since the present calculations do not include period nonuniformity, the interfacial roughness estimated from the fitting may be overestimated if the effect of period distribution is taken into account.

4 Summary

In this study, La/(C/B/C) multilayers with C interlayers inserted between the La and B layers were fabricated and evaluated. Fresnel-coefficient-based material selection confirmed that La and B are a promising material pair because of their low absorption and large refractive-index contrast. Comparison of the experimental reflectance with calculated values indicated an interfacial roughness of 1.02–1.04 nm, which exceeds the thickness of the C layer and is comparable to that of the B layer. In addition, the measured reflectance peak widths were more than twice the calculated values, suggesting the presence of period nonuniformity caused by changes in the sputtering rates during deposition. Therefore, suppressing interfacial reactions and intermixing at the La interfaces, stabilizing the sputtering rates during long deposition processes, and improving the period uniformity are essential for achieving

high-reflectance La/(C/B/C) multilayers in the BEUV wavelength region.

References

- [1] D. Kuznetsov, A. Yakshin, J. Sturm, R. van de Kruijs, E. Louis, and F. Bijkerk, *Opt. Lett.* **40**, 3778–3781 (2015).
- [2] M. W. Chase Jr., *J. Phys. Chem. Ref. Data*, Monograph **9**, 1–1951 (1998).
- [3] R. H. Schumm, D. D. Wagman, S. Bailey, W. H. Evans, and V. B. Parker in National Bureau of Standards (USA) Technical Notes, 270 (1973).
- [4] T. Ejima, *Jpn. J. Appl. Phys.* **65**, 050802 (2026).
- [5] N. I. Chkhalo, S. Künstner, V. N. Polkovnikov, N. N. Salashchenko, F. Schäfers, and S. D. Starikov., *Appl. Phys. Lett.* **102**(1), 4–7 (2013).
- [6] D. L. Windt, *Computers in Physics* **12**(4) 360–70 (1998).

*takeo.ejima.e7@tohoku.ac.jp

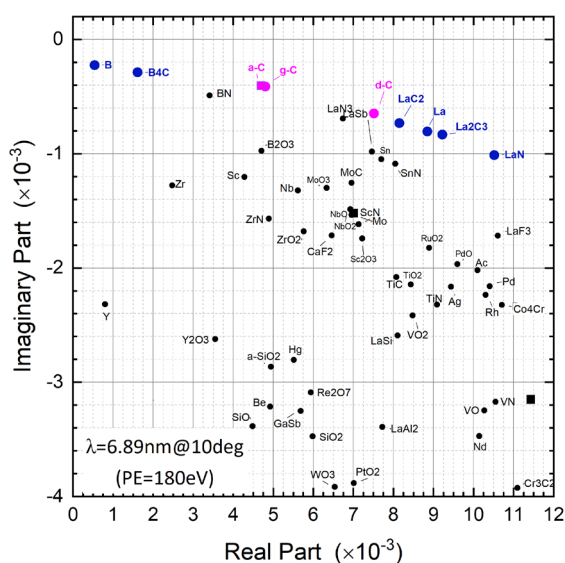


Fig. 1: Fresnel coefficient map at wavelength 6.89 nm and normal incidence angle of 10°. Carbon related materials exist between La and B series.

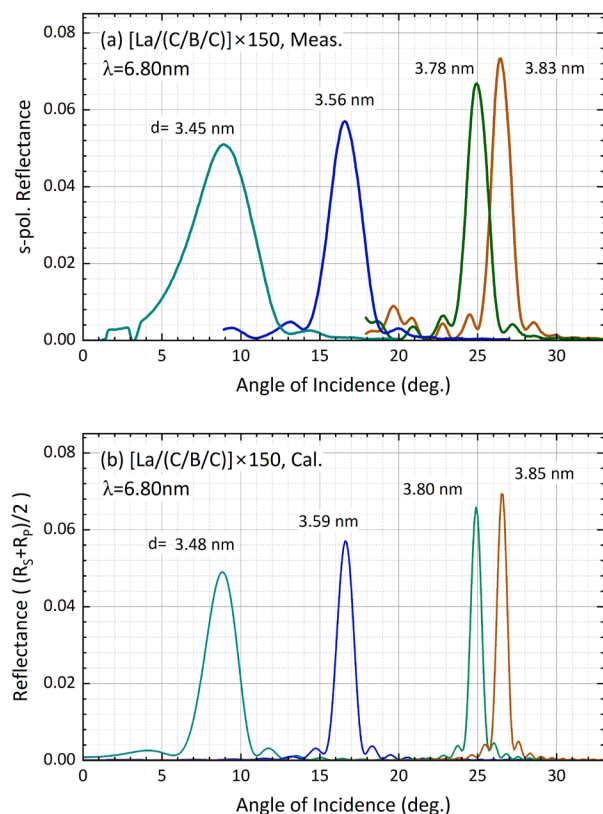


Fig. 2: Measured and calculated reflectance of La/(C/B/C) multilayers: (a) measured reflectance and (b) calculated reflectance. The calculations were performed to reproduce the measured peak positions and peak heights.