

Multilayer Coating for Extreme Ultraviolet Experiments

NTT Advanced Technology Corporation, Atsugi, Kanagawa, Japan

E-mail: nano-sales@ml.ntt-at.co.jp

Web: <https://keytech.ntt-at.co.jp/>

Abstracts: *Extreme ultraviolet (EUV) multilayer coatings have been designed and fabricated for kinds of experiments including high-order harmonics application. In this paper, we present recent result obtained with Ru/B₄C, Mo/Si, Zr/Al-Si, SiC/Mg multilayer coatings, at corresponding wavelength region of 8 nm – 50 nm. These multilayer coatings can be easily customized depended on experimental requirements, including the incident angle, peak wavelength, and bandwidth.*

1. Introduction

Extreme ultraviolet (EUV) lights are powerful tool not only for basic research fields but also industrial fields. High order harmonics and x-ray free electron lasers (XFELs) have been used for various kinds of ultrafast and high-intense optical experiments. For industrial fields, plasma based light sources have realized EUV lithography and EUV inspection as high-volume manufacturing tools. Since the pioneering demonstration of EUV multilayer in 1976 [1], research and developments of multilayers have been progressed. Especially, a Mo/Si multilayer realized the peak reflectivity of more than 60% at wavelength of 13 nm region [2], and Mo/Si based coating for high-thermal tolerance [3-5], narrow bandwidth and broad bandwidth have been demonstrated. In addition, many multilayers such as Mg and Sc based multilayers for >25 nm region [6, 7] and Mo based multilayer for 8 nm – 12 nm wavelength [8-10] have been reported.

In this paper, we report recent demonstration of our EUV multilayer coatings for wavelength region of 8 nm – 50 nm. A Ru/B₄C multilayer optimized for incident angle of 45 degrees has a possibility of using as a reflective polarizer for investigating magnetic material properties. Around 13.5 nm, high reflectivity Mo/Si, narrowband MoSi₂/Si and broadband Ru/Si multilayers will work for EUV lithography related applications. For longer than 17 nm region, narrowband Zr/Al-Si and SiC/Mg multilayers can separate a single-order harmonic spectra from high-harmonics discrete spectrum, and broadband Mo/Si multilayers will be used for attoscience experiments.

2. Multilayer Coatings for 8–12 nm region

As coating materials for 8–12 nm multilayer, Ru and B₄C were selected. Fig. 1(a) – (c) show the calculated reflectivities of Ru/B₄C multilayer comparing with other material combinations. Each multilayer is optimized for peak wavelength of (a) 8 nm, (b) 10 nm and (c) 12 nm, respectively, and for incident angle of 0 degree. Note that the calculation assuming no surface oxidation and no inter-layer roughness, and assuming bulk optical constants. As shown these figures, although Pd/B₄C multilayers have slightly higher reflectivity at 8 nm and 10 nm region than those of Ru/B₄C multilayers, at 12 nm region, a Ru/B₄C has higher reflectivity. It is because that although the absorption coefficient of Pd increases at longer wavelength, that of Ru is still low at 12 nm region. It means that Ru/B₄C multilayers are more flexible for coating designs. In addition, the material price of Ru is more stable than that of Pd. Thus, we selected Ru and B₄C as

a multilayer material combination for 8 nm – 12 nm wavelength region although Pd/B₄C based multilayer coating has been demonstrated [11].

Fig. 2 shows measured reflectivity profile of a Ru/B₄C multilayer. The reflectivity was measured at Advanced Light Source BL 6.3.2, Laurence Berkeley National Laboratory [12]. It shows good agreement with a calculated profile assuming interlayer roughness of 0.4 nm. The peak reflectivity of 33% is comparable with a conventional report of a Pd based multilayer [11]. This 45-degree multilayer has p-polarization reflectivity of ~0.1%, and the multilayer will work not only as a steering mirror or a focusing mirror but also as a reflective polarizer.

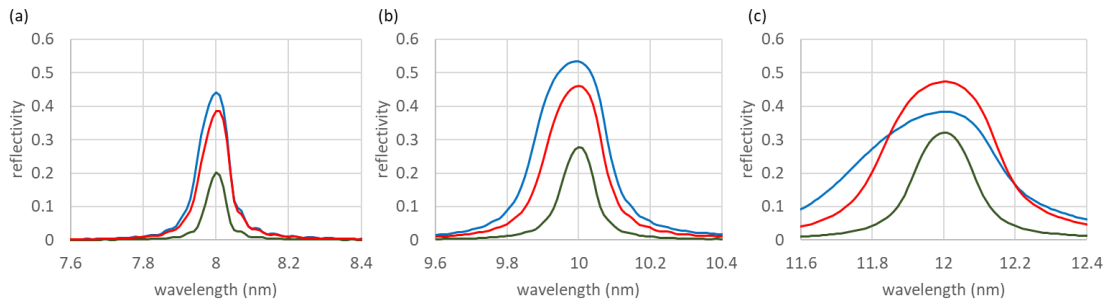


Fig. 1 Calculated reflectivity profiles of Pd/B₄C (blue line), Ru/B₄C (red line) and Mo/B₄C (green line) multilayers designed for (a) 8 nm, (b) 10 nm and (c) 12 nm, respectively.

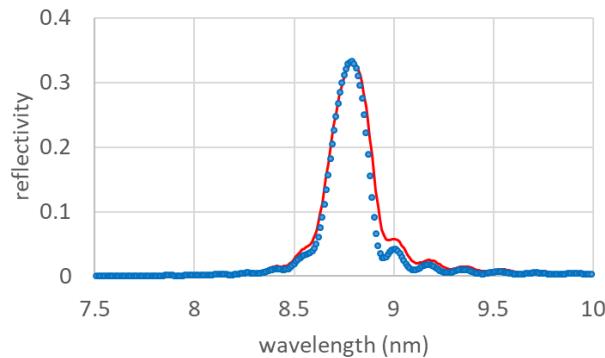


Fig. 2 A measured reflectivity profile of a Ru/B₄C multilayer (blue dots) designed for incident angle of 45 deg. and peak wavelength of 8.8 nm, Red line is a calculated profile assuming inter-layer diffusion of 0.4 nm.

3. Multilayer Coatings for 13.5 nm region

For wavelength region of longer than Si L-edge (~12.4 nm), Si has been used as a low index material of multilayer. A Mo/Si multilayer is one of most standard multilayers for 13.5 nm region. The mirrors are used in many kinds of applications such as EUV lithography, astronomy and attosecond science. Figure 3 is a measured reflectivity of a standard Mo/Si multilayer mirror we fabricated. The reflectivity reaches to almost 70% at normal incident angle and ~65% at incident angle of 45 degrees. A capping layer works for surface protection from oxidization, contamination, and ion irradiations [3, 4], and barrier layers work for increasing thermal

tolerance of multilayer [5]. We have several materials for capping layer and barrier layer, such as Ru, B₄C, C, TiO₂ and SiO₂.

MoSi₂/Si multilayer mirror has narrow-bandwidth around 13.5 nm region, and Ru/Si multilayer mirror has broad bandwidth in this wavelength region. In Fig. 4(a) and (b), calculated reflectivity profiles of a narrowband MoSi₂/Si multilayer and a broadband Ru/Si multilayer are shown comparing with those of Mo/Si multilayers. Although Mo/Si multilayers can be modified for narrowband and broadband reflections, MoSi₂/Si and Ru/Si multilayers show good properties for narrowband and broadband mirrors, respectively.

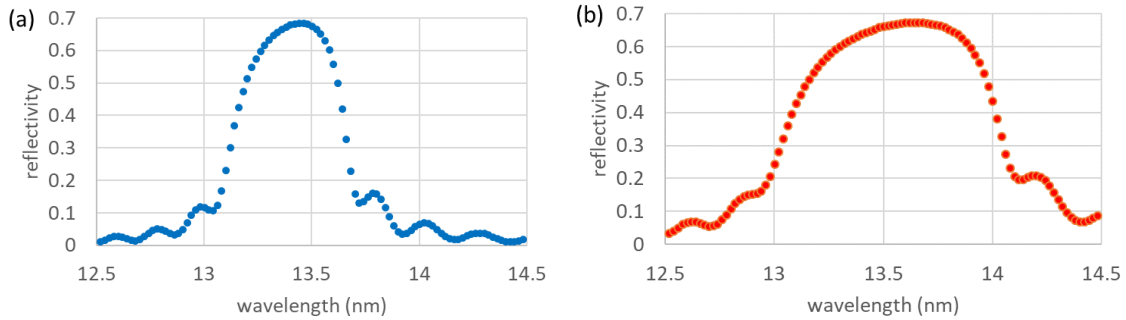


Fig. 3 Measured reflectivities of Mo/Si multilayers. (a) Incident angle of 6 deg. and (b) incident angle of 45 deg., respectively.

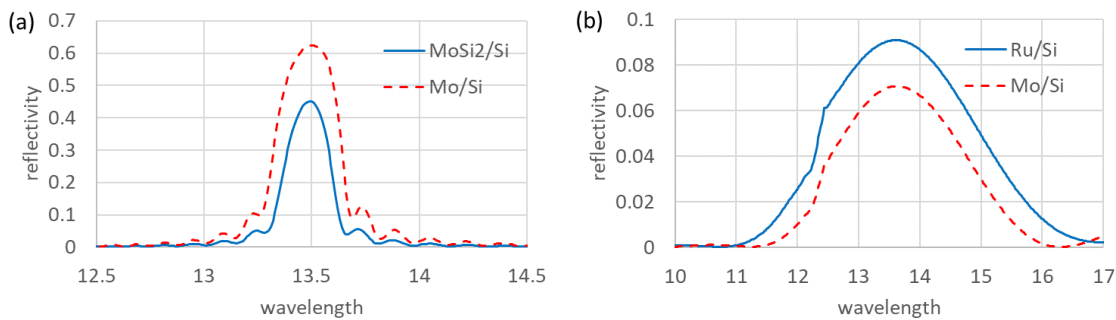


Fig. 4 Calculated reflectivity profiles of (a) a MoSi₂/Si narrowband multilayer and (b) a Ru/Si broadband multilayer. In the calculations, inter-layer roughness was assumed 0.7 nm. Mo/Si multilayer profiles are also shown by red dashed lines for references.

4. Multilayer Coatings between 17 nm and 25 nm

Al is a well-known as a low absorption material in wavelength longer than 17 nm. Al thin film filter is a standard optical component to separate high-order harmonics spectrum from a visible fundamental beam. For a synchrotron application, Al thin filters are installed to separate a 1st order diffraction from higher order diffractions of EUV gratings. On the other hand, there are limited investigations about multilayer coatings based on Al [13-15]. It is because that Al is easily oxidized and easily crystallized. Both oxidation and crystallization affect reflectivity decreasing of multilayers, thus, more stable Si based multilayers, especially Mo/Si multilayers have been used in this wavelength region. However, due to the absorption of Mo and Si, it is

difficult to obtain enough reflectivity by using these material combinations. We have demonstrated hypereutectic Al-Si based multilayer coatings to realize narrowband high reflectivity mirrors in wavelength region of 17 nm – 25 nm [16]. Hypereutectic Al-Si is well-known on mechanics engineering fields as a high thermal conductivity and a good strength material [17], and the EUV multilayer coatings are expected having advantages compering with pure-Al based multilayers.

Fig. 5 is calculated reflectivity profiles of Zr/Al-Si, C/Al-Si and Y/Al-Si multilayers, respectively. In the calculation, we select the Al-Si ratio as 0.7 and 0.3. The value is larger than the eutectic point (12.7% of Si). Calculated reflectivity profiles of Zr/Al-Si high-reflectivity, narrowband and broadband coatings are shown in Fig. 6 comparing with thots of Mo/Si multilayer coating. In the figure 45-deg. profiles are also shown.

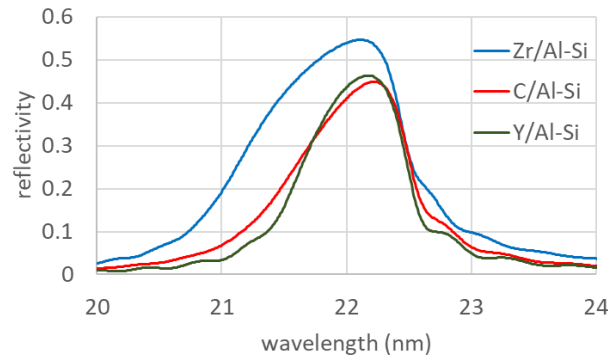


Fig. 5 Calculated reflectivity of Zr/Al-Si (blue-line), C/Al-Si (red line) and Y/Al-Si (green line), respectively.

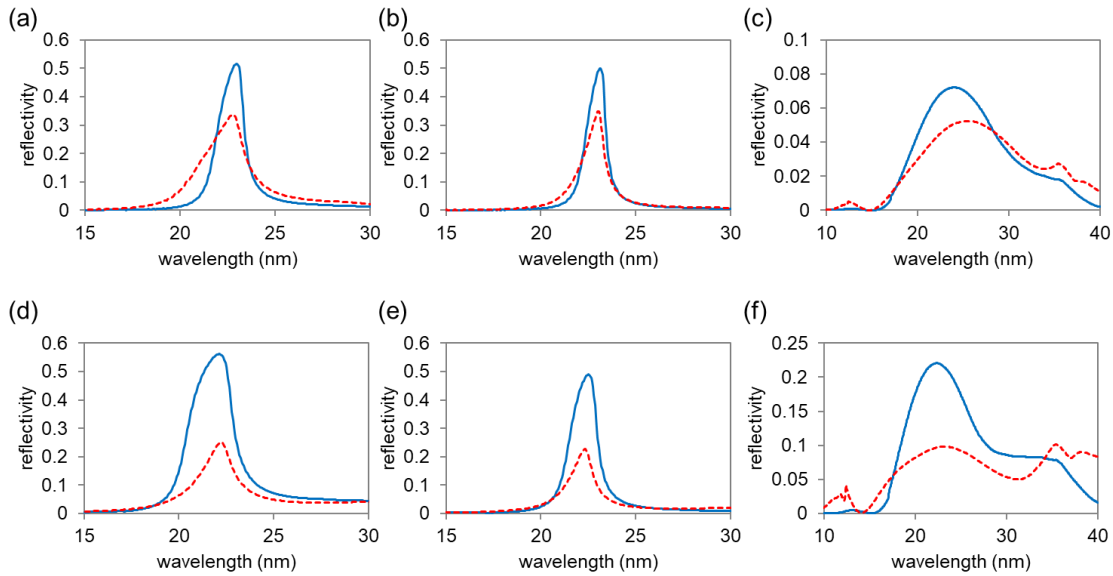


Fig. 5 Calculated profiles of (a) (d) high-reflectivity, (b) (e) narrowband reflectivity, and (c) (f) broadband reflectivity Zr/Al-Si (blue lines) and Mo/Si (red dashed lines) multilayers. (a), (b) and (c) are AOI=0 deg., and (d), (e) and (f) are AOI=45 deg., respectively.

Note that into these calculations in Fig. 5 and Fig. 6, ideal condition such as no-surface oxidation, no inter-layer roughness, and optical constants of bulk materials are assumed.

Measured reflectivities of a narrowband Zr/Al-Si multilayers at incident angle of 45 deg. are shown in Fig. 7, peak wavelength of (a) 18 nm and (b) 21 nm. Estimate surface oxidation and inter-layer roughness are 4 nm and 0.5 nm. The peak reflectivity and the full-width half maximum (FWHM) bandwidth of this narrowband mirror were 45% and 0.7 nm at wavelength of 18 nm, and 34% and 0.8 nm at wavelength of 21 nm, respectively.

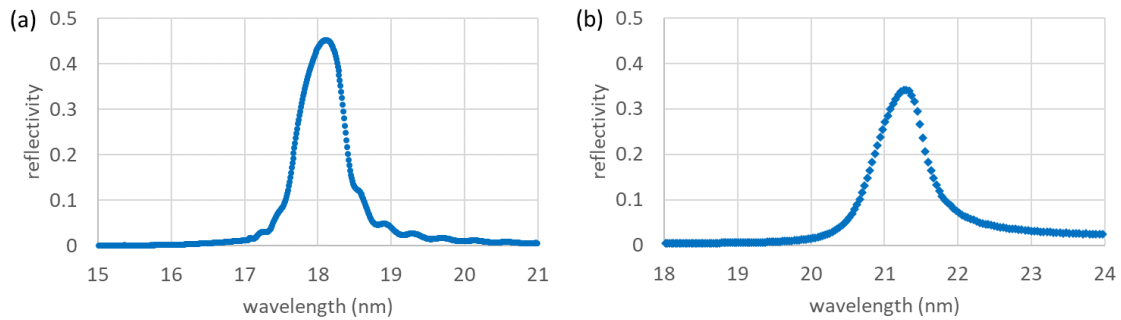


Fig. 7 Measured reflectivity of Zr/Al-Si multilayer designed for (a) 18 nm and (b) 21 nm at incident angle of 45 degrees.

5. Multilayer Coating for 25 nm – 50 nm region

Wavelength region of 25 nm – 50 nm is particularly important for high-harmonics, astronomy, x-ray laser, and other applications. Especially, by using high-harmonics, many kinds of basic material research such as attosecond dynamics measurements, angle-resolved photoelectron spectroscopy and two-photon absorption are demonstrated.

SiC/Mg multilayer coatings have been used for narrowband high-reflectivity mirrors [6]. Comparing other material combinations such as Mo/Si and B₄C/Mo/Si multilayers, the SiC/Mg multilayer has an advantage for narrow bandwidth. It is sometime strongly required from high-harmonics experiments, especially single-order separation. Fig. 8 is a measured reflectivity profile of a SiC/Mg multilayer mirror. The peak wavelength was 42.1 nm, peak reflectivity was 51%, and the FWHM bandwidth was 3.0 nm, respectively. The bandwidth (2.15 eV) is narrow enough to separate a single order harmonics spectra generated from 800 nm pump laser, which is discretely generated at 3.1 eV.

On the other hand, Mo/Si based broadband coatings are proposed and reported. To optimize the layer thicknesses, number of layers and a capping layer, the bandwidth and reflection range are controlled depended on the applications. Multilayers with two different periodic length stacks can broad the bandwidth, and a specific designed capping layer can extend the reflective tail to the total reflection range. In Fig. 9, designed reflectivity profiles of (a) two-blocks multilayer [18] and (b) a multilayer with tail enhanced capping layer are shown. These multilayers have limited number of layers (typically less than 20), and the layer structures are simple, thus, the spectral phase of these multilayers have almost linear. Therefore, these broadband multilayers can be used in a variety of applications requiring pulse duration measurements in the sub-100 as region [19].

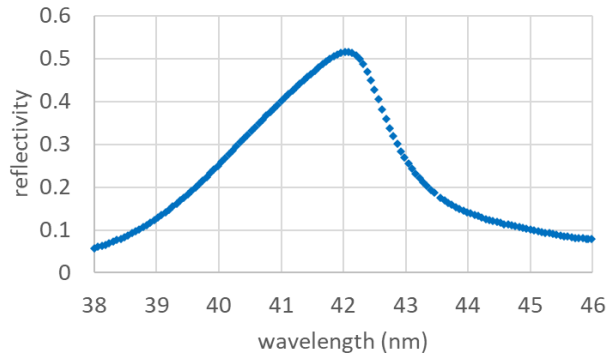


Fig. 8 Measured reflectivity of SiC/Mg narrowband multilayer. Incident angle is 6 deg.

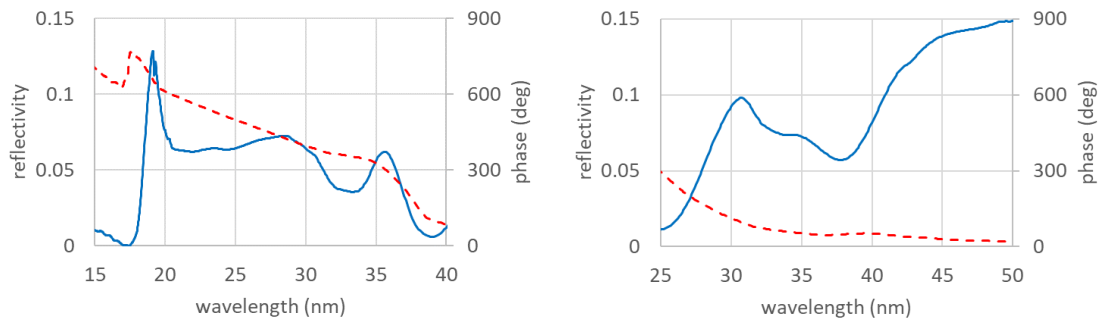


Fig. 9 Calculated reflectivity profiles (blue line) and reflective phase profiles (red dashed line) of Mo/Si based broadband multilayers, (a) a two-block multilayer and (b) a specified capping layer. Incident angles are 6 deg.

6. Conclusion

In this report, we demonstrated various kinds of multilayer coatings for high-reflectivity, narrowband reflectivity, and broadband reflectivity. For 8 nm – 12 nm region, Ru/B₄C shows good property, and around 13.5 nm region, Mo/Si, MoSi₂/Si and Ru/Si show various properties. For 20 nm region, Zr/Al-Si multilayers are useful and longer than 25 nm region, narrowband SiC/Mg and broadband Mo/Si will be selectable for depended on the applications. These multilayers are used in various research fields, such as high-order harmonics, astronomy, plasma physics and EUV lithography.

- [1] E. Spiller, "Reflective multilayer coatings for the far uv region," *Appl. Opt.* **15**, 2333 (1976).
- [2] T. W. Barbee, et al., "Molybdenum-silicon multilayer mirrors for the extreme ultraviolet," *Appl. Opt.* **24**, 883 (1985).
- [3] J. H. Underwood, et al., "Tarnishing of Mo/Si multilayer x-ray mirrors," *Appl. Opt.* **32**, 6985 (1993).

- [4] M. Singh and J. J. M. Braat, "Capping layers for extreme-ultraviolet multilayer interference coatings," *Opt. Lett.* **26**, 259 (2001).
- [5] H. Takenaka and T. Kawamura, "Thermal stability of Mo/C/Si/C multilayer soft X-ray mirrors," *J. Electron Spectrosc. Relat. Phenom.* **80**, 381 (1996).
- [6] H. Takenaka, et al., "Soft-x-ray reflectivity and heat resistance of SiC/Mg multilayer," *J. Electron Spectrosc. Relat. Phenom.* **144-147**, 1047 (2005).
- [7] M. Grisham, et al., "Damage to extreme-ultraviolet Sc/Si multilayer mirrors exposed to intense 46.9-nm laser pulses," *Opt. Lett.* **29**, 620 (2004).
- [8] K. M. Skulina, et al., "Molybdenum/beryllium multilayer mirrors for normal incidence in the extreme ultraviolet," *Appl. Opt.* **34**, 3727 (1995).
- [9] B. Sae-Lao and C. Montcalm, "Molybdenum-strontium multilayer mirrors for the 8–12-nm extreme-ultraviolet wavelength region," *Opt. Lett.* **26**, 468 (2001).
- [10] C. Montcalm, et al., "In situ reflectance measurements of soft-x-ray/extreme -ultraviolet Mo/Y multilayer mirrors," *Opt. Lett.* **20**, 1450 (1995).
- [11] D. L. Windt and E. M. Gullikson, "Pd/B₄C/Y multilayer coatings for extreme ultraviolet applications near 10 nm wavelength," *Appl. Opt.* **54**, 5850 (2015).
- [12] J. H. Underwood, et al., "Calibration and standards beamline 6.3.2 at the Advanced Light Source," *Rev. Sci. Instrum.* **67**, 3372 (1996).
- [13] D. L. Windt, and J. A. Bellotti, "Performance, structure, and stability of SiC/Al multilayer films for extreme ultraviolet applications," *Appl. Opt.* **48**, 4932 (2009).
- [14] E. Meltchakov, et al., "Development of Al-based multilayer optics for EUV," *Appl. Phys. A* **98**, 111 (2010).
- [15] Q. Zhong, et al., "Enhancement of the reflectivity of Al/Zr multilayers by a novel structure," *Opt. Express*, **21**, 14399 (2013)
- [16] M. Hatayama, et al., "Wide-range narrowband multilayer mirror for selecting a single-order harmonic in the photon energy range of 40–70 eV," *Opt. Express* **24**, 14546 (2016).
- [17] H. Ye, et al., "An overview of the development of Al-Si-Alloy based material for engine applications," *Eng. Perf.* **12**, 288 (2003).
- [18] M. Hatayama, et al., "Broadband extreme ultraviolet multilayer mirror for supercontinuum light at a photon energy of 35-65 eV," *Appl. Opt.* **48**, 5464 (2009).
- [19] H. Mashiko, et al., "Extreme ultraviolet supercontinua supporting pulse durations of less than one atomic unit of time," *Opt. Lett.* **34**, 3337 (2009).