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Aperiodic multilayer mirrors for efficient broadband reflection in the extreme ultraviolet

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Abstract Recent extreme ultraviolet sources using high-harmonic generation in a rare gas make new optics developments necessary. We report on the study and development of multilayer structures with efficient reflectivity in the 35–75 eV energy range. We have optimized, deposited and characterized two aperiodic broadband mirrors consisting of a Mo, Si and B_4C thin-film stack. We used the needle procedure in order to optimize mirror reflectivity. The magnetron sputter deposited multilayers have been calibrated and characterized using Cu K_α grazing incidence X-ray reflectometry. Reflectivity measured at near-normal incidence on a synchrotron radiation source reaches 12% with a full width at half maximum of nearly 40 eV. Experimental results are compared with theoretical simulation using available optical constants for Mo, Si and B_4C in this spectral range.

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1 Introduction

Optics in the extreme ultraviolet (EUV) field are difficult to design because of strong absorption at these wavelengths for most materials, but some theoretical work dealt with this [1]. However, periodic Mo/Si multilayers were found to have interesting properties, as reported by Barbee et al. [2]. These multilayers appear to have a maximal theoretical reflectivity of 75% at 13.5 nm for near-normal incidence. Due to recent interest in EUV lithography applications [3], a lot of work was done to improve efficiency of Mo/Si mirrors and approach the theoretical limit. Reflectivity values higher than 0.7 have already been reported [4]. One major point to address to improve reflectivity is the quality of interfaces. The composition and thickness of Mo/Si interfaces have been extensively studied [5–7]. Several approaches were proposed to improve reflectivity taking into consideration these interfacial properties. One approach consists in modeling the silicide at the interfaces [8]. Another approach consists in sputtering a thin layer of another material to prevent interdiffusion [9]. In previous papers, we have shown that boron carbide (B_4C) can improve the interfacial properties [5] and can also play the role of a third material [10], which should improve reflectivity compared with two-material multilayers.

Besides EUV lithography, EUV optics became a key element in the field of ultra-short light pulses. Indeed, high spectral bandwidth is required to achieve attosecond pulses, which can only be done with EUV photons. These high-energy photons are generated by a pulsed fs laser in a rare gas [11, 12] and need special optics to be manipulated. Broadband mirrors, made of aperiodic Mo/Si stacks, have already been deposited and characterized in the energy range 80–100 eV [8]. Experimental reflectivity reported can reach 15% for a nearly 15 eV full width at half maximum (FWHM) [13]. These kinds of mirrors are of main interest for several applications, such as generation of attosecond pulses from high-harmonic generation in a rare gas [14]. Moreover, development of broadband mirrors for lower energies (typically 35–75 eV) is also useful for astrophysics spectroscopy, where this wavelength region is rich in emission lines [15–17].

In this paper, we report on the possibility to design and deposit efficient broadband mirrors in the spectral range 35–75 eV by using three-material aperiodic multilayers.

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In the first part, we discuss optimization of broadband mirrors in the EUV range starting at 35 eV up to 75 eV using the three-material system Mo/Si/B_4C. In the second part, we describe the realization process, using a RF and DC magnetron sputtering system. In the last part, we present the reflectivity measured at the BEAR beamline at the Elettra facility in Italy [18], and compare with the simulation results.

2 Broadband mirror design

We optimize multilayer mirrors for maximum reflectivity using the three materials Mo/Si/B_4C, growth conditions of which are well controlled and routinely used for EUV optics [19] in the laboratory. We use the commercial thin-film design program TFCalc (Software Spectra Inc.) as it is versatile and makes it possible to enter optical constants of the materials, enter new materials, choose the optimization procedure between gradient or simplex methods and allow a wide variety of possible targets. The optimization procedure includes a 'needle' procedure. The needle procedure adds randomly one or more new layers in the stack of any specified material after a local optimization and starts again a new optimization until it reaches the criteria. This method ensures exploring a wider region in the parameter space, but a seed remains needed for faster and more reliable convergence. In all the cases, the calculation procedure remains the minimization of a merit function, as in [8, 20–22]. At the end of the optimization process, layers with zero thicknesses are automatically removed but still some layers can have non-realistic thicknesses, and some constraints can also be given to the program.

We optimize two mirrors at 10° incidence in s polarization and in the 35–75 eV range with different types of reflectivity targets. We use a gradient method with the needle optimization, using the optical parameters given in [23]. A first optimization process is done without any constraints of any kind; we run a last optimization procedure using a gradient method with constraints on the minimum deposited thicknesses, in particular for the B_4C barrier layers, to have realistic values. The first mirror (mirror A) maximizes the reflectivity between 40 and 75 eV with a flat profile and the seed was a periodic multilayer with maximum reflectivity at 50 eV. The second mirror (mirror B) is a multichannel mirror for high-harmonic filtering; the seed was made of thick Si spacers inside a periodic multilayer structure. The multichannel mirror can be seen as a generalization of the dual-channel mirror that we have developed for astrophysics [24]. It maximizes the reflectivity only for the odd high-harmonic wavelengths, starting from the 23rd to the 43rd generated by a 800-nm fs laser in an atomic vapor, which corresponds to the 35–66 eV energy range. Using this mirror at a different incidence angle makes it possible to select the even harmonics without changing the contrast.

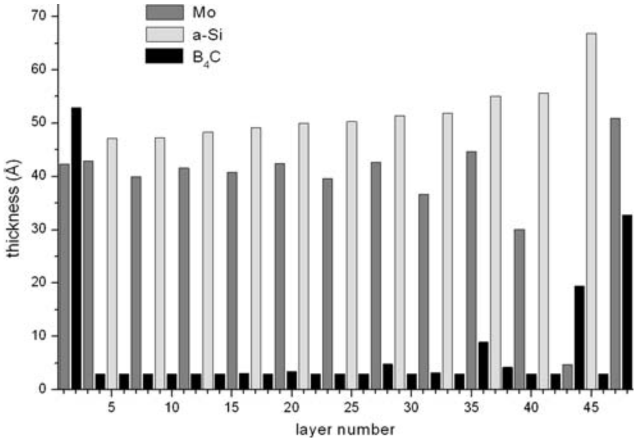

Fig. 1 Sequence of layers starting from the substrate of the mirror A with a flat reflectivity between 35 eV and 75 eV

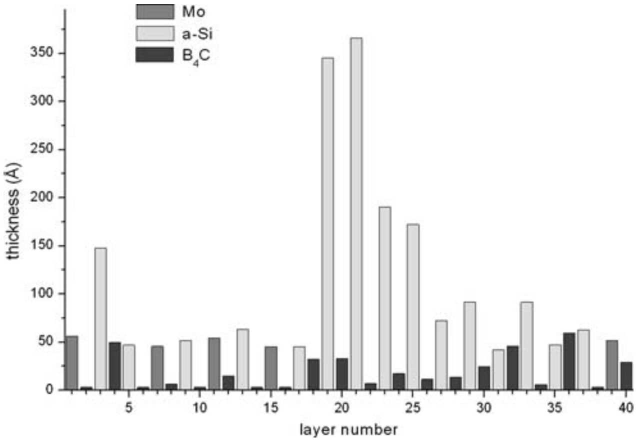

Fig. 2 Sequence of layers starting from the substrate of the mirror B reflecting the odd high harmonics from H_{23} to H_{43}

The designed multilayers are presented in Figs. 1 and 2. The first sequence shows the result for a flat reflectivity profile (mirror A), where it appears that Mo and Si have comparable thicknesses and B_4C is used mainly as an interfacial barrier layer. We can notice a slight tendency of the Si thicknesses to decrease from the surface. The second sequence shows the layer thicknesses for a multipeak reflectivity profile (mirror B). It appears that thick Si layers are needed near the center of the structure. They act as spacers between the front and back aperiodic three-material structures.

3 Sample preparation

The gap that may appear between results of optimization calculations and what can be grown is addressed by some constraints on the thicknesses of layers in the optimization process. First of all, because of the materials chosen, interdiffusion of Si and Mo at the interfaces may be a problem. In order to prevent period contraction and limit intermixing, we use a thin B_4C layer of 3 Å as a barrier layer at every Mo/Si interface that could appear in the first optimization procedure. But, B_4C is not only used as a barrier layer but also as a material participating in the reflectivity properties of the stack, which appears in the sequences in Figs. 1 and 2 with layer thicknesses larger than 3 Å. The samples are grown on a 600-µm-thick Si substrate and end with a thick enough B_4C layer to prevent the underlying layers from oxidation.

The multilayers were deposited in a DC and RF magnetron sputtering system. The chamber of preparation has a nominal vacuum of 2×10^{-8} Torr. The sputtering was performed with Ar gas at a pressure of 2 mTorr. The RF target powers were 150 W for both Si and B_4C and a direct current of 0.06 A for Mo. The sample is rotated while passing over the targets and the number and speed of scans determine the thickness of material sputtered. Due to the aperiodic concept, every layer thickness needs to be specified individually in terms of scans and rotational speeds over the target. The speeds of growth were characterized with periodic structures and found to be around 5 Å/scan, 13 Å/scan and 23 Å/scan for B_4C, Mo and Si respectively at 1°/s rotational speed. We performed reflectivity measurements in X-ray grazing incidence at the Cu K_α line to control the calibration of our material deposition.

4 Results and discussion

The mirror A was characterized in grazing incidence and the results are shown in Fig. 3. A small correction of +1.3% in Mo layer thicknesses is needed to match with a very good agreement the simulation to the measurements. This small discrepancy was used to recalibrate with more accuracy the speed of growth of Mo. This new calibration was used for mirror B.

The mirror B was also characterized in grazing incidence; the results are shown in Fig. 4. The results are in excellent agreement with the simulation, meaning that the thicknesses are what we expected.

Reflectivity measurements were performed at the BEAR beamline at the Elettra facility. Figure 5 shows the results for the flat reflectivity profile between 40 and 75 eV. We measured a maximum reflectivity of around 0.12 (black squares), that is to say, 0.04 less than expected first (dotted line); it is due to interface residual roughness and/or intermixing. The full width at half maximum is 38 eV, which is amongst the larger reported in this energy range. We fitted the measurements with new optical constants available for B_4C [25] and Mo [26] and the already well-known ones for

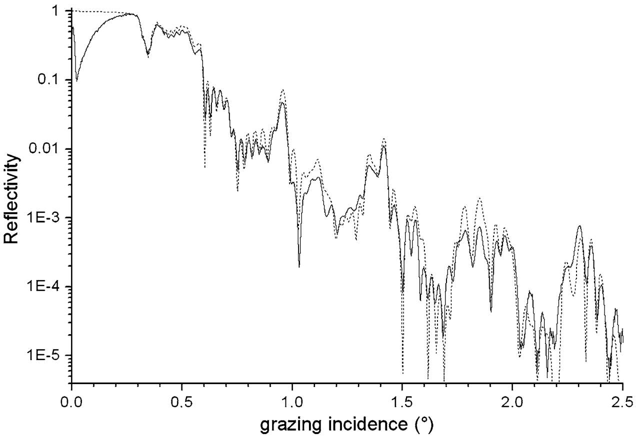

Fig. 3 Reflectance at Cu K_α line of the flat reflectivity profile sample. The *solid line* represents the measured data and the *dotted line* is the calculation with a 1.3% thickness correction on the Mo layers

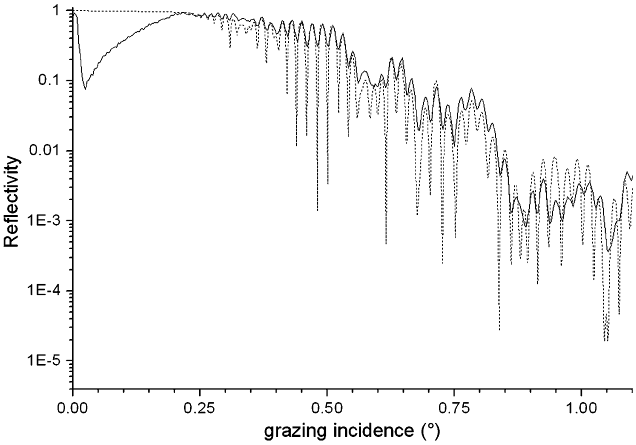

Fig. 4 Reflectance at the Cu K_α line of the multipeak profile sample. The *solid line* represents the measured data and the *dotted line* is the calculation

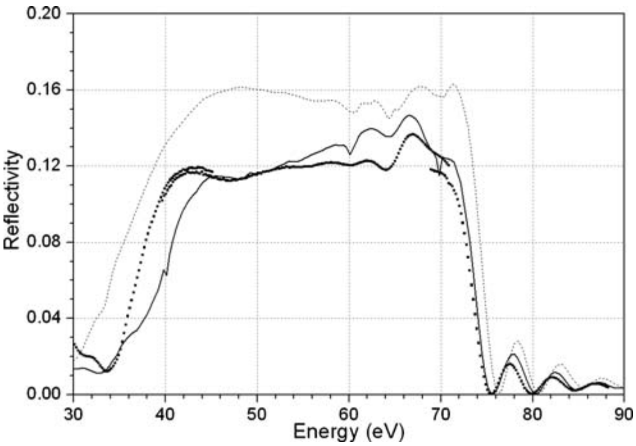

Fig. 5 Mirror A: reflectivity. *Dotted line*: results of optimization process. *Black squares*: measurement data. *Solid line*: simulation results with Mo corrected thicknesses and optical constants from [26] and B_4C new optical constants from [25]

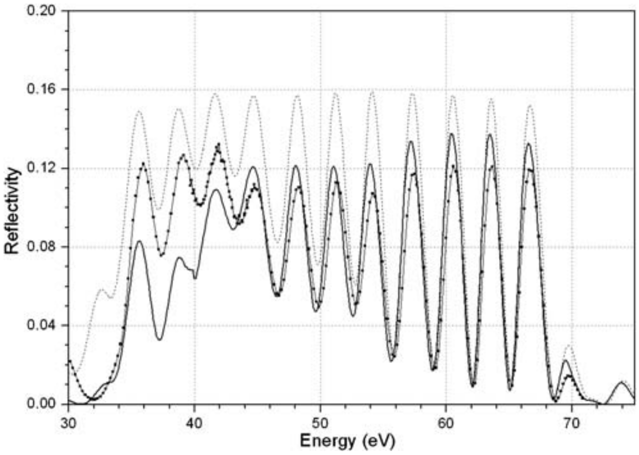

Fig. 6 Mirror B: reflectivity. *Dotted line*: results of optimization process. *Black squares*: measurement data. *Solid line*: simulation results with Mo optical constants from [26] and B_4C optical constants from [25]

Si [23]. We also tuned the thicknesses of Mo and Si so as to match the minima of reflectivity at 77 and 82 eV with measurements (solid line), where Mo optical constants are the most reliable. The shape of the reflectivity simulation reproduces quite well the measurements, but for energies lower than 50 eV discrepancies becomes significant, due to bad knowledge of optical constants of deposited materials, especially for Mo, which is difficult to measure in this energy region.

Figure 6 shows the results for the multichannel profile. We measured a maximum reflectivity of around 0.12 (black squares), that is to say, 0.04 less than expected first (dotted line), mainly due to interface residual roughness once again. We fitted the measurements with new optical constants available for B_4C [25] and Mo [26] and there was no need for thickness tuning (solid line). The positions of minima and maxima matched very well. If the results are in excellent agreement with measurements, this is nevertheless not the case for energies lower than 45 eV, where important discrepancies appear between simulations and measurements. The results show efficient filtering for harmonics above 50 eV with a maximum contrast of 85%.

The important discrepancies between simulations and measurements appearing below 50 eV come from the optical constants of Mo. In particular, the data for Mo in the literature [26] does not fit well with that of our sputtered Mo.

5 Conclusion

We have presented new developments in wide broadband mirrors for the EUV energy range that could be interesting for attosecond light pulses and high-harmonic management. These mirrors were conceived and realized in our laboratory and measured at the BEAR beamline at Elettra, Italy. Reflectivity measurements showed very good agreement with our simulations, with some discrepancies at lowest energies coming from a bad knowledge of our Mo optical constants.

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