Enhancement of soft X-ray reflectivity and interface stability in nitridated Pd/Y multilayer mirrors

Dechao Xu, Qiushi Huang, Yiwen Wang, Pin Li, Mingwu Wen, Philippe Jonnard, Angelo Giglia, Igor V. Kozhevnikov, Kun Wang, Zhong Zhang, et al.

To cite this version:

Dechao Xu, Qiushi Huang, Yiwen Wang, Pin Li, Mingwu Wen, et al.. Enhancement of soft X-ray reflectivity and interface stability in nitridated Pd/Y multilayer mirrors. Optics Express, 2015, 23 (26), pp.33018-33026. 10.1364/OE.23.033018 . hal-01270612

HAL Id: hal-01270612
https://hal.sorbonne-universite.fr/hal-01270612

Submitted on 8 Feb 2016

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

Distributed under a Creative Commons Attribution 4.0 International License

Enhancement of soft X-ray reflectivity and interface stability in nitridated Pd/Y multilayer mirrors

Dechao Xu,[1] Qiushi Huang,[1,7] Yiwen Wang,[1] Pin Li,[1] Mingwu Wen,[1] Philippe Jonnard,[2,3] Angelo Giglia,[4] Igor V. Kozhevnikov,[5] Kun Wang,[6] Zhong Zhang,[1] and Zhanshan Wang[1,8]

[1]*Key Laboratory of Advanced Micro-Structured Materials MOE, Institute of Precision Optical Engineering, School of Physics Science and Engineering, Tongji University, Shanghai 200092, China*
[2]*Sorbonne Universités, UPMC Univ Paris 06, Laboratoire de Chimie Physique - Matière et Rayonnement, 11 rue Pierre et Marie Curie, F-75231 Paris cedex 05, France*
[3]*CNRS UMR 7614, Laboratoire de Chimie Physique - Matière et Rayonnement, 11 rue Pierre et Marie Curie, F-75231 Paris cedex 05, France*
[4]*CNR Istituto Officina Materiali 34149 Trieste, Italy*
[5]*Shubnikov Institute of Crystallography, Russian Academy of Sciences, Leninskiy pr. 59, Moscow 119333, Russia*
[6]*College of Mechanical Engineering, Tongji University, Shanghai 200092, China*
[7]*huangqs@tongji.edu.cn*
[8]*wangzs@tongji.edu.cn*

Abstract Pd/Y multilayer mirrors operating in the soft X-ray region are characterized by a high theoretical reflectance, reaching 65% at normal incidence in the 8-12 nm wavelength range. However, a severe intermixing of neighboring Pd and Y layers results in an almost total disappearance of the interfaces inside the multilayer structures fabricated by direct current magnetron sputtering and thus a dramatic reflectivity decrease. Based on grazing incidence X-ray reflectometry and X-ray photoelectron spectroscopy, we demonstrate that the stability of the interfaces in Pd/Y multilayer structures can be essentially improved by adding a small amount of nitrogen (4-8%) to the working gas (Ar). High resolution transmission electron microscopy shows that the interlayer width is only 0.9 nm and 0.6 nm for Y(N)-on-Pd(N) and Pd(N)-on-Y(N) interfaces, respectively. A well-defined crystalline texture of YN (200) is observed on the electron diffraction pattern. As a result, the measured reflectance of the Pd(N)/Y(N) multilayer achieves 30% at λ = 9.3 nm. The peak reflectivity value is limited by the remaining interlayers and the formation of the YN compound inside the yttrium layers, resulting in an increased absorption.

©2015 Optical Society of America

OCIS codes: (340.7470) X-ray mirrors; (340.7480) X-rays, soft x-rays, extreme ultraviolet (EUV); (310.1860) Deposition and fabrication.

References and links

1. E. Louis, A. E. Yakshin, T. Tsarfati, and F. Bijkerk, “Nanometer interface and materials control for multilayer EUV-optical applications,” Prog. Surf. Sci. **86**(11-12), 255–294 (2011).
2. D. S. Kuznetsov, A. E. Yakshin, J. M. Sturm, R. W. E. van de Kruijs, E. Louis, and F. Bijkerk, “High-reflectance La/B-based multilayer mirror for 6.x nm wavelength,” Opt. Lett. **40**(16), 3778–3781 (2015).
3. J.-M. André, P. Jonnard, C. Michaelsen, J. Wiesmann, M.-F. Ravet, F. Delmotte, F. Bridou, and E. Filatova, “La/B_4C small period multilayer interferential mirrors for the analysis of boron,” X-Ray Spectrom. **34**(3), 203–206 (2005).
4. P. Boerner, C. Edwards, J. Lemen, A. Rausch, C. Schrijver, R. Shine, L. Shing, R. Stern, T. Tarbell, A. Title, C. Jacob Wolfson, R. Soufli, E. Spiller, E. Gullikson, D. McKenzie, D. Windt, L. Golub, W. Podgorski, P. Testa, and M. Weber, “Initial Calibration of the Atmospheric Imaging Assembly (AIA) on the Solar Dynamics Observatory (SDO),” Sol. Phys. **275**(1-2), 41–66 (2012).

#251252 Received 1 Oct 2015; revised 3 Dec 2015; accepted 3 Dec 2015; published 14 Dec 2015

5. M. H. Hu, K. Le Guen, J. M. André, P. Jonnard, E. Meltchakov, F. Delmotte, and A. Galtayries, “Structural properties of Al/Mo/SiC multilayers with high reflectivity for extreme ultraviolet light,” Opt. Express **18**(19), 20019–20028 (2010).
6. Q. Zhong, Z. Zhang, R. Qi, J. Li, Z. Wang, K. Le Guen, J. M. André, and P. Jonnard, “Enhancement of the reflectivity of Al/Zr multilayers by a novel structure,” Opt. Express **21**(12), 14399–14408 (2013).
7. F. Eriksson, N. Ghafoor, L. Hultman, and J. Birch, “Reflectivity and structural evolution of Cr/Sc and nitrogen containing Cr/Sc multilayers during thermal annealing,” J. Appl. Phys. **104**(6), 063516 (2008).
8. M. Prasciolu, A. F. G. Leontowich, K. R. Beyerlein, and S. Bajt, “Thermal stability studies of short period Sc/Cr and Sc/B_4C/Cr multilayers,” Appl. Opt. **53**(10), 2126–2135 (2014).
9. E. M. Gullikson, F. Salmassi, A. L. Aquila, and F. Dollar, “Progress in short period multilayer coatings for water window applications,” The 8th International Conference on The Physics of X-Ray Multilayers Structures, (2006).
10. A. J. Corso, P. Zuppella, D. L. Windt, M. Zangrando, and M. G. Pelizzo, “Extreme ultraviolet multilayer for the FERMI@Elettra free electron laser beam transport system,” Opt. Express **20**(7), 8006–8014 (2012).
11. B. Kjornrattanawanich and S. Bajt, “Structural characterization and lifetime stability of Mo/Y extreme-ultraviolet multilayer mirrors,” Appl. Opt. **43**(32), 5955–5962 (2004).
12. V. V. Medvedev, A. E. Yakshin, R. W. E. van de Kruijs, and F. Bijkerk, “Phosphorus-based compounds for EUV multilayer optics materials,” Opt. Express **5**(6), 1450–1459 (2015).
13. B. Sae-Lao and C. Montcalm, “Molybdenum-strontium multilayer mirrors for the 8-12-nm extreme-ultraviolet wavelength region,” Opt. Lett. **26**(7), 468–470 (2001).
14. C. Montcalm, P. A. Kearney, J. M. Slaughter, B. T. Sullivan, M. Chaker, H. Pépin, and C. M. Falco, “Survey of Ti-, B-, and Y-based soft x-ray-extreme ultraviolet multilayer mirrors for the 2- to 12-nm wavelength region,” Appl. Opt. **35**(25), 5134–5147 (1996).
15. D. L. Windt and E. M. Gullikson, “Pd/B_4C/Y multilayer coatings for extreme ultraviolet applications near 10 nm wavelength,” Appl. Opt. **54**(18), 5850–5860 (2015).
16. D. L. Windt, “Reduction of stress and roughness by reactive sputtering in W/B_4C X-ray multilayer films,” Proc. SPIE **6688**, 66880R (2007).
17. A. V. Vinogradov and B. Ya. Zeldovich, “X-ray and far uv multilayer mirrors: principles and possibilities,” Appl. Opt. **16**(1), 89–93 (1977).
18. R. A. Alqasmi, S. Paasch, and H.-J. Schaller, “Thermodynamic properties of Pd–Y and Pd–Gd intermetallic phases,” J. Alloys Compd. **283**(1-2), 173–177 (1999).
19. U. Diebold, “The surface science of titanium dioxide,” Surf. Sci. Rep. **48**(5-8), 53–229 (2003).
20. M. Li, W. Hebenstreit, and U. Diebold, “Morphology change of oxygen-restructured TiO2 (110) surfaces by UHV annealing: Formation of a low-temperature (1×2) structure,” Phys. Rev. B **61**(7), 4926–4933 (2000).
21. S. Bajt, D. G. Stearns, and P. A. Kearney, “Investigation of the amorphous-to-crystalline transition in Mo/Si multilayers,” J. Appl. Phys. **90**(2), 1017–1025 (2001).
22. N. Ghafoor, F. Ericsson, E. Gullikson, L. Hultman, and J. Birch, “Incorporation of nitrogen in Cr/Sc multilayers giving improved soft x-ray reflectivity,” Appl. Phys. Lett. **92**(9), 091913 (2008).
23. S. Nannarone, F. Borgatti, A. DeLuisa, B. P. Doyle, G. C. Gazzadi, A. Giglia, P. Finetti, N. Mahne, L. Pasquali, M. Pedio, G. Selvaggi, G. Naletto, M. G. Pelizzo, and G. Tondello, “The BEAR Beamline at Elettra,” AIP Conf. Proc. **705**, 450–453 (2004).
24. B. L. Henke, E. M. Gullikson, and J. C. Davis, “X-ray interactions: photoabsorption, scattering, transmission, and reflection at E=50-30000 eV, Z=1-92,” At. Data Nucl. Data Tables **54**(2), 181–342 (1993).
25. P. Kempter, N. H. Krikorian, and J. C. McGuire, “The crystal structure of yttrium nitride,” J. Phys. Chem. **61**(9), 1237–1238 (1957).

1. Introduction

In spite of impressive progress in the fabrication technology of multilayer mirrors for soft X-ray (SXR) and extreme ultraviolet (EUV) radiation, these high-quality mirrors were developed for several spectral regions only. Among them are the Mo/Si multilayers for EUV lithography (λ = 13.5 nm; experimentally achieved reflectivity at normal incidence R = 70.3%) [1], La-containing multilayers for “beyond EUV” lithography (λ ~6.7 nm, R = 64%) [2, 3], Al-containing multilayers for solar astronomy (λ ~17-30 nm, R = 53%) [4–6] and multilayers for SXR microscopy in the “water window” region (λ ~2.3-4.4 nm, R_{max} = 32%) [7–9]. At the same time, all of these multilayers are poorly suited for the 8-12 nm spectral region, which is of particular interest for solar astronomy [4] and a free-electron laser beamline [10]. Figure 1 shows that the theoretical peak reflectivity of the well-studied Mo- and La-containing multilayers does not exceed 40%, as a rule. Some increase in the reflectivity can be obtained by the use of a lower absorbing yttrium spacer. The experimentally achieved reflectivity of the Mo/Y multilayer is equal to 38.4% at λ = 9.5 nm [11].

#251252 Received 1 Oct 2015; revised 3 Dec 2015; accepted 3 Dec 2015; published 14 Dec 2015
© 2015 OSA

The most promising absorbing material in the 8-12 nm spectral region is palladium, whose absorptivity is almost the same as that of Mo, while the real part of the polarizability exceeds that of Mo by a factor of 1.5 (at λ = 9.5 nm). As a result, the calculated reflectivity of the Pd/B_4C mirror is higher than that of Mo- and La-containing multilayers in the 8-11 nm spectral interval (see Fig. 1). Replacement of the B_4C spacer with less absorbing B or Y gives an additional rise in the reflectivity of up to 65% for the Pd/Y multilayer at λ = 9.5 nm. The maximal possible reflectance is achieved for Pd/Sr or Pd/BP [12] multilayers (the last one is not shown in Fig. 1), exceeding 70%.

Unfortunately, the high reactivity of Sr results in a fast deterioration of the multilayer structure [13]. The deposition of BP compound, as well as pure B, is a critical issue for direct current (DC) magnetron sputtering, which is a standard technique for the fabrication of SXR and EUV multilayer mirrors. Therefore, we consider the Pd/Y multilayer as the best candidate for a highly reflective mirror in the 8–12 nm spectral range from both theoretical and technological points of view.

The Pd/Y multilayers were first proposed and experimentally studied by Montcalm et al in [14], where the main problem of practical fabrication of these multilayers was also indicated, namely, the severe intermixing and compound formation between Pd and Y layers destroying the optical contrast (dielectric constant variation) at the interfaces. If sharp interfaces are produced between the Pd and Y layers, high reflectance of the multilayer can be expected. Recently, an introduced thin B_4C barrier layer was used to mitigate the interdiffusion between Pd and Y, and the experimental reflectivity was increased up to 43% at λ = 9.35 nm [15]. Chemical passivation of the layers, e.g. by nitridation, is another way to reduce the interdiffusion and compound formation at the interfaces. This approach was already successfully applied to several multilayer systems including La/B [2], W/B_4C [16], and Cr/Sc [7]. In this paper, we demonstrate that the nitridation of both Pd and Y layers can also increase the interface stability and enhance the soft X-ray reflectivity. The near normal incidence SXR reflectance from the Pd(N)/Y(N) multilayer is found experimentally to be equal to 30% at a wavelength of 9.3 nm.

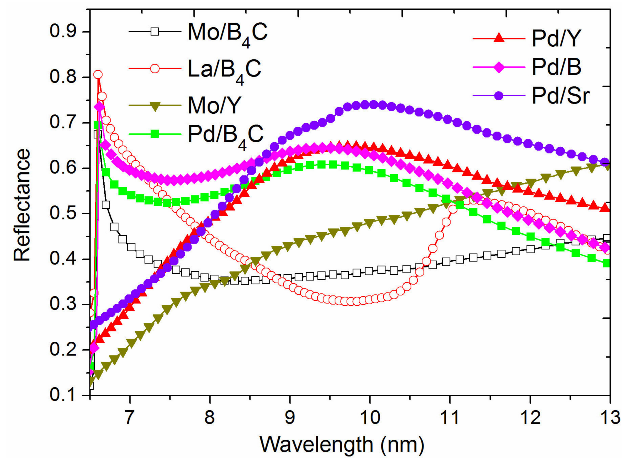

Fig. 1. Maximal normal incidence reflectance of different multilayer mirrors for operation in the 8-12 nm wavelength range. The calculations were performed for semi-infinite multilayer structures with the optimized thickness of absorbing and spacing layers [17]. Neither interlayers nor interfacial roughness were taken into account.

2. Fabrication and structural characterization of Pd/Y multilayers

The studied Pd/Y multilayer mirrors were designed for operation at the 9.5 nm wavelength, with a d-spacing of approximately 4.9 nm and the ratio of the Pd layer thickness to the d-spacing γ = ~0.5. The multilayers were fabricated by using the DC magnetron sputtering

#251252 Received 1 Oct 2015; revised 3 Dec 2015; accepted 3 Dec 2015; published 14 Dec 2015
© 2015 OSA

technique. The base pressure before deposition was ~3.0 × 10-5 Pa. Both high purity argon and a mixture of argon and nitrogen were used as the working gas. Nitrogen was used to passivate the interfaces between the Pd and Y layers and to enhance their stability due to the possible formation of yttrium nitride (YN) because the Gibbs free energy of Pd-Y compound formation is between −60 and −91.7 kJ/mol (at 1073 K) [14, 18] and the energy of YN formation is −269 kJ/mol (at 298 K) [14]. Three different mixing ratios, i.e., the ratios of the partial pressure of N2 to the total pressure in the bottle, were used: 4%, 6%, and 8% nitrogen. The total working pressure in the deposition chamber was set to 0.1 Pa. The multilayers were deposited onto superpolished silicon wafers with a root-mean-squared (rms) surface roughness of 0.2 nm.

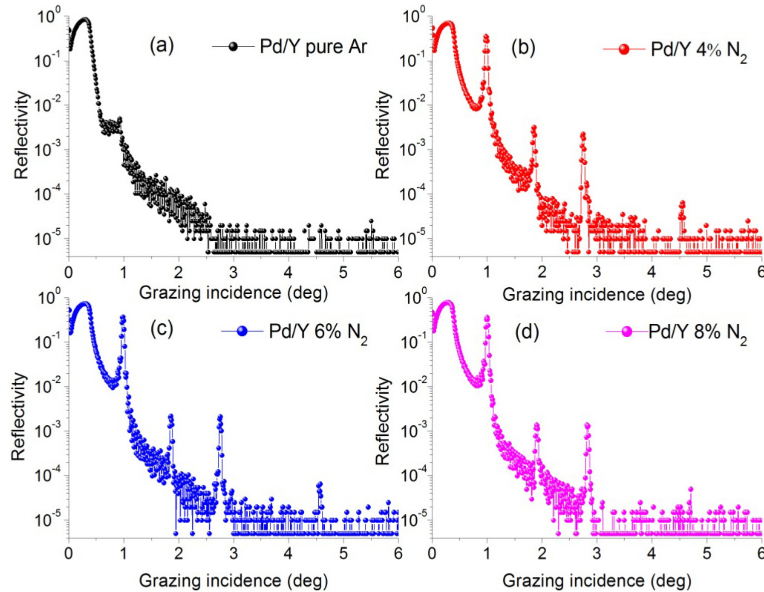

Fig. 2. X-ray reflectivity (at λ = 0.154 nm) versus the grazing angle of Pd(N)/Y(N) multilayers fabricated by magnetron sputtering with the use of pure Ar as the working gas (a) and a mixture of Ar + N2, where the content of nitrogen is 4% (b), 6% (c) and 8% (d).

At first, a set of Pd/Y and Pd(N)/Y(N) multilayers consisting of 20 bilayers was fabricated to study the internal structure of the mirrors by grazing incidence X-ray reflectometry (GIXR) at the Cu-Kα emission line (λ = 0.154 nm) and by X-ray photoelectron spectroscopy (XPS). The GIXR curves of four Pd/Y multilayers deposited in pure Ar, Ar + 4% N_2, Ar + 6% N_2 and Ar + 8% N_2 are shown in Fig. 2. Only the very low 1st Bragg peak is observed for the multilayer deposited in pure argon (Fig. 2(a)). The same behavior of reflectivity was also detected in Ref [14, 15]. This fact indicates that, probably, the Pd and Y layers are almost totally intermixed such that interfaces between neighboring layers are barely perceptible. However, after adding 4% nitrogen to the working gas, the layered structure is drastically improved and the 5th Bragg peak is quite observable in the GIXR curve (Fig. 2(b)) (the 4th Bragg peak is suppressed here). We can conclude that adding a small amount of nitrogen to the working gas reduced significantly the intermixing of Pd and Y layers. A further increase in the nitrogen content up to 6% and 8% only slightly affects the GIXR curves. In particular, the 1st order Bragg peak reflectance is equal to (36 ± 1)% for all three curves in Fig. 2(b), 2(c), 2(d), demonstrating that the interface width in the Pd(N)/Y(N) multilayers remains almost the same as the nitrogen content increases from 4% to 8%.

To study the effect of nitridation on the interface stability and the change in chemical composition of the layers, Pd/Y multilayers fabricated with both pure Ar and Ar + 4% N_2

#251252 Received 1 Oct 2015; revised 3 Dec 2015; accepted 3 Dec 2015; published 14 Dec 2015
© 2015 OSA

working gas were analyzed using X-ray photoelectron spectroscopy (XPS). The AXIS Ultra DLD instrument and Al Kα radiation (E = 1486 eV) were used for the analysis. A cap layer of 2.5 nm thick Pd was deposited onto the top of both the multilayers to avoid oxidation. Ion etching (2 keV Ar^+) was applied to determine the atomic concentration of Y and Pd with depth. The ion spot size was 3 × 3 mm^2, and the etching rate of the Pd(N)/Y(N) multilayer was approximately 1 nm/min.

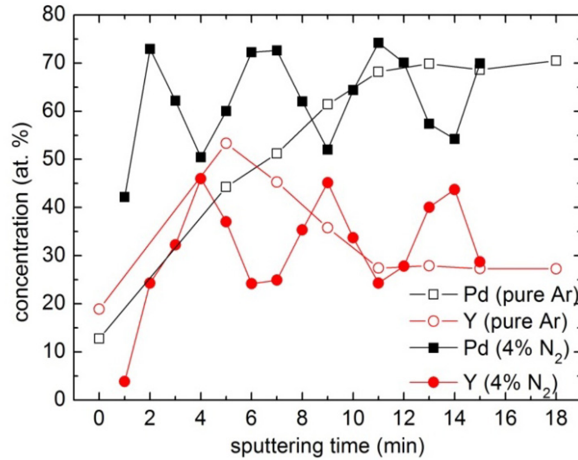

Fig. 3. XPS sputter depth profiles of the Pd/Y multilayer and the Pd(N)/Y(N) multilayer fabricated with 4% N2 content.

The measured concentration profiles of Pd and Y are displayed in Fig. 3. In the case of the Pd/Y multilayer fabricated with pure Ar, the ratio of the atomic concentrations of Pd and Y is kept constant (~2.4) starting at a depth of approximately 10 nm. This value is very close to the ratio (~2.3) of the deposited atoms of Pd to Y, assuming the bulk density of materials and the same thickness of Pd and Y layers prescribed when designing the structure with abrupt interfaces. This fact clearly demonstrates a complete intermixing of layers inside the Pd/Y structure. The concentration ratio is quite different near the surface, where the concentration of Y becomes even higher than that of Pd. Probably, this can be explained by the intermixing of Pd and Y, oxidation of Y on the surface, and diffusion of Y atoms from the interior of the structure to the surface, driven by the oxidation process. The last effect was earlier observed for other metal systems [19, 20]. The low concentrations of Pd and Y on the surface are explained by the high oxygen and carbon content caused by the oxidation of Y and the formation of an adhesion layer consisting mainly of water and hydrocarbon molecules.

Adding 4% nitrogen to the working gas resulted in the formation of a clearly observed periodic Pd(N)/Y(N) structure and hence an essential mitigation of interdiffusion between the Pd and Y layers. In particular, the concentration of yttrium reduced to zero on the surface. This fact explains the appearance of well-pronounced Bragg peaks in the reflectivity curve shown in Fig. 2(b).

The internal structure and morphology of the multilayer deposited with 4% nitrogen in the working gas were characterized by transmission electron microscopy (TEM) using the FEI Tecnai G2 F20 equipment of Materials Analysis Technology Inc.. Both bright field and dark field images were obtained using a transmitted and diffracted beam, respectively. The bright field image (Fig. 4(a)) shows the layered structure, while the dark field image (Fig. 4(b)) reveals the crystalline grains. A periodic layered structure is clearly observed in Fig. 4(a), where the light layers are yttrium and the dark ones are palladium. The periodic structure is consistent with the XPS results. The thickness of both Y and Pd layers was found to be approximately 2.4 nm. Atomic planes with different orientations are seen inside the yttrium layers, giving an indication of crystalline grains. Despite the regular layered structure, the

#251252 Received 1 Oct 2015; revised 3 Dec 2015; accepted 3 Dec 2015; published 14 Dec 2015
© 2015 OSA

intermixing areas at both the Pd-on-Y and Y-on-Pd interfaces are still observed. The interlayer width was estimated to be 0.9 nm and 0.6 nm at the Y-on-Pd and Pd-on-Y interface, respectively.

The selected area electron diffraction (SAED) pattern is shown in Fig. 4(c). The two bright spots and two discontinuous diffraction rings indicate the existence of polycrystalline grains in the multilayer structure. The two bright spots in the outer ring were identified as YN (200) diffraction. These two spots appear in the same direction as the diffraction pattern from the multilayer (near the central spot). Thus, we can conclude that after adding nitrogen to the working gas, a part of the yttrium reacted with nitrogen and formed YN polycrystalline grains, a large part of them being oriented parallel to the multilayer interfaces. The dark field image (Fig. 4(b)) demonstrates that crystalline grains (YN) appear mainly inside the Y layers. The grain size along the multilayer growth direction is limited by the layer thickness, while the lateral size is a few nanometers. The second ring in Fig. 4(c) is most likely to be identified as Y(002) diffraction. The diffraction pattern of palladium was not observed, which indicates that the palladium layer is in an amorphous state. This fact explains the asymmetrical interfaces observed in Fig. 4(a), as the crystallites in the Y layers suppress the interlayer formation during the growth of Pd-on-Y [21].

In addition, energy dispersive X-ray spectroscopy (EDX) was used to analyze the chemical composition inside layers of the studied samples. The atomic concentration of nitrogen contained in the palladium layers was estimated to be less than 10%, while the nitrogen content in the yttrium layers is noticeably higher and reaches 16~19%. This is consistent with the SAED results, which show that YN is formed inside the yttrium layers.

The experimental results thus demonstrate clearly that the nitridation of Pd/Y multilayer structure suppresses heavily the interdiffusion of Pd and Y layers. The TEM and SAED data indicate that an essential part of yttrium layers consists of YN and pure Y crystalline grains. As a diffusion of an amorphous material (Pd) occurs mainly along the grain boundaries of a polycrystalline material (Y, YN), while it occurs over the whole interface area in the case of two neighboring amorphous materials, we conclude that crystallization of yttrium layers is one of the possible factors leading to stabilization of interfaces. Nevertheless, we cannot exclude that a part of yttrium layers is still in amorphous state. Following [22] we can assume that interdiffusion of Pd and amorphous Y is also suppressed due to incorporation of considerable amount of nitrogen into both materials and formation of strong chemical metal-nitrogen bonds decreasing mobility of atoms.

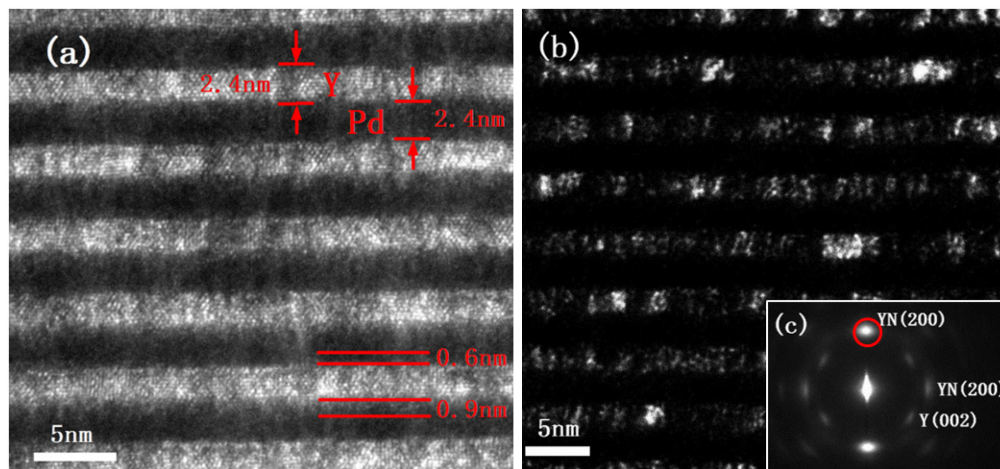

Fig. 4. TEM bright field image (a), dark field image (b) and electron diffraction pattern (c) of the Pd(N)/Y(N) multilayer fabricated with 4% N_2 content in the working gas. The multilayer surface is on the top of images (a) and (b).

4. Soft X-ray reflectivity measurement

For the SXR reflectivity measurements, three Pd(N)/Y(N) multilayers consisting of 80 bilayers were fabricated with different contents of nitrogen in the working gas (4%, 6%, and 8%). The number of bilayers was large enough to provide reflectivity approaching the maximal possible value at $\lambda = \sim 9.5$nm. The multilayer fabricated without adding nitrogen to the working gas was not measured because preliminary experiments indicated the disappearance of the layered structure due to the intermixing of materials. The measurements were performed at the Bending magnet for Emission Absorption and Reflectivity (BEAR) beamline at the ELETTRA synchrotron [23]. The spectral dependence of the reflectivity at the 5 degree incidence angle (off normal) is shown in Fig. 5. The multilayer fabricated with the addition of 4% nitrogen to the working gas demonstrates 29.3% reflectivity at $\lambda = 9.3$ nm. A further increase of the nitrogen content leaves the reflectivity curve almost unchanged: the peak reflectivity of the mirrors fabricated with 6% and 8% nitrogen content in the working gas is equal to 30.0% and 28.7%, respectively. A small shift of the reflectivity peak in the 6% case is caused by a slightly different period of the sample.

Despite the significantly improved internal structure of the Pd(N)/Y(N) multilayers, the measured SXR reflectance is only half of the theoretical value of 62% at $\lambda = 9.3$ nm. The discrepancy can be caused by the following reasons. First, according to the TEM results, the interlayers of considerable width (0.6 nm and 0.9 nm) are still left at the Pd(N)-on-Y(N) and Y(N)-on-Pd(N) interfaces after nitridation of the multilayer structure. The interlayers reduce the reflectivity by almost 20% (in absolute units) compared with the ideal structure with abrupt interfaces. As seen in Fig. 6, the calculated peak reflectivity of the Pd/Y multilayer with interlayers being taken into account is only 43% instead of 62% for the ideal mirror with abrupt interfaces.

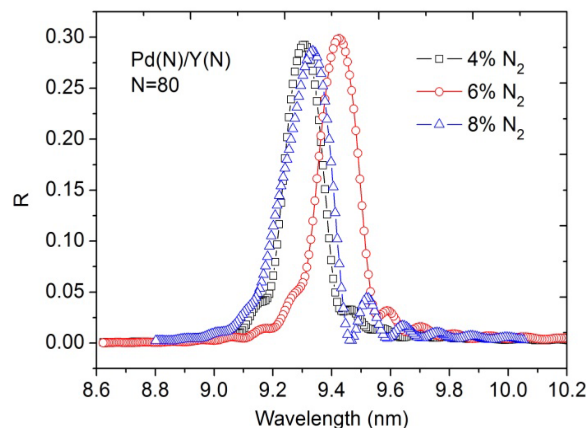

Fig. 5. The measured reflectivity versus the radiation wavelength of the Pd(N)/Y(N) multilayers fabricated by magnetron sputtering with different nitrogen contents (4%, 6%, and 8%) in the working gas (Ar).

Second, adding nitrogen to the working gas results in nitridation of both materials of the multilayer structure and the formation of crystalline YN inside the yttrium layers, which enhances the interface stability. However, impurities essentially increase the SXR radiation absorption in the spacing (Y) layers. Assuming the Y layer is changed into YN with a density of 4.4 g/cm^3 (the same as the density of pure Y), the absorptivity of the spacing layers is almost doubled compared with the pure Y spacer [24]. Then, the peak reflectivity decreases to 33%, as illustrated by Fig. 6. Evidently, increasing the YN layer density results in a further decrease in the reflectivity to 25.5% at a 5.6 g/cm^3 YN density (the value corresponding to the cubic crystalline structure of YN [25]). Although the comparison is based on the assumption that all Y layers are transformed into YN ones, which is not the real case, Fig. 6 clearly

#251252 Received 1 Oct 2015; revised 3 Dec 2015; accepted 3 Dec 2015; published 14 Dec 2015
© 2015 OSA

indicates that the nitridation of Y layers causes reflectance loss compared to the pure Y layers case. Note that the effect of impurities introduced into the absorbing layers (Pd) is much weaker. Therefore, the effects of interlayers in the Pd(N)/Y(N) structure and the incorporation of nitrogen into the Y layers may explain the lower value of the measured reflectance compared with that of the ideal Pd/Y mirror.

These effects may also explain an independence of the measured SXR reflectivity on the nitrogen concentration in the working gas (see Fig. 5). On the one hand, an increasing nitrogen concentration results in a nitridation enhancement and thus in an increasing stability of interfaces and a reflectivity rise. On the other hand, increasing content of YN in yttrium layers leads to increasing absorption of an incident SXR radiation, so that these two opposite processes are balancing each other retaining the reflectivity almost constant.

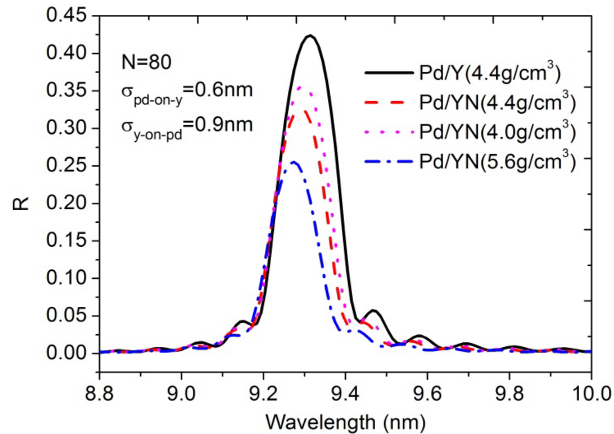

Fig. 6. The calculated reflectivity versus the radiation wavelength of the Pd/Y and Pd/YN multilayers, with the different YN densities indicated in the graph. Interlayers of 0.6 nm and 0.9 nm thickness were taken into account at the Pd-on-Y and Y-on-Pd interfaces, respectively. The optical constants of materials were taken from the CXRO website [23].

5. Summary

Soft X-ray Pd/Y multilayer mirrors designed for operation in the 8-12 nm wavelength region were fabricated by the direct current magnetron sputtering technique with the use of pure Ar and a mixture of Ar and nitrogen (4% to 8% N_2) as the working gas. The periodic structure of the multilayers fabricated with the pure Ar working gas was demonstrated, using GIXR and XPS, to have disappeared due to the severe intermixing of the Pd and Y layers. At the same time, adding a small amount of nitrogen to argon increased the stability of the interfaces and essentially mitigated the interdiffusion of Pd and Y mainly due to the formation of YN crystallites in the yttrium layers. The strong texture of YN was clearly observed in the SAED pattern, and the well-pronounced periodic structure of the Pd(N)/Y(N) multilayer was shown through XPS analysis. However, interlayers of considerable width (0.6 nm and 0.9 nm) still remained at the Pd(N)-on-Y(N) and Y(N)-on-Pd(N) interfaces and were observed in the TEM images. The interlayers, on the one hand, and the increased absorption of the spacing layers due to the introduction of nitrogen, on the other hand, reduced the SXR reflectivity by half at λ = 9.3 nm, from 62% for the ideal Pd/Y structure with abrupt interfaces down to 30% (measured experimentally). One of possible ways for further reflectivity increase is to use a time-dosed nitridation of yttrium layers at initial and final stages of their growth only. We would expect that such a process allows to obtain relatively sharp Pd-Y interfaces and a smaller amount of nitrogen inside Y layers. Notice that the similar process was used successfully in [2] under development of high reflective B/LaN multilayer mirrors.

#251252 Received 1 Oct 2015; revised 3 Dec 2015; accepted 3 Dec 2015; published 14 Dec 2015
© 2015 OSA

Acknowledgments

This work was supported by the National Natural Science Foundation of China (No. 11443007, No. 11505129), NSAF (No. U1430131), the National Key Scientific Instrument and Equipment Development Project (No. 2012YQ13012505), Shanghai Pujiang Program (No. 15PJ1408000), and the Fundamental Research Funds for the Central Universities. The SXR reflectivity measurements were carried out at the ELETTRA synchrotron in the framework of Proposal No. 20145172.

#251252 Received 1 Oct 2015; revised 3 Dec 2015; accepted 3 Dec 2015; published 14 Dec 2015
© 2015 OSA