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Microstructure evolution and hard x-ray reflectance of ultrathin Ru/C multilayer mirrors with different layer thicknesses

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Abstract

Nanoscale Ru/C multilayers are essential reflective optics in the hard x-ray region of 7–20 keV. To understand the layer growth behavior and develop ultrathin Ru/C multilayer mirrors with periods smaller than 3.0 nm, multilayers with different periods of 6.2–1.5 nm were fabricated and studied. It is found that the average interface width started to increase obviously when the period became smaller than 2.5 nm while the surface roughness of different multilayers remained almost the same. The intrinsic stress of the multilayer gradually decreased with decreasing period and reached a very low value of −82 MPa at d = 2.3 nm. High reflectance of 54% and 65% (at E = 8.04 keV) were demonstrated for the multilayers with periods of 2.5 nm and 3.0 nm, respectively, whereas that for 1.9 nm period was significantly lower. To further analyze the layer microstructure, x-ray diffraction and transmission electron microscopy were used. The polycrystallized structure of Ru remained similar for the multilayers with period less than 2.5 nm, while a non-continuous layer growth and severe intermixing between Ru and C were observed for the multilayer with period of 1.9 nm. The increased intermixing between Ru and C was found to be the main reason for the larger interface width and lower reflectance of the multilayers with period smaller than 2.5 nm. It also indicated that the layer thickness threshold for a Ru/C multilayer growing with good layer quality is 1.0–1.2 nm.

1. Introduction

Nanoscale multilayers are widely used as x-ray monochromators in synchrotron radiation facilities, plasma diagnostics and other related fields [1–4]. Compared with crystal monochromators, multilayer monochromators have a two orders of magnitude larger bandwidth which can bring much higher integral photon flux [5–7]. The period (d) of multilayer can also be easily adapted to work at different energies and incident angles.

Ru/C multilayer is one of the ideal candidates for the high-flux monochromator that operate in the energy range of 7–20 keV. Compared with other possible multilayers in this energy range, such as Ru/B4C and Pd/B4C [8, 9], the intrinsic stress of Ru/C multilayer is much lower, which assures improved stability and preservation of the precise surface figure of substrates. The layer structure and thermal stability of Ru/C multilayers with a relatively large period have been studied before. Nguyen et al fabricated Ru/C multilayers with the period of 3.5, 5.0, and 10.0 nm for normal incidence reflectors at soft x-ray wavelengths. It was found that the deposited Ru layer was polycrystallized with the crystallite size on the order of a few nanometers at room temperature [10, 11]. After annealing at 600 °C, the period of Ru/C multilayer (d = 3.5 nm) increased by 14%, the roughness became larger, and the Ru layers have clearly agglomerated [11]. Hui et al studied the microstructures of Ru/C multilayers with period of 3.0, 4.0, and 5.0 nm and their stability under high and low temperature environment. They found that the grain sizes in Ru layers increased from 1.27 nm to 1.62 nm as the period increased from 3.0 nm to 5.0 nm, and the interfacial roughness and interdiffusion were strongly correlated to the thickness and...
crystallinity of Ru. The interface width of the as-deposited multilayer is around 0.31–0.36 nm which decreased to 0.29–0.31 nm after soaking in the low temperature environment of LN₂. For the multilayer with period of 5.0 nm, the interface roughness increased after annealed at 600 °C, and an asymmetric interface structure was observed before and after annealing [12–14]. Störmer et al studied the effects of deposition process on Ru/C multilayers with period of ~2.8 nm. It was found that higher sputtering power and lower sputtering pressure can improve the interface quality, and the reflectivity of the Ru/C multilayer increased from 40% to approximately 60% at 8 keV after process optimization. A 500 mm long laterally graded Ru/C multilayer mirrors was fabricated for applications in synchrotron beamlines [15].

Notwithstanding the aforementioned studies, the change in the layer structure with a decreasing period, from 3.0 nm down to around 1.0 nm, and the smallest thickness of the multilayer achievable with good interface quality have not been studied systematically. This is of particular interest for hard x-ray multilayer monochromators as a smaller period will enable the mirror to work at larger grazing incidence angle and higher photon energy. It will also increase the saturated number of bilayers contributing to the Bragg reflection and provide a higher energy resolution [16]. Meanwhile, the growth behavior of such ultrathin multilayers can be significantly different with the thicker ones, since the layer thickness is approaching the threshold of continuous growth of thin film. In this paper, the microstructures and reflectivities of Ru/C multilayers with periods of 6.2 nm down to 1.5 nm are systematically characterized, and the physical mechanism for the evolution of the structure is discussed. The layer thickness threshold of Ru and C for growing with a good interface quality and high reflectivity is found to be 1.0–1.2 nm, using the current magnetron sputtering technique.

2. Methods

Two groups of samples were designed and fabricated in this paper. The first group consists of 6 Ru/C multilayer samples with periods of approximately 1.5 nm (S1), 1.8 nm (S2), 2.3 nm (S3), 2.5 nm (S4), 3.1 nm (S5), and 6.2 nm (S6). The samples S1–S4 consisted of 30 bilayers, whereas S5–S6 consisted of 20 bilayers. This is to study the evolution of layer microstructures with decreasing period. To make the fitting procedures easier, less number of bilayers were selected to reduce the number of free parameters. The second group consists of 3 multilayer samples with periods of approximately 1.9 nm (S2-M), 2.5 nm (S4-M), and 3.0 nm (S5-M). A saturated number of bilayers was selected based on theoretical simulations, which were 300, 150, and 100 bilayers, respectively. This is to study the maximum achievable experimental reflectivity of the ultrathin Ru/C multilayers. Thus, they need to be fabricated with large number of bilayers. The reason to choose these three periods is that they approximately represent the different growth modes based on the characterization results of the first group. The ratio of the Ru layer thickness to the period was approximately 0.5. The top layer of all samples was the C layer. All Ru/C multilayer samples were fabricated using the direct current (DC) magnetron sputtering technique (the equipment was home designed and made in China) on super-polished Si (100) wafers with a surface roughness of 0.2 nm (root-mean-square, RMS). The substrate was not heated and electrically grounded. The Ru and C layer are deposited alternatively by moving the substrate back and forth from the different sputtering area. The base pressure before deposition was 5.0 × 10⁻⁶ Pa. During the deposition process, Ar with a purity of 99.999% was used as the sputtering gas with a working pressure approximately 0.14 Pa. The deposition rates of the Ru and C layers were approximately 0.070 nm s⁻¹ and 0.040 nm s⁻¹, respectively.

After deposition, the multilayers were characterized by grazing incidence x-ray reflectometry (GIXR) at the Cu-Kα emission line (λ = 0.154 nm). The GIXR curves were fitted based on a simple two-layer model with a genetic algorithm by using IMD software (version 5.0 [17]) to determine the structural information, including the thickness, density, and interface width of Ru layer and C layer. The hard x-ray reflectance at 8.04 keV was also obtained from the GIXR measurements. The lab-based x-ray diffractometer (Bede D1, Durham, UK) was equipped with a Si (220) crystal monochromator, which provided high monochromaticity and a beam divergence of approximately 0.007°. The Nevot-Croce factor was used to simulate the effect of the interface width (including both roughness and interdiffusion) on reflectivity. The surface morphology of the multilayers was measured using atomic force microscopy (AFM, Bruker Dimension Icon, Billerica, USA). The Fizeau interferometer (HI-MARC, NJ, CHN) was applied to measure the surface contours and radii of curvature of the substrates before and after deposition [18]. Next, the intrinsic stress of the multilayers was calculated using the Stoney equation [19]. X-ray diffraction measurements (XRD, Bruker D8 Advance, Karlsruhe, GER) were used to study the crystallization change of multilayers. The measurement was performed using the theta–2theta (θ–2θ) scan and grazing incident x-ray diffraction (GIXRD) modes. GIXRD was performed at a grazing incident angle of 1°. The crystal structure was analyzed by comparing the angular positions of diffraction peaks with the powder diffraction file (PDF) of the International Centre for Diffraction Data (ICDD). The layer microstructure and crystallization change were further characterized by transmission electron microscopy (TEM). The TEM samples were prepared by a focused ion beam and the measurements were performed with a FEI Talos F200X.
The high-angle annular-dark-field measurement (HAADF) was also used to investigate the internal structure, which used an annular detector to locate scattered electrons at high angles [20, 21]. Selected area electron diffraction (SAED) measurements were used to characterize the crystallization of the multilayer and a large electron beam was used to cover the whole multilayer stack during the measurement.

3. Results

The GIXR curves of S1 to S6 Ru/C multilayers are shown in figure 1. All GIXR curves were fitted to obtain the layer thicknesses, interface widths, and layer densities. The detailed measurements and structural parameters are listed in table 1. It can be observed that the Bragg peaks gradually shifted to larger angles and peak reflectivity reduced when the period decreased. Figure 2 has been drawn to analyze the change in the average interface width more intuitively. The interface width listed in table 1 and the red curves in figure 2 showed that the average...
The interface width of the Ru/C multilayer increased with decreasing period. The multilayer with a period of 6.2 nm (S6) had an average interface width of 0.27 nm, which was used as a reference for high quality multilayer. The multilayer with \( d = 2.5 \) nm (S4) showed an average interface width of 0.28 nm. After the period reduced to below 2.5–2.3 nm, the average interface width of the Ru/C multilayer increased evidently. Asymmetric interfaces seemed to be observed from the fitted data of all six samples where the interface width of Ru-on-C was slightly larger. It should be noted that the value of interface width fitted in this paper is the RMS value, which is widely used in the community and consists of both the roughness and intermixing. The real width of the transition area between the two materials is several times larger. This will be further discussed in the following.

Based on the GIXR fitted models, refractive index profiles (real part) of S1 to S5 samples were calculated using GenX software (GenX-2.4.10) \[22\] as depicted in figure 3. The refractive index profiles provide a more realistic description of the electron density/optical constants variation inside the layer structure. The ideal profile of the Ru/C multilayer with bulk density and perfect interface (dash line) is also shown for comparison. For sample S6, there was a plateau of the refractive index profile at the center of both C and Ru layers. The index of C was close to the standard values obtained with the bulk material density. However, that of Ru was larger than the standard values. This can be attributed to the lower density of the layers resulting from the slightly porous layer structure and the interdiffusion of C into Ru. The fitted layer density of Ru was approximately 85% of the bulk value (table 1). For sample S5, the plateau area significantly reduced due to the smaller thickness. The index of Ru at the plateau area was a little larger than S6 which can be affected by the interfacial diffusion of carbon atoms. For sample S4 with \( d = 2.5 \) nm, the index at the center of each layer and the slope of the index profile at the transition areas were still the same as that for S5; however, the plateau disappeared. It indicated that there can be no pure Ru or C layer left in the stack and the interdiffusion started to penetrate through an entire period. For
sample S3 with $d = 2.3$ nm, the index difference between Ru and C layers decreased obviously, leading to a reduced optical contrast of the multilayer compared with the thicker ones. The slope of the profile at transition areas also decreased slightly. It indicated that intermixing between Ru and C began to increase at this layer thickness. For sample S2 with $d = 1.9$ nm, the index profile further deviated from that of thick multilayers (S5) with lower optical contrasts and smaller transition slopes. At $d = 1.5$ nm (S1), the optical contrast decreased to a significantly low amplitude, implying an almost complete intermixing. The low optical contrast and large interface width will significantly reduce the experimental reflectivity of multilayers.

The surface morphologies of samples S1 and S5 can be seen in the AFM images in figure 4. The surface contour image was measured over a 2 $\mu$m $\times$ 2 $\mu$m area, with 512 $\times$ 512 points. The average roughness values of S1 and S5 were approximately 0.25 nm (RMS) and 0.23 nm (RMS), respectively, which was similar to that reported by Störmer [15]. The surface roughness remained significantly small for various multilayers, which indicated that the layers with even sub-nanometer thicknesses were grown in a significantly smooth mode.

The internal stress of the multilayer structures was determined by the sample-curvature measurement. To improve the accuracy of the results, two samples of each structure were deposited using the same process. The average stress values for S3, S5, and S6 are depicted in figure 5. The stress of multilayers with various periods remained in a compressive state. The stress decreased gradually from $-685.36$ MPa to $-82.57$ MPa with decreasing period. The reduced stress with a smaller period was also observed in other multilayers such as W/Si [23]. The stress of the Ru/C multilayer with a period of 3.1 nm (S5) was $-319.95$ MPa, which was significantly less than that of the Ru/B$_4$C multilayer [8]. The small compressive stress of the small period multilayer can avoid obvious deformation of the high-precision mirror substrate in the application.

The XRD results with fitted curves of S1, S5, and S6 samples are shown in figure 6. The intensity of the diffraction peaks in figures 6(a), (b) was weak and the signal-to-noise ratio was not good, which was mainly due
to the small number of bilayers \((N = 20–30)\) of the three samples. Figure 6(a) showed the XRD pattern measured in the GIXRD scan mode, which was applied to analyze the structure of the grains with the crystallographic plane not parallel to the multilayer surface; the diffraction peaks of S1 and S5 were almost the same, whereas the diffraction peak of S5 showed a slightly high intensity. Based on the fitted results, the peaks were mainly identified as coming from \((002)\) crystallographic planes and partly from \((100)\) crystallographic planes of hexagonal lattice of polycrystalline Ru, while the C layers of all samples are amorphous. For S6, the diffraction peak shifted slightly toward larger angles, which can be identified as diffractions from \((002)\) and \((101)\) crystallographic planes of Ru while the one from \((100)\) crystallographic plane of Ru disappeared. The grain size of S6 also increased slightly based on the narrower peak. Figure 6(b) showed the XRD results measured in the \(\theta–2\theta\) scan mode, in which only the crystallographic plane parallel to the multilayer surface can be detected. Sample S6 had a strong peak, which corresponded to the diffractions from \((002)\) and \((101)\) crystallographic planes of Ru. The diffraction intensity reduced obviously with decreasing period, which led to a very weak peak of S5 corresponding to the diffraction from \((002)\) crystallographic plane of Ru. For S1, the diffraction intensity was almost at the noise level, indicating that the Ru/C multilayer with \(d = 1.5\) nm (S1) had almost no Ru grains with crystallographic planes grown parallel to the multilayer interface. A decreased crystallization with decreasing period was also observed in W/Si multilayers [23]. Based on the fitting results, the interplanar distances of the \((002)\) and \((101)\) crystallographic planes of Ru was consistent with that provided by the PDF of ICDD (0.214 nm and 0.206 nm, respectively). While the one of \((100)\) crystallographic plane of Ru was about 0.85% smaller than the theoretical one (0.234 nm), which may either be caused by the intercalation of C atoms into the Ru grains or a measurement error.

Based on the diffraction measurement results, the reduction in stress as the period decreased from 6.2 nm to 3.1 nm can be further explained by two reasons: (i) the reduced crystallization and different orientation that may bring less strain from the grains [24]; and (ii) the less atomic peening effect during the growth of each Ru and C layer after the initial growth stage, as the thickness of each layer was significantly reduced [19, 23]. As the period decreased from 3.1 nm to 1.5 nm, the interfacial stress was considered to be more dominant than the intrinsic stress of each layer. The increased intermixing of Ru and C atoms may lead to a small stress for multilayers with smaller periods [23].
To study the x-ray performance of the Ru/C multilayer, multilayers with a saturated number of bilayers were characterized. The detailed structural parameters are presented in Table 2. The GIXR curves of all three samples (S2-M, S4-M, and S5-M) are shown in Figure 7(a). The measured first-order Bragg peaks with the fitted data are shown in Figure 7(b).

Table 2. Detailed structural parameters of Ru/C multilayers with saturated number of bilayers.

| Sample No. | Bilayers | Average Thickness (nm) | Period Drift (nm) | Average Interface Width (σ (nm)) | Δθ/θ (%) | Experimental Reflectivity (%) |
|------------|----------|------------------------|-------------------|----------------------------------|----------|-------------------------------|
| S2-M       | 300      | Ru 0.902               | 0.056             | 0.395                            | 1.28     | 21.8                          |
|            |          | C 0.967                |                   |                                  |          |                               |
| S4-M       | 150      | Ru 1.270               | 0.045             | 0.294                            | 1.72     | 54.2                          |
|            |          | C 1.256                |                   |                                  |          |                               |
| S5-M       | 100      | Ru 1.542               | 0.045             | 0.287                            | 2.47     | 64.9                          |
|            |          | C 1.607                |                   |                                  |          |                               |

Figure 7. GIXR curves at $E = 8.04\,\text{keV}$ of Ru/C multilayers (a) S2-M, S4-M, and S5-M. The measured first-order Bragg peaks with the fitted data are shown in (b).

To study the x-ray performance of the Ru/C multilayer, multilayers with a saturated number of bilayers were characterized. The detailed structural parameters are presented in Table 2. The GIXR curves of all three samples (S2-M, S4-M, and S5-M) are shown in Figure 7(a). The measured first-order Bragg peaks are given in Figure 7(b) together with the fitted data. Owing to the long sputtering time of the thick multilayers, thickness drifts were included in the layer structure model to account for the broadening of the Bragg peaks.

For the Ru/C multilayer with a period of 3.1 nm (S5-M), the peak reflectivity was 65%, which was almost the same as that of the nitridated Ru/B,C multilayer [8] and that reported by Störmer [4, 15]. Based on the full width half maximum of the peak width, the angular/energy bandwidth was estimated to be $\Delta\theta/\theta = 2.5\%$. The fitted average interface width was almost the same as the multilayers with smaller bilayers. No accumulation of interface roughness was observed when the number of bilayers increased. The period drift was approximately 45 pm. For sample S4-M, a high reflectance of 54% was demonstrated with an angular bandwidth of 1.7%,
which was similar to that of the Pd/B,C multilayer [7]. The peak reflectance can be 3%–4% higher (reaching 58%) if the divergence of the incident beam (0.007°) can be reduced to a few microradians, which can be easily achieved in the synchrotron beamlines. The fitted period drift was also 45 pm. If the drift is also corrected, the peak reflectance of S4-M can reach 65%. The theoretical reflectance curve using the fitted layer structure, but with the collimated incident beam and no thickness drift, is also presented in figure 7(b). It indicated that the Ru/C multilayer with \( d = 2.5 \text{ nm} \) (S4-M) still had a significantly good optical property. When the period decreased to 1.87 nm (S2-M), reflectivity significantly dropped to 22%. A relatively large period drift of 56 pm was found from the fitted result, which can be caused by a significantly large number of deposited bilayers \((N = 300)\). The average interface width was also increased to 0.40 nm. Owing to the smaller period of 1.87 nm and the resultant narrower bandwidth, the effects of the thickness drift and interface width on the reflectance were more prominent, which caused low reflectance. Nevertheless, if the beam divergence and thickness drift can be corrected, the peak reflectance of S2-M with the current interface structure can be close to 42%. With further optimization, the Ru/C multilayer with a period of approximately 1.9 nm may still be useful for applications in hard x-ray monochromators and imaging systems.

To analyze the internal structure of the multilayers with larger number of bilayers further, XRD measurements were performed on S2-M, S4-M, and S5-M samples using two scan modes. The diffraction intensity of all samples (figures 6(c), (d)) was stronger than the one shown in figures 6(a), (b), which can be easily understood from the larger number of bilayers \((N = 100–300)\). For the GIXRD mode (figure 6(c)), the diffraction peaks of samples S2-M and S4-M were almost the same, which can be identified as diffraction from (100) and (002) crystallographic planes of Ru. Whereas that of S5-M slightly shifted to higher angles centered at 42.15°, and the diffraction peak of the (002) crystallographic plane of Ru was enhanced. For the 0–20 mode (figure 6(d)), the diffraction peaks of S2-M and S4-M were also similar; while the intensity of S5-M increased and its peak position shifted to 42.15°, it is mainly coming from the diffraction of (002) crystallographic plane of Ru. The result is consistent with the multilayer with less bilayers. Through fitting the diffraction peaks of samples S2-M, S4-M, and S5-M, it is found that the size of grains with crystallographic planes non-parallel to the layer interface increased slightly from \( \sim 1.3 \text{ nm} \) to \( \sim 1.7 \text{ nm} \) (both for (100) and (002) crystallographic planes of Ru) as the multilayer period increased from 1.9 nm to 3.1 nm, and the grains with crystallographic planes parallel to interfaces remained the same size of \( \sim 1.8 \text{ nm} \). Overall, the polycrystallization state changed minorly especially as the period decreased from 2.5 nm to 1.9 nm which should not bring significant effect to the interface structure.

High-resolution TEM (HRTEM) was performed for both samples of S2-M and S5-M. The intensity of the HAADF measurement is a function of the atomic number; atomic number contrast images are obtained [25]. Heavy elements produce strong signals, resulting in a bright contrast. On the contrary, light elements result in a dark contrast, which is contrary to HRTEM. As a complementary method, HAADF can clearly show the positions of Ru and C atoms and the thickness ratio of various elements.

The HAADF and HRTEM results of S2-M and S5-M are presented in figure 8. The HAADF results in figures 8(a), (b) indicated that the ratio of the Ru layer thickness to the period was close to 0.5 for both S2-M and S5-M. The interface of S5-M was relatively flat and sharp. However, the interface of S2-M was wavy with small fluctuations. The local variations of the imaging contrast in each layer indicated an intermixing of Ru and C atoms. This can be further observed in the HRTEM images (figures 8(c), (e)). In figure 8(e), continuous Ru and C layers with a thickness of approximately 1.5 nm were formed despite interdiffusion between the layers. The SAED pattern displayed a broad diffraction ring (figure 8(f)), which can be attributed to (002) and (100) crystallographic planes of Ru, according to the calculated interplanar distances \((0.213–0.237 \text{ nm})\). In figure 8(c), a noncontinuous growth of layers with a thickness of approximately 0.9 nm was observed and a complete intermixing occurred at some positions (indicated by circles). This explained the significantly reduced optical contrast and deviation of the reconstructed refractive index from the layers with bulk density (figure 3). An asymmetric interface was clearly observed in S5-M sample where the interface width of Ru-on-C was slightly larger. This is consistent with the fitting results of the GIXR measurements. It can be caused by the strong bombardment of heavy Ru atoms and the polycrystalline structure of Ru layers which bring more interdiffusion at Ru-on-C interface than C-on-Ru [26]. Compared to S5-M, an even weaker diffraction ring of S2-M was shown in the SAED image (figure 8(d)), which can be also attributed to (002) and (100) crystallographic planes of Ru. This is consistent with the XRD results.

4. Discussion

Based on our experimental results, the evolution of the microstructure and initial growth state of Ru/C multilayers are summarized and discussed as below. Only a small part of the periodic multilayer is shown here in figure 9.
For the Ru/C multilayer with period \( d \leq 1.9 \) nm, i.e., the layer thickness was no more than 0.95 nm, the deposited Ru and C atoms were not enough to form a continuous layer, and there was severe intermixing between Ru and C across the entire multilayer (figure 9(a)). This actually resulted in a complete mixture of Ru and C with gradually varied concentration of the two materials (figures 8(a), (c)) and significantly low optical contrast (figure 3) [27]. Nevertheless, this may not indicate an island growth mode of Ru or C layers at the initial stage because the surface morphology of the ultrathin multilayer was still very smooth, as revealed by AFM measurements (figure 4) [28]. The intermixing of two materials can relax the mismatch of the two structure and lead to a small stress (figure 5).

When the period increased to \( d = 2.3–2.5 \) nm, i.e., the layer thickness reached 1.0–1.2 nm, the intermixing of Ru and C reduced gradually with an improved optical contrast (figure 3). The obtained refractive indices at the center of Ru and C layers were similar to those of multilayers with a larger period. The average interface width significantly reduced to 0.32–0.28 nm (figure 2). The sample with \( d = 2.5 \) nm also showed a high reflectance of 54\% at 8 keV. Nevertheless, as there was no plateau of the index profile inside each layer, there was still no layered structure in the stack. Zameshin et al studied the growth properties of Ru on the C layer using the low-energy ion-scattering technique. It was found that the change in surface atomic composition with an increase in the thickness of the Ru layer during deposition exhibited different behaviors at below and above 1.0 nm [29]. The threshold of the Ru thickness of \( \sim 1.0 \) nm was very close to our results, which implied that the diffusion effect of C can be significantly reduced beyond this thickness value (figure 9(b)).

When the period of the multilayer increased to 3.1 nm, more complete layers of Ru and C were formed with relatively sharp and flat interfaces (figures 8(b), (d)). The interface width reduced slightly compared to the case of \( d = 2.5 \) nm. A plateau of the refractive index profile was observed at the center of Ru and C layers (figure 3),

![Figure 8. HAADF (a), (b), HRTEM (c), (e) and SAED (d), (f) results of Ru/C multilayers S2-M and S5-M.](image-url)
indicating the existence of pure Ru and C thin layers or a thin mixed layer of Ru and C of constant composition. Based on the observed deep intermixing and segregation effects of C in [29], the latter one can be closer to reality. This was also supported by the still obvious interdiffusion at the Ru-on-C interfaces seen in the TEM image (figure 8(d)). Nevertheless, the average interface width of the multilayer was very small and it led to high reflectance at 8 keV (figure 7). Notably, all multilayers with \( d = 3.1 \text{–} 1.5 \text{ nm} \) displayed similar polycrystalline structure of Ru, and the boundaries among tiny Ru grains could facilitate the diffusion of C atoms.

5. Conclusions

Ru/C multilayers with small period of 6.2 nm down to 1.5 nm were fabricated and systematically characterized. It was found that the average interface width of the multilayers started to increase as the period reduced to below 2.5 nm. The surface roughness and polycrystalline structure of all Ru/C multilayers with periods smaller than 2.5 nm remain almost the same, which indicate the roughness and nanograin growth should not be the reason for the increased interface widths. The TEM results showed a clear layered structure of the multilayer with period of 3.1 nm while strong interdiffusion and noncontinuous layer growth of the multilayer with period of 1.87 nm. The measured reflectivities (at \( E = 8.04 \text{ keV} \)) of the Ru/C multilayer with \( d = 2.5 \text{ nm} \) and 3.0 nm reached quite high values of 54% and 65%, respectively. For the multilayer with period of 1.87 nm, the reflectivity significantly dropped to 22% due to the poor interface structure and thickness drift over the stack. Based on the comprehensive characterization, it is concluded that the severe intermixing is the main reason for the large interface width of the ultrathin multilayers, and the actual internal structure of the multilayer with period smaller than 2.5 nm is a complete mixture of Ru and C with gradually varied composition. The smallest layer thickness of Ru and C for growing with relatively small intermixing using the current DC magnetron sputtering technique is 1.0 nm–1.2 nm. Further optimization of the deposition process, like optimizing the kinetic energy of the deposited Ru and C atoms to promote an earlier formation of a continuous layer, may further reduce this thickness threshold. This study provides useful guidance for the future development of ultrathin Ru/C multilayers, in order to work at larger grazing incident angle, higher working energy, with higher energy resolution, than currently achieved. The result can also act as a reference for the comparison and selection of high performance multilayer mirrors for the hard x-ray region.

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Data availability statement
The data that support the findings of this study are available upon reasonable request from the authors.

Ethical statement
The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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