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Effects of sputtering power and annealing temperature on surface roughness of gold films for high-reflectivity synchrotron radiation mirrors

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Abstract Gold films deposited by direct current magnetron sputtering are used for synchrotron radiation optics. In this study, the microstructure and surface roughness of gold films were investigated for the purpose of developing high-reflectivity mirrors. The deposition process was first optimized. Films were fabricated at different sputtering powers (15, 40, 80, and 120 W) and characterized using grazing incidence X-ray reflectometry, X-ray diffraction, and atomic force microscopy. The results showed that all the films were highly textured, having a dominant Au (111) orientation, and the film deposited at 80 W had the lowest surface roughness. Subsequently, post-deposition annealing from 100 to 200 °C in a vacuum was performed on the films deposited at 80 W to investigate the effect of annealing on the microstructure and surface roughness of the films. The grain size, surface roughness, and their relationship were investigated as a function of annealing temperature. AFM and XRD results revealed that at annealing temperatures of 175 °C and below, microstructural change of the films was mainly manifested by the elimination of voids. At annealing temperatures higher than 175 °C, grain coalescence occurred in addition to the void elimination, causing the surface roughness to increase.

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1 Introduction

Metal single layers are widely used as X-ray mirrors in synchrotron radiation beam lines [1, 2]. In comparison with other metal single layers, gold films have some distinct advantages. The first and most crucial point is that gold films have chemical stability and high reflectivity at the energy region of 50–2000 eV [3]. Second, gold films can be cleaned by plasma cleaning for reuse. The long-term operation of synchrotron beam lines will be affected by contamination with hydrocarbon molecules on the optics in the beamline vacuum chambers, which is responsible for a loss of reflectivity of the optics [4, 5]. For the second phase of construction of the Shanghai synchrotron radiation facility [6], mirrors that can operate in the 50–2000 eV energy range are required. Thus, it is important to decrease the surface roughness and improve the reflectivity effectively for the application of gold films.

Optimization of the deposition parameters [7] and post-deposition annealing [8–11] are frequently used to decrease the surface roughness of metallic films. Typical deposition parameters include base pressure, sputtering pressure, and substrate temperature. Tang et al. [12] prepared 500-nm-thick gold films using magnetron sputtering. Results showed that the surface roughness decreased as the deposition temperature changed from 100 to 200 °C and then increased at deposition temperatures beyond 200 °C, which indicated that the deposition temperature has a significant influence on the surface morphology. However, there are limited data available on the effect of the sputtering power on the surface roughness of gold films, which may also influence the surface roughness. Annealing has been widely used to regulate the microstructure of as-deposited metallic films. Several studies have been devoted to the annealing of evaporated gold films, but not many have studied the annealing of gold films deposited by magnetron sputtering [13]. Plaza et al. [14] investigated annealing effects on the microstructure of sputtered gold layers. Their results demonstrated that annealing affects the grain structure of the layers; an annealing temperature of 300 °C results in a larger grain size and smoother surfaces, but generates some cracks in the film surface. Huang et al. [15] fabricated gold films with thicknesses of approximately 110 nm, which were sputter-deposited on unheated glass substrates coated with a sputtered binding layer of Cr. It was found that when the annealing temperature reaches 200 °C and the annealing time exceeds 30 min, chromium atoms markedly diffuse into the gold layer, which may affect the characteristics of the gold films. In conclusion, gold films with a thickness more than 100 nm have been intensively studied in recent decades. However, gold films with a thickness of 50 nm are presently required for the mirrors for the Shanghai synchrotron radiation facility.

In the present work, the effect of sputtering power on the surface roughness was studied with the aim of increasing the reflectivity of gold films with 50 nm thickness deposited by direct current (DC) magnetron sputtering. Further investigations were then conducted to examine the effect of annealing at 100–200 °C on the surface roughness of the gold films.

2 Experimental details

All the films were fabricated by DC magnetron sputtering. The 4-inch (101.6 mm) circular sputtering sources face the substrate. The perpendicular distance between sputtering sources and substrate was 9 cm. The thickness of the films depended on the time that the substrate stayed above the sputtering sources.

All samples were deposited on super-polished Si (100) substrates (the surface roughness was less than 0.2 nm) that were used for structure characterizations. A 5-nm-thick transition chromium layer was deposited first, followed by the gold film. The base pressure before deposition was 7.0×10^{-5} Pa. During the deposition process, argon with a purity of 99.999% was used as the sputtering gas at a working pressure of approximately 0.15 Pa.

To investigate the effect of the sputtering power on the surface roughness of gold films, the ~ 50-nm-thick films were deposited at powers of 15, 40, 80, and 120 W. To decrease the surface roughness of gold films further, the films with the lowest roughness were selected by grazing incidence X-ray reflectometry (GIXR) and atomic force microscopy (AFM). Then, the as-deposited samples were annealed in a vacuum of 1.0×10^{-4} Pa at 100, 125, 150, 175, or 200 °C for 1 h.

GIXR was carried out on a D1 high-resolution X-ray diffractometer using a Cu-Kα source (λ = 0.154 nm). The fitting calculations of GIXR curves were carried out by using IMD software [16] to determine structural information about the gold films, including thickness, surface roughness, and density. AFM was used to obtain the surface morphology information with a scanning probe microscope using a Bruker Dimension Icon system in Peak-force Tapping mode. The AFM has a noise level of 0.002 nm and the ability to measure an ultra-smooth surface with high-spatial-frequency roughness as low as the sub-nanometer scale. Power spectrum density (PSD) functions of the surfaces obtained from AFM images were used to determine the differences in surface morphologies of the gold films. In addition, the root-mean-square (RMS) micro-roughness can be calculated by integrating the PSD curve. All samples were measured three times at different positions, and then, their average RMS micro-roughness values were calculated. X-ray diffraction (XRD) was performed on a D8 Advance X-Ray diffractometer using Cu-Kα source (λ = 0.154 nm) with θ–2θ scanning.

3 Results and discussion

The GIXR curves of the four samples and the fitting data are shown in Fig. 1 and Table 1. As can be seen, the surface of the gold film deposited at 15 W was rough, with a

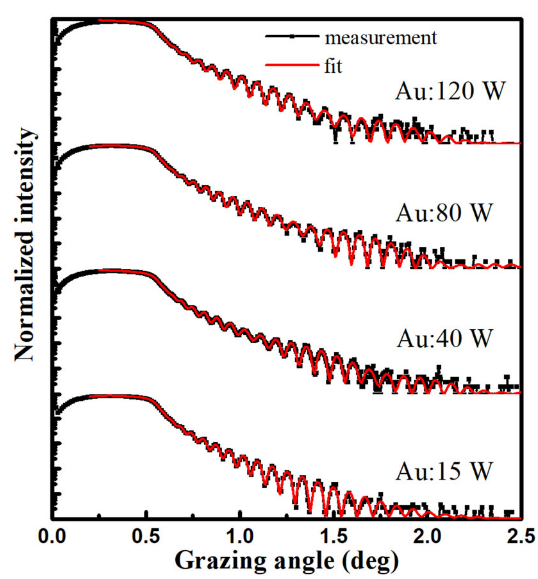

Fig. 1 (Color online) GIXR measurements (black spots) and fitting curves (solid red lines) for gold films with different sputtering powers

Table 1 GIXR fitting data for all the gold films with different sputtering powers

Power (W)	Thickness (nm)	Density (%)	Surface roughness (nm)	Deposition rate (nm/s)
15	49.10	99.99	0.96 ± 0.01	0.19
40	48.23	99.99	0.76 ± 0.03	0.51
80	47.34	99.99	0.69 ± 0.02	1.03
120	44.28	99.99	0.80 ± 0.03	1.57

surface roughness of approximately 0.96 nm. The surface roughness decreased to a minimum value of 0.69 nm from 15 to 80 W and then increased to 0.80 nm from 80 to 120 W. The deposition rates of these four samples are given in Table 1, showing that the sputtering rate increased as sputtering power increased.

Figure 2 shows 1 μm × 1 μm AFM images of the gold films deposited at: (a) 15 W, (b) 40 W, (c) 80 W, and (d) 120 W. The as-deposited films are characterized by dome-like surface grains with voids distributed randomly around the grain boundaries. With the increase in sputtering power, it can be seen from the AFM diagrams that there are very slight changes in grain size except for the sample deposited at 15 W, for which the grain size is obviously larger than that of the other three samples. To compare the surface morphology of the gold films quantitatively, PSD curves were calculated from the AFM images. Because the surface of the metallic single layer is isotropic, there is no difference between the horizontal and vertical directions of the PSDs. The PSD functions in the horizontal direction shown in Fig. 2e were directly obtained from the measured AFM diagrams shown in Fig. 2a–d. For the gold film deposited at 15 W, the PSD over the entire spatial frequency range was obviously higher than that of gold films deposited at 40, 80, or 120 W. When the sputtering power increased from 15 to 80 W, the GIXR fitting results exhibited a decreased surface roughness. The PSD of the gold film deposited at 80 W was lower than that of the other samples. Thus, a sharp decrease in RMS micro-roughness from 0.745 to 0.480 nm appeared, while the sputtering power increased from 15 to 80 W. As for the gold film deposited at 120 W, the PSD curve and RMS micro-roughness value are nearly identical to those of the 40 W film. From Fig. 2f, the variation trend of surface roughness tested by AFM is same as that obtained using

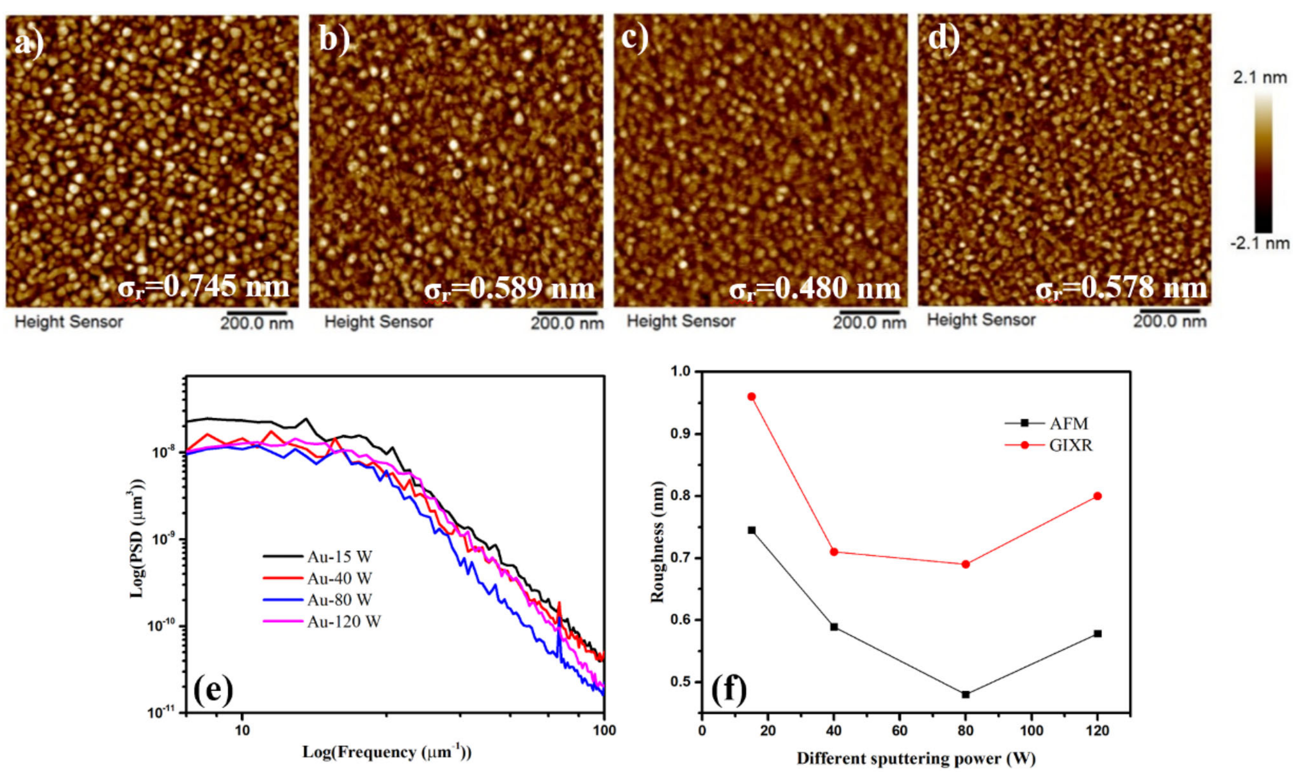

Fig. 2 (Color online) **a–d** AFM images of the surface of the gold films deposited at different sputtering powers: **a** 15 W, **b** 40 W, **c** 80 W, **d** 120 W. **e** PSD curves of the AFM images shown in **a–d**. **f** Surface roughness of gold films versus sputtering power

GIXR, which proves that the surface roughness fitted from GIXR is reliable.

The XRD patterns of as-deposited gold films are shown in Fig. 3. The results of all coated samples reveal that only diffraction peaks of gold are visible in the XRD patterns. This means that there are no alloying phases in the gold film, and the interlayer of Cr is too thin to be detected by conventional XRD. Two peaks appear for all samples at the diffraction angles of approximately 38.28° and 81.72°. The crystal planes can be determined by referring to the standard PDF cards, which reveal that all the films are highly textured, with the Au (111) orientation dominating, which is consistent with a previous study [12]. At 15 W of deposition power, the diffraction peak was strong, and the full width at half maximum (FWHM) of the diffraction peak was narrow. With increased sputtering power, the diffraction peak became weaker, and the FWHM of the diffraction peak became wider. However, when the sputtering power reached 120 W, the FWHM of the diffraction peak became narrower and the crystallization was again enhanced. The size of crystal particles can be deduced from the Scherrer formula:

$$D = \frac{\kappa\lambda}{B\cos\theta}, \quad (1)$$

where D is the mean size of the crystal particles perpendicular to the crystal plane direction; κ is a dimensionless shape factor with a value of 0.89; λ is the X-ray wavelength, which is equal to 0.154 nm; B is the FWHM of the diffraction peak, whose unit is radians; and θ is the Bragg angle. The crystal size perpendicular to the (111) crystal plane directions of the films deposited at 15, 40, 80, and 120 W were 24.68, 22.66, 22.18, and 23.76 nm, respectively. This means that the larger crystal particles caused

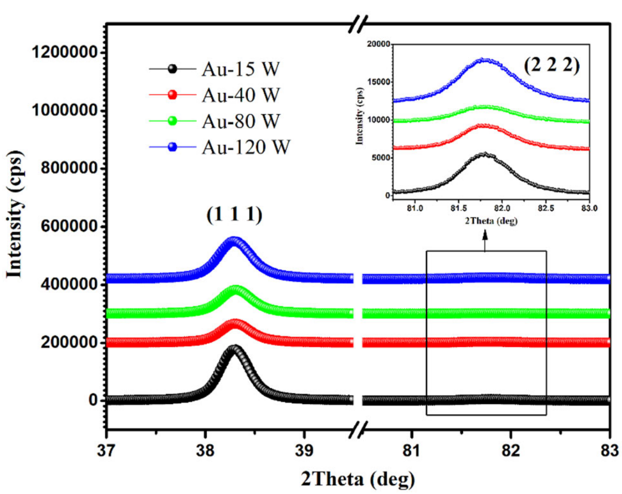

Fig. 3 (Color online) XRD results of gold films deposited at different sputtering powers

the gaps between the crystal particles to be larger, which directly increases the surface roughness [17].

Based on the results obtained from different sputtering powers, post-deposition annealing at 100–200 °C in vacuum was performed on the films deposited at 80 W. Figure 4a–d shows 1 μm × 1 μm AFM images of gold films annealed at 100, 125, 175, and 200 °C, respectively. Compared to the as-deposited film shown in Fig. 2c, all the films showed a general trend for the small grains to form agglomerates, which means the annealing reduces the roughness to form a considerably smoother surface [14]. Moreover, it is notable that the grain undulation of the sample annealed at 125 °C was gentler than that of other samples.

In Fig. 4e, the PSD functions in the horizontal direction are displayed. For the gold film annealed at 125 °C, the PSD over the entire spatial frequency range was obviously lower than that of gold films annealed at 100, 175, and 200 °C. For the gold film annealed at 200 °C, when the spatial frequency was more than 39 μm^{-1}, the PSD curve was higher than that of other samples. In addition, the RMS micro-roughness was calculated by integrating the PSD curve. The RMS roughness decreased down to a minimum value of 0.413 nm from 100 to 125 °C and then increased to 0.486 nm from 125 to 200 °C. In Fig. 4f, the variation of surface roughness difference before and after annealing obtained from the GIXR and AFM images is presented. The difference of surface roughness $\Delta\sigma_r$ shown in the figure is obtained by

$$\Delta\sigma_r = \sigma_a - \sigma_b, \quad (2)$$

where σ_a and σ_b are surface roughness after annealing and before annealing, respectively. When the annealing temperature was higher than 100 °C and lower than 200 °C, the roughness of the gold film after annealing was lower than that before annealing. When the annealing temperature was lower than 100 °C or higher than 175 °C, the change in surface roughness was not obvious. It would appear that the surface roughness is closely related to the change in the morphology of the gold film [18]. Compared to the as-deposited film, the gaps between grains were gradually eliminated with the increase in the annealing temperature (from 125 to 175 °C), so the film surface became smooth [19]. However, the surface roughness at annealing temperatures around 100 and 200 °C exhibited little change.

To study the microstructure changes and the possible mechanism for the change in the surface roughness, the samples were measured with XRD and the results are shown in Fig. 5. All the films show predominately Au (111) orientations, indicating that the grain orientation was not affected by annealing. In addition, the position of the (111) peak shifts toward the right, which indicates that

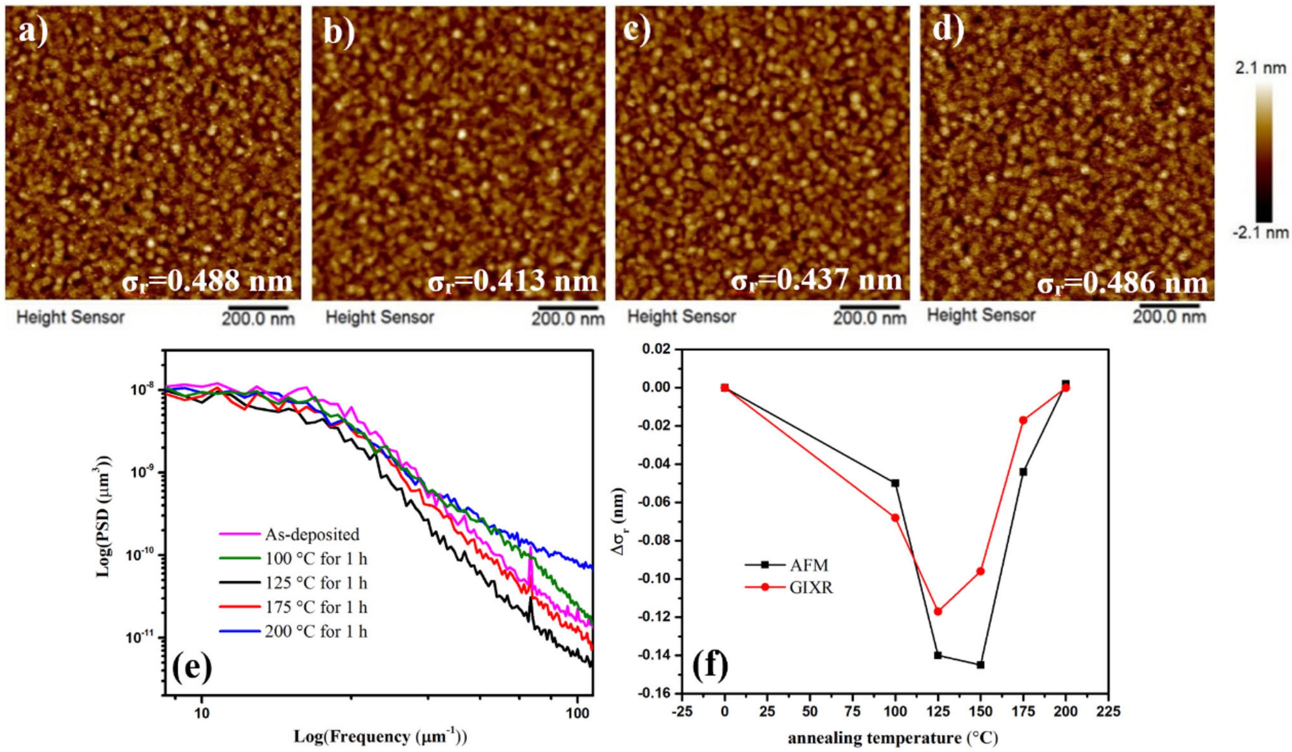

Fig. 4 (Color online) **a**–**d** AFM images of the surface of the gold films with different annealing temperatures: **a** 100 °C, **b** 125 °C, **c** 175 °C, and **d** 200 °C. **e** PSD curves of the AFM images shown in **a**–**d**. **f** Surface roughness difference before and after annealing of gold films versus annealing temperature

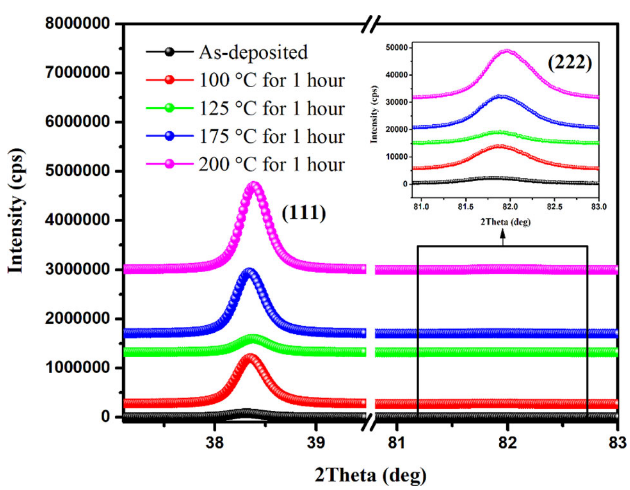

Fig. 5 (Color online) XRD results of gold films with different annealing temperatures

tensile stress increased with the annealing temperature [19]. Moreover, the crystallization becomes more obvious after annealing, and the (111) textures are also enhanced. The grain size ranged from 22.18 to 28.68 nm and reached a minimum at the annealing temperature of 125 °C. This effect agrees with prior observations on thermally evaporated Au films, where annealing has been reported to promote grain growth [20] and to form smoother surfaces.

4 Conclusion

We prepared four gold films by DC magnetron sputtering with different sputtering powers (15, 40, 80, and 120 W) to optimize the surface roughness of the gold films. The results show that the change in power has an effect on the surface roughness of gold films. These films were first characterized by GIXR fitting, which showed that the surface roughness is a maximum at 15 W of sputtering power and is a minimum at 80 W. The samples were also characterized using AFM, for which the variation trend of surface roughness is consistent with that of GIXR. The variation trend was interpreted as being caused by the microstructural changes in the gold film surface. For sputtering powers up to 80 W, the crystal particle size of gold films becomes smaller, which causes the gaps between the crystal particles to become smaller resulting in a smoother film surface. Moreover, larger crystal particles also lead to a rougher surface. It is worth mentioning that the optimal sputtering power will be system dependent.

To further decrease the surface roughness of the gold films, we annealed the samples prepared at a sputtering power of 80 W. The annealing temperature in the present study was in the range of 100–200 °C. Once again, the samples were characterized using GIXR and AFM before and after annealing. The results show that the change trend of the roughness difference obtained by these two tests is basically consistent. The crystal structure and the crystal orientation of the films were examined by XRD, which revealed that with increasing annealing temperature, the grain size increases initially, followed by a decrease, but then increases again. For the gold films annealed at moderate temperatures (175 °C and below), the grain growth increases and the grains expand laterally so that the gaps between them become shallow (the gap depth decreases) and smoother, so the surface roughness is lower than that of the as-deposited samples [14, 17]. Further increasing the annealing temperature (higher than 175 °C) did not change the roughness; the influence of gap and grain size on the roughness is offsetting each other; and the influence of grain size dominates, so the film surface becomes rougher.

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