Thickness dependence of grain size and surface roughness for dc magnetron sputtered Au films*

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The grain size and surface morphology of sputtered Au films are studied by x-ray diffraction and atomic force microscope. For as-deposited samples the grain growth mechanism is consistent with the two-dimensional (2D) theory, which gives relatively low diffusion coefficient during deposition. The annealing process demonstrates the secondary grain growth mechanism in which the thickness dependence of grain boundary energy plays a key role. The surface roughness increases with the increase of grain size.

Keywords: grain size, surface morphology, Au film, magnetron sputtering
PACC: 6890, 7360D

1. Introduction

Au films are widely used in surface coating and material sciences due to their chemical inertness and high conductivity. Therefore, investigating the growth kinetics of the material is meaningful for both the basic research and technical application.

Many factors, including growth competition, grain boundary migration, surface diffusion, twinning, etc., may influence the microstructure and morphology of thin films. Wild *et al.*,[1] Dammers and Radelaar,[2] Paritosh *et al.*,[3] developed algorithms for simulating the growth of faceted crystals based on the van der Drift model in two dimensions. Dammers and Radelaar demonstrated that the average grain size D is proportional to the square root of the film thickness d, $D \propto \sqrt{d}$. Thijssen[4] took into account the influence of actual environment of grain growth, simulated the polycrystalline growth in (2+1) dimensions and predicted $D \propto d^{2/5}$. Under the constraint of a constant surface area, Tu *et al.*[5] presented a simple kinetic model of linear rate of grain growth during film deposition. At elevated deposition temperatures when surface diffusion is significant, the growth exponent is independent of temperature and equals 0.25, a value of the critical exponent for noisy Mullins diffusion equation.[6]

The experimental side[7−13] is even more complicated than the theoretical predictions. The reported exponent of the thickness dependence spans from as low as 0.02 up to 0.99. There were cases that different exponents were obtained for different thickness ranges in the same deposition condition.

In the present work, the thickness dependences of grain size and surface roughness in a wide range from $\sim$ 10 nm to $\sim$ 1000 nm are investigated in both the as-deposited and the post-annealed states. We find that in the whole thickness range the exponent for as-deposited samples equals 0.52, supporting the 2D growth mechanism. The exponent decreases to 0.37 for 300-°C annealed films.

2. Experiment

The Au films of 99.995% purity were deposited by dc magnetron sputtering on Si(100) substrates covered by SiO_2 layer of about 500 nm. The SiO_2–Si wafers were pre-cleaned in an ultrasonic cleaner by sequentially using acetone, ethanol and distilled water. Before sputtering, the substrates were baked *in-situ* to 150 °C for about 16 h to degas the surfaces. The system base pressure is approximately 6×10^{-8} Pa. To study the dependences of structural and morphological properties on films thickness, a series of films with thickness 10 nm∼1000 nm were prepared at a constant deposition rate of 0.8 nm·s^{-1} (The thinnest film

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of 10 nm with deposition rate 0.4 nm·s^{-1}). For each thickness, two samples from the same batch were studied, one of them was post-annealed for 5 h at 300 °C in a vacuum of 10^{-4} Pa. The structure of gold films was characterized by x-ray diffractometer (XRD M18AHF, Mac Science Co., Japan) using CuKαI radiation with $\lambda = 1.5405$ Å (1 Å=0.1 nm) and the surface morphology information collected by a scanning probe microscope (SPA-400, Seiko Instrument, Japan) in the atomic force microscope (AFM) mode.

3. Results and discussion

The XRD patterns of as-deposited Au films are shown in Fig. 1(a). Only one sharp peak at 38.19° is observed, indicating that all films are highly textured with Au-(111) orientation parallel to the film surfaces. This implies that the films growing process is self-orientated. The annealing-induced changes of XRD pattern for 100 nm sample are shown in Fig. 1(b). Annealing shrinks the diffraction peak width, or, equivalently, increases the grain size, consistent with AFM image observation.

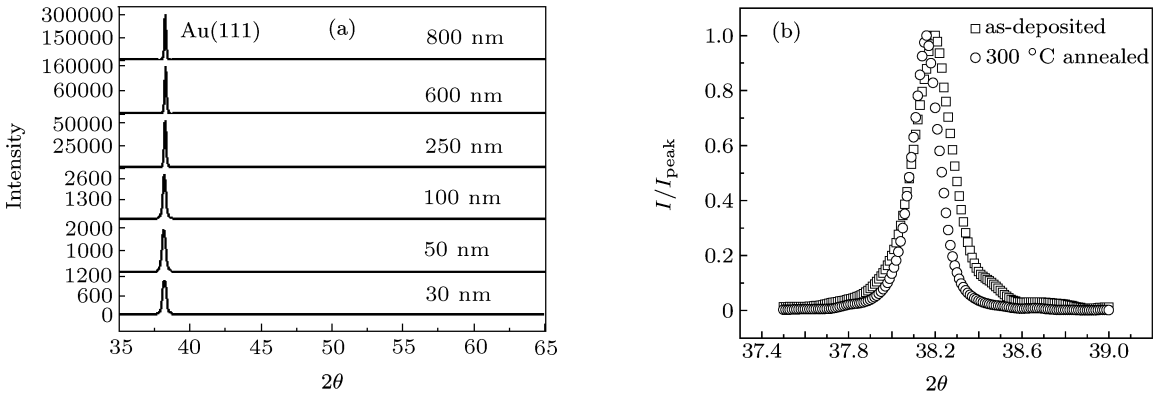


Fig. 1. (a) The XRD of different Au films. (b) The annealing-induced changes of 100-nm Au film.

The surface morphologies of 30 nm, 400 nm, 900 nm are shown in Fig. 2. These images clearly show the nanostructured nature of the films and how the granular structure changes with the film thickness. A distribution of grain sizes is visible in all cases. These surfaces have a characteristic fractal auto-affine symmetry,[14] with a "cauliflower morphology" where smaller grains aggregate to form larger grains.

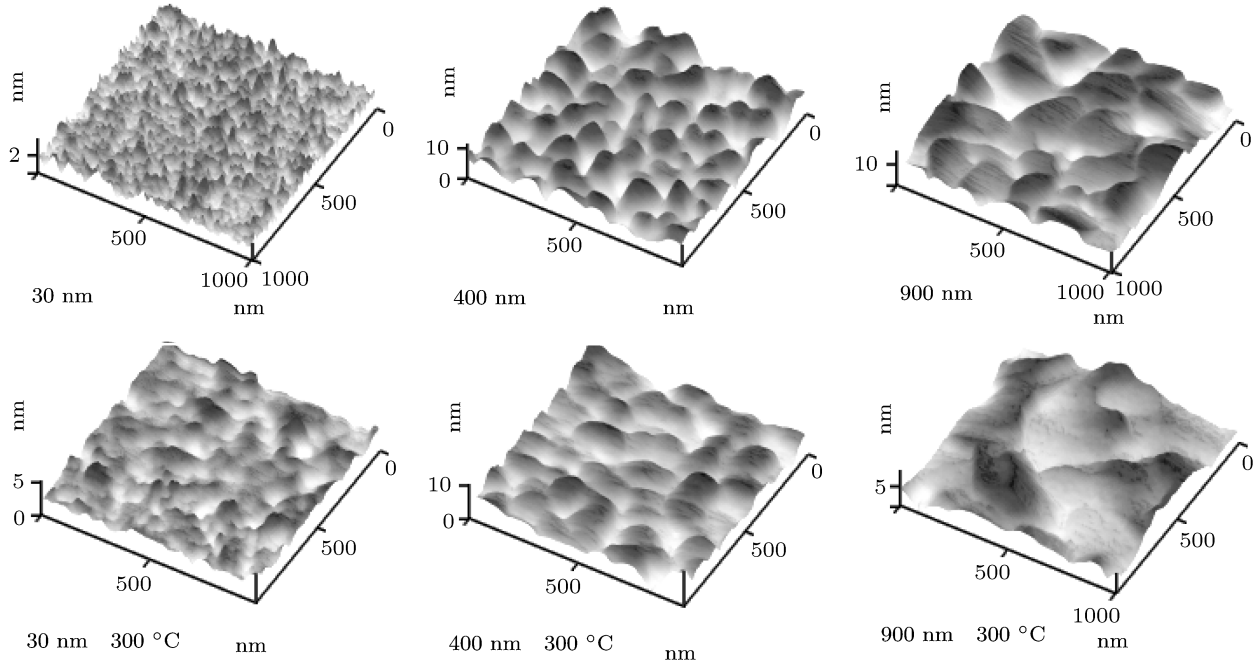


Fig. 2. The AFM topographic images of Au thin films with thickness 30 nm, 400 nm, and 900 nm before and after 300 °C annealing.

In Fig, 3, the variation of grain size before and after annealing obtained from the AFM images is presented in log–log scale. The fit lines shown in the figure are obtained by

$$D(d) = 4.3 \times d^{0.52} \quad \text{(as-deposited)},$$
$$D(d) = 14.8 \times d^{0.37} \quad \text{(300 °C annealed)}.$$

For the as-deposited films, the grain size grows with film thickness in a way close to what the van der Drift 2D model expects. This result shows that for our Au films the main constraint of grain growth is the grain boundary energy, not the thickness effect.[15]

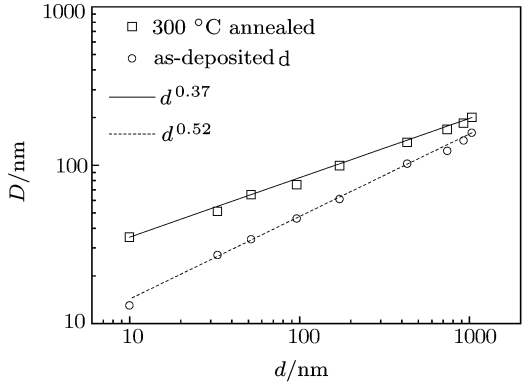


Fig. 3. The variation of grain size before and after annealing obtained from the AFM images and the solid line and dash line are the linear fits (in log–log scale).

Annealing is a process related to secondary grain growth in the film. Thompson discussed the secondary grain growth mechanism and came to the conclusion that the secondary grain growth is driven by the reduction of the total grain boundary energy. Since the grain boundary energy is film thickness-dependent, the secondary grain growth rate increases when the film thickness is reduced. The consequence of this factor is to slow down the thickness-dependence of grain size and hense to decrease the exponent, in our case, to 0.37.

The surface roughness ($R_{\rm rms}$) obtained from AFM varies with film thickness as

$$R_{\rm rms} = 0.0118 \times d^{\beta}, \quad \beta = 0.6, \quad \text{(as-deposited)}$$

the β value obtained here is not far from the exponent for grain size change. However, both values are somewhat larger than that for evapourated films reported in previous work. We believe the relatively low diffusion coefficient in room deposition sputtered films could be one of the reasons for this difference.

The annealing-induced variation of surface roughness is shown in Fig. 4(b). The overall trend, i.e., $R_{\rm rms}$ increases with d, holds, but the data are too scattered to draw a definate conclusion.

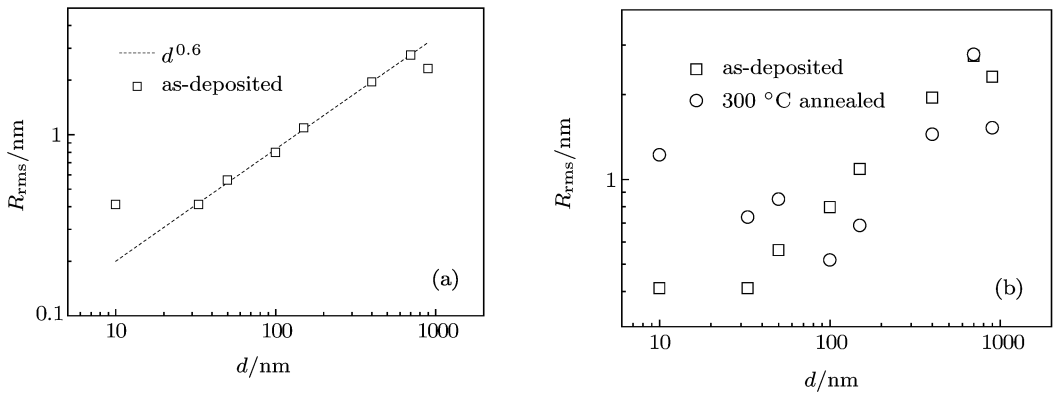


Fig. 4. (a) The surface roughness ($R_{\rm rms}$) variation with film thickness. (b) The variation of surface roughness after 300 °C annealing.

4. Conclusion

In this paper, the morphological grain size and surface roughness as a function of the film thickness of magnetron sputtering Au films have been investigated. The grain growth mechanism revealed here could be useful for further improvement of the film structure and futute applications in suitable cases.

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