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Sol–gel processing for epitaxial growth of ZrO_2 thin films on Si(100) wafers

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Abstract

High quality epitaxial thin films of t-ZrO_2, were fabricated on Si(100) wafers via sol–gel processing. The thin films exhibited high epitaxial quality (FWHM$\sim$0.3°) and excellent surface morphology (rms roughness$\sim$14 Å); thus, they are expected to be very useful as epitaxial buffer layers.

Keywords: Sol–gel; Epitaxial growth; ZrO_2 thin films

1. Introduction

Particular interest in zirconia (ZrO_2) thin films for thin film devices arises from the fact that an epitaxial thin film of ZrO_2 can serve as epitaxy buffer layers between Si wafers and various functional thin films with appropriate lattice parameters [1–4]. ZrO_2 possesses similar lattice parameters ($a\sim5.12$ Å, $c\sim5.3$ Å) to Si (a$\sim$5.4 Å). It has been reportedly found that many electronic, optic and magnetic thin films with a perovskite-like structure (lattice parameters ranging 3.8$\sim$4.2 Å) can grow epitaxially on ZrO_2 buffered Si(100), either in cube-to-cube or 45° rotation. In this paper, we report the sol–gel fabrication of high quality epitaxy thin films of ZrO_2 on Si(100) wafers.

2. Experimental

Stock solution of ZrO_2 was prepared from 0.4 M of zirconia propoxide ($Zr(OCH_2CH_2CH_3)_4$) dissolved in *n*-propanol:Cl_3COOH:H_2O = 2:2:1. The solution was then aged at room temperature for 24 h. Thin films were coated using spin caster. The 1×1 cm coupons of Si(100) wafer were cleaned and etched using a conventional RCA process and used as substrates. The coated films were pyrolized on a hot plate pre-heated at 400°C, and the coating was repeated (typically two times) to attain a proper thickness ($\sim$1000 Å). Finally, thin films were annealed at 800–1000°C in air for 3 h. The four circle X-ray diffractometer was employed for structural characterization. The symmetric scan (2θ-θ) combined with rocking scan (ω) and ϕ scan were used to examine out-of-plane and in-plane orientation, respectively. Microstructure of the films was examined by tapping mode atomic force microscopy (TM-AFM).

3. Results and discussion

Fig. 1(a) shows XRD symmetric scan of the thin films annealed at 800°C in air for 3 h. Only the (001) peak of t-ZrO_2 was observed. It was reported that undoped zirconia sustain tetragonal structure if the grain size is fairly small (< 500 Å), since large free energy for transformation from tetragonal to monoclinic structure is required to overcome huge surface energy [5]. This is probably the case for our thin films, as will be verified by AFM characterization.

Fig. 1(b) displays the rocking scan (ω) of the same films. The full-width-at-half-maximum (FWHM) value was estimated to be $\sim$0.3°, thus the films were found to have good out-of-plane orientation. Fig. 2(c) shows

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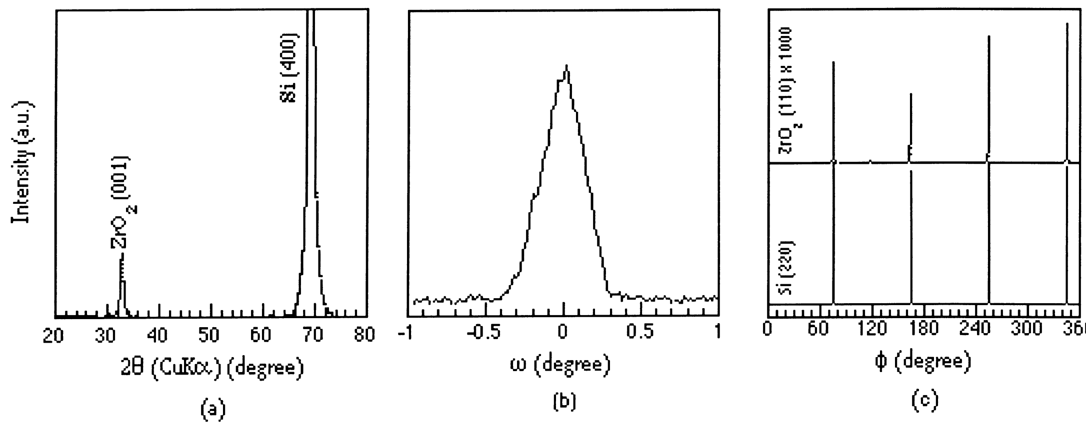

Fig. 1. Four-circle X-ray diffraction pattern of ZrO_2 thin film annealed at 800°C in air for 3 h: (a) symmetric (b) rocking and (c) ϕ scan.

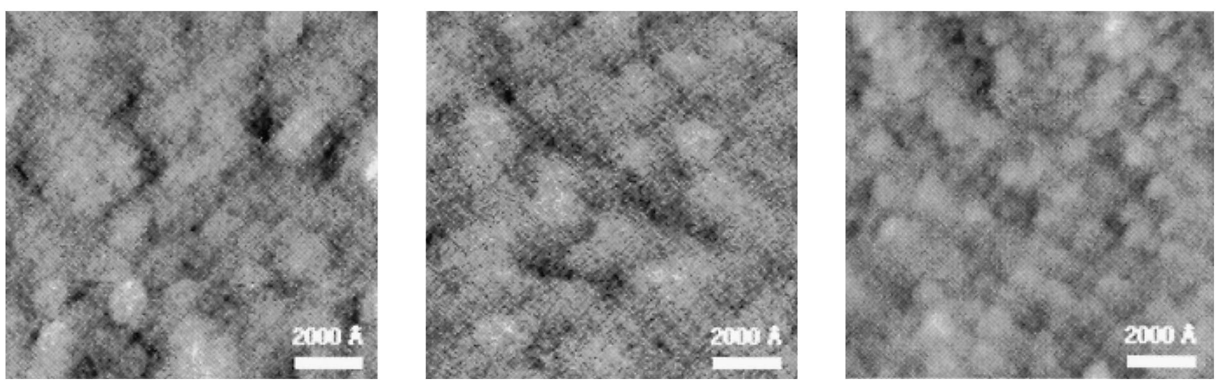

Fig. 2. Atomic force micrographs of ZrO_2 thin films annealed at (a) 800°C (b) 900°C and (c) 1000°C.

XRD ϕ scan of the (110) peak of the film (corresponding axial configurations are $2\theta = 22.3°$, $\chi = 45°$ and $\omega = 11.15°$). The observed peaks were at every 90°, which is consistent with the 4-fold symmetry of out-of-plane orientation [001]. The same scan on the (220) peak of the Si wafer is shown at the bottom of the plot. The (220) peaks of Si are located at the exactly the same position to those of ZrO_2 (110). From these, ZrO_2 thin films were confirmed to grow on Si(100) in cube-to-cube manner (i.e. ZrO_2[100](001)/Si[100](100)). The same epitaxial relation was found in all films annealed at 800–1000°C.

Fig. 2 shows the atomic force micrographs of the ZrO_2 thin films which were annealed at (a) 800, (b) 900 and (c) 1000°C. The films annealed at 800°C exhibited very excellent microstructure with uniform and spherical grains of 500 Å in size and rms roughness of only 14 Å. The increasing annealing temperature slightly increased rms roughness up to 18 Å, while the grain sizes were almost invariant, as shown in Fig. 2.

4. Conclusion

We could fabricate ZrO_2 epitaxial thin films on Si(100) via a sol–gel processing. The thin films exhibited high epitaxiality (the FWHM values in the rocking scan were ~0.3°) and excellent surface morphology (rms roughness was 14–18 Å), which makes the film highly versatile as epitaxial buffer layers on Si(100) wafers for various functional thin films.

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