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IN-SITU X-RAY REFLECTIVITY STUDY ON GROWN DYNAMICS
OF SPUTTER DEPOSITED GOLD ON SILICON*

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IN-SITU X-RAY REFLECTIVITY STUDY ON GROWTH DYNAMICS OF SPUTTER DEPOSITED GOLD ON SILICON

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ABSTRACT

We performed in-situ x-ray reflectivity measurements to study the growth dynamics of gold sputter deposited onto silicon, using an x-ray scattering chamber equipped with a faced magnetron source where the substrate is held at a right angle relative to the sputtering guns. By operating the guns at low power (1 watt) and under 20 mTorr Ar, we could control the gold deposition rate to less than $1\text{\AA}/\text{sec}$. The observed $\bar{x}$ -ray reflectivity for gold deposited onto a silicon substrate held at room temperature may be consistent with nucleated island growth for average gold particle sizes less than 50 $\text{\AA}$. Above 50 $\text{\AA}$, the reflectivity data indicates that the gold film uniformly covers the silicon surface, and that as the film thickness is increased the gold-vacuum interface gets progressively rougher. Detailed analysis of room temperature data is in progress, as is the temperature dependence on the roughness of the gold vacuum interface.

INTRODUCTION

The growth dynamics of thin solid films is sensitively dependent upon the deposition method and conditions. Thin solid film formation by particle deposition under nonequilibrium conditions has been an active area of theoretical and computer simulation research for over ten years^[1]. In spite of this, there has been only a limited number of experimental works done. The synchrotron x-ray reflectivity technique is a sensitive structural probe on microscopic length scales. Here we report preliminary results of the dependence of growth conditions on sputter deposited gold films on silicon studied by *in-situ* x-ray reflectivity.

EXPERIMENTAL

Synchrotron x-ray reflectivity measurements were performed on beamline X6B, at the National Synchrotron Light Source, Brookhaven National Laboratory. The wavelength of the incident x-rays was 1.55 $\text{\AA}$. Two pairs of tantalum slits were used to define the unfocused incoming beam to 2mm by 2mm. An x-ray compatible sputtering chamber, whose details are presented elsewhere^[2], was used with a faced magnetron sputtering system. The chamber was mounted on a Huber four circle diffractometer and had a base pressure of 5×10^{-5} torr. Research grade argon gas was used for sputtering, and an Ortec high voltage power supply, tunable from

0-3000 volts at 0-20 mA, powered the sputtering guns. The deposition rate was 0.2-1 Å/sec for data presented here. The reflectivity scans were recorded after sputtering for a fixed time. Silicon samples were ultrasonically cleaned for five minutes each in acetone, methyl alcohol, and deionized water. They were then baked in vacuum to 475 K in order to remove surface contaminants such as hydrocarbons and water.

RESULTS

The x-ray reflectivity geometry is shown in figure 1 for x-rays incident upon a film having index of refraction n_1 , which was grown on a semi-infinite substrate with index of refraction n_2 . The reflection amplitude for an ideally sharp interface can be written down as [3]

$$A = \frac{r_{01} + r_{12}e^{-iQD}}{1 + r_{01}r_{12}e^{-iQD}}$$

where r_{ij} is the Fresnel reflection amplitude of classical optics, and L the layer thickness. The deviation of the data from this equation is a sensitive measure of interfacial roughness.

Figure 2(a) and (b) show the reflectivity data as a function of momentum transfer recorded at 300 K, 10 mTorr Ar gas pressure and 400 K, 10 mTorr Ar gas pressure, respectively. In figure 2(a), the top most reflectivity was recorded after sputtering for two minutes. Below the critical momentum transfer ($Q_c = (4\pi/\lambda)\sin\theta_c$) incident x-rays experience total external reflection. The values (Q_c) for gold and silicon are 0.080 and 0.032 Å⁻¹ respectively. The two minute data clearly shows the silicon critical angle, which may be consistent with gold islands partially covering the silicon up to gold particle sizes of 50 Å. The oscillations in the data are due to interference between the silicon-gold and gold-vacuum interfaces and the frequency of oscillation is proportional to film thickness. The roughness of the gold-vacuum interface is manifested by a decrease in the reflected intensity above Q_c . The data suggests that the gold surface roughness increases with increasing deposition time.

In the early growth stages, up to approximately 100 Å, the 400 K data is very similar to the 300 K data, but the data exhibit marked differences thereafter. For example, the oscillation frequency of the 300 K data uniformly increases up to 60 minutes of deposition, indicating that a film with a relatively smooth surface is growing. However, the 400 K film oscillation frequency does not uniformly increase after 14 minutes of deposition. Speculatively, this is suggestive of a wetting transition, where the 300 K gold film is growing uniformly thick, but the 400 K gold may be growing in bulk-like droplets.

Figure 3 shows powder diffraction scans of four gold/silicon samples measured using copper $K\alpha_1$ radiation from an in-house rotating anode source. The films are approximately 3000 Å thick, and were sputtered, from top to bottom, at 1 mTorr Ar pressure, 300 K, 1 mTorr, 220 K, 10 mTorr, 300K, and 10 mTorr, 220 K. From figure 3, and corresponding rocking curve scans, it was determined that the films made at 1 mTorr are two dimensional(2D) powders made up of randomly oriented crystallites with (111) faces parallel to the substrate surface. The films made at 10 mTorr are also 2D powders, but the crystallites have a distribution of(111) and (200) faces oriented parallel to the substrate surface. Data recorded at 400 K, 10 mTorr is consistent with the low temperature 10 mTorr results.

SUMMARY

We have exhibited the practicality of performing in-situ x-ray reflectivity studies for sputter deposition. Preliminary results indicate that film crystallite orientation, and growth mode are sensitive to deposition conditions. Significantly, the surface roughness of a growing film increases with time or, equivalently, with film thickness. This result is independent of the variation of deposition conditions studied here. An investigation of the dynamic scaling behavior of growing thin solid films under a variety of sputter deposition conditions is in progress.

ACKNOWLEDGEMENTS

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Figure 1: In-situ specular x-ray reflectivity geometry for x-rays incident upon a sputter deposited film with complex index of refraction n_1 .

Figure 2: Reflectivity versus Q for gold sputter deposited on silicon. (a) The substrate temperature was 300K and the deposition times from top to bottom were 2, 4, 6, 8, 10, 12, 20, 24, 28, 36, 44, 60, 90, and 120 minutes. (b) Substrate temperature was 400K and the deposition times were, from top to bottom, 1, 2, 4, 6, 8, 10, 12, 14, 16, 18, 28, 40, 70, and 130 minutes.

Figure 3: Powder diffraction of gold films sputtered deposited on silicon to a thickness of approximately 3000 Å. The deposition conditions were; (a) 220K and 1 mTorr Ar pressure, (b) 300 K and 1mTorr Ar pressure, (c) 230 K and 10 mTorr Ar pressure, and (d) 300 K and 10 mTorr Ar pressure.

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