*Superficies y Vacío **9**, 280-282, Diciembre 1999*

Thickness effects on aluminum thin films

P. Quintana, A. I. Oliva, O. Ceh and J. E. Corona
Centro de Investigacion y de Estudios Avanzados del IPN-Unidad Merida, Departamento de Fisica Aplicada, A.P. 73 Cordemex, 97310, Merida,Yucatan, Mexico

M. Aguilar
Instituto de Ciencia de Materiales (CSIC), Cantoblanco, 28049 Madrid, Spain.

Aluminium thin films grown with different thickness on silicon substrates were analyzed by atomic force microscopy and grazing incidence x-ray techniques. Stress on surface film was found to be greater as the thickness increases because of the substrate history. Roughness measurements made on the AFM images shown a linear increase with thickness and an asintotic behavior with time after growth. As the film thickness increases, the (111) peak intensity increases relative to the (200) and (220) peaks.

1. Introduction

Aluminum thin films and Al alloys are widely used for electronic device applications [1-3] and comprise the majority of the interconnections used in the semiconductor chips. The physical properties of the Al thin films depend strongly on their microstructure, which can be characterized using different techniques [4]. The smoothness of the surface morphology of thin films is rather difficult to study since the microelectronic devices are becoming smaller and thinner on a submicron scale.

Atomic force microscopy (AFM) has been applied for the characterization of thin film devices. Primarily as a high-resolution profilometer for measuring edge angles, step-heights, surface microstructure, roughness, and in measuring lateral variations in dopant profiles. It is a suitable technique to characterize the surface texture of aluminum thin film in this range scale, which has an isolated character of its surface oxide; nevertheless AFM does not gives information about the surface structure.

By using x-ray diffraction techniques we can study the microstructural parameters, such as the degree of crystallinity, crystallographic orientation, crystallite size, phase identification, etc., important characteristics for the design, development and failure analysis of devices containing thin films. Specifically, x-ray grazing incidence is a technique suited for the study of thin surface layers. The advantage of this technique is that the penetration depth of the probe beam can be very nearly kept constant for all diffraction peaks studied, and can be kept at a defined low value by setting the grazing angle of incidence appropriately. This small depth of penetration and the availability of a wide array of diffraction peaks are particularly useful for studying very thin films.

Free evaporation is one of the easiest techniques for Al deposition; however because of the poor control of the atoms on the substrate is very common to obtain stressed thin films after deposition [5]. The measurement of the stress level of the film, can give the key to know more about the precision on the composition measurements and the degree of relaxation [6]. High quality silicon wafers are known to be flat for substrate applications and are normally used for Al deposition for electronic applications.

In this work we studied the effects of the thickness of Al thin films, as well as the stress effects when they were prepared by evaporation, and characterized by atomic force microscopy. Structure properties measured by grazing incidence x-ray diffraction are presented as a function of the film thickness.

2. Experimental

Aluminum thin films were prepared by evaporation in a conventional vacuum evaporation system. The chamber pressure during evaporation was about 10^{-6} torr. The material used for film preparation was high-purity (99.99 %) Al rod (from Balzers). The films were grown on silicon (100) substrates without previous treatment. The substrate was maintained at room temperature (RT) during sample preparation.

During film growth the distance substrate-source was 120 mm. Films were rectangular (2x10 mm) shape with mean thickness ranged from 0.06 µm to 3 µm. The average deposition rate was about 2 nm/s. The interconnects of the very large scaling integration (VLSI) devices have typical thickness of about 1 µm and widths that are measured in microns. Our samples have widths in the millimeter range because the necessity to have large areas for x-ray studies.

After preparation the films were kept into the chamber at vacuum for about 2 hours. This was made to be sure that the film temperature be uniform so that no thermal stress will appear when taken out from the chamber.

Thin films were not annealed to eliminate the intrinsic stress because the intrinsic elastic stresses in the film are homogeneous. Therefore, they should not lead to gradients that would be relaxed by the material transport.

Topographic images were obtained with a Park Scientific AFM/STM, model Autoprobe CP, using a high-resolution scanner in the force constant mode. The scan speed was 1 Hz and the image resolution 256 x 256 pixels. Measurements were made at room temperature (RT) and atmospheric pressure. We used commercial micro-levers from Park Scientific, coated with gold. The thermal response measured from these cantilevers is about 0.33 ms when they are coated with Al [7]. Giving that Al has a similar ρC_p property than gold for the same thickness, then, similar thermal response can be expected for aluminum. An important key, is to compare AFM images obtained with similar conditions in the gain of the feedback loop, the applied force and with the same cantilever. Different images obtained with different conditions can not be compared between them because of the different values obtained for the surface roughness.

The crystallinity of the films was studied with a Siemens D-5000 x-ray diffractometer. Grazing incidence geometry, and Cu monochromatic radiation, $\lambda = 1.5418$ Å was used. The maximum diffraction data were registered with a step size of 0.005° every 6 s with an aperture slit of 0.05 mm and with an incidence angle of 0.5°. Diffractograms were obtained with the sample at RT. Reflections were processed using Diffrac AT software.

3. Results

Al films with different thickness were grown on Si (100) substrate in order to study the films quality. AFM images were obtained from the substrate. Sometimes, substrate effects are not taken into account [8], but recently, some works have demonstrated that the statistical correlation between the roughness of different interfaces, plays an important role to describe the film roughness growth. In our case, initial substrate roughness was 1.1 nm for the showed image. Thus, the substrate characteristics can affect the first stage growth of the films, according to the nature of the mismatch between substrate and the first atoms impinging.

After each film was grown, we obtained AFM images and compared between them. Figure 1 shows the corresponding AFM images for the smallest thickness (0.06 µm) and for the largest thickness (3 µm). As it can be seen, surface characteristics are very different due to the contrasted differences in the growing conditions.

From Figure 1, we can deduce that growing conditions play an important role during the evolution of the structure. For small thickness, AFM images presents small nucleation sites because of the short time of growth.

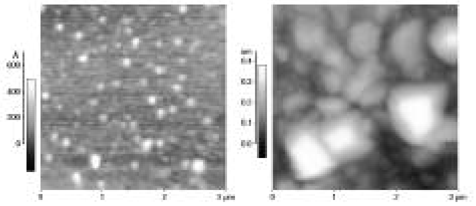

Figure 1. AFM images (3x3 µm^2) of Al film deposited on Si with 0.06 µm (left), and 3 µm (right) thickness.

However, as the thickness increases, grain size increases and film morphology seems to be uniform with large grains. Similar results were obtained on the surface morphology for a ratio between substrate temperature (T_s) and the melting point (T_m) of the film material larger than 0.3 [9]. From the fundamental structure forming phenomena, film growth evolves from nucleation islands to the growth of the continuous film, leading to large grains formed by the coalescence phenomena between grains. However, electronic applications will strongly depend on the surface roughness obtained and the structural properties.

The structural properties were measured by grazing incidence x-ray diffraction. Figure 2 shows the (220) maximum reflection for the thinnest and the thickest Al film for as grown condition. Al films have three mainly peaks, however a reflection with higher 2θ value has been used for a better appreciation. It can be deduced that the thinnest Al film is more stressed than the thickest one, since the main peak appears far from the line which correspond to the (220) standard peak. As the film thickness increases the surface stress diminishes.

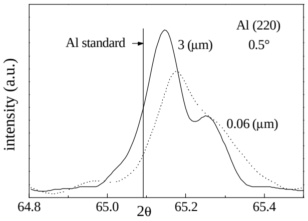

Figure 2. Diffractogram of the thinnest and the thickest Al film for the (220) peak studied in this work. Film stress is related by the shift-phase between both peaks and Al standard.

However, structural properties also change, as can be seen from the difference on the second peak reflection, which appears at higher 2θ values, compared between the two different thin films thickness. This peak is related with the mechanical stress between grains and the peaks position are related with the structure of the bulk. By the method used for film preparation, stresses always appear in the film surface. As the thickness increases, film “forgets” the substrate structure information and time to time, nucleus coalesces in crystals, crystals form grains, and grains fill the channels between them. The filled channels initiate the thicker growth and thus, the continuous film. Each growth stage will present different properties. It can also be observed that with higher thickness, the intensity of the peak increases, this effect favors the quality in the crystallinity on the material.

In order to find differences between them, we measured the three main diffraction peaks (111), (200) and (220) of the Al samples with different thickness. Figure 3 shows the relation between (111)/(200) and (111)/(220) peaks, considering the ratio of the peak area for the maximum diffraction, as a function of the film thickness; as the film thickness increases, the main (111) peak dominates both for the parallel direction as for the perpendicular direction.

However, the difference can be seen more clearly for the perpendicular direction. According to the JCPDS, the Al intensity ratio between the (111) and (200) reflections for a randomly oriented sample should be 2.12

[10]. If we take the ratio between the peak area for both relations, we found values between 1 and 10. From the results, all prepared films demonstrated to have the preferred (111) orientation, and this preference increases with the film thickness.

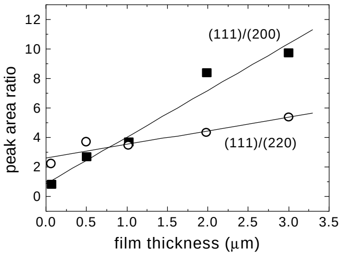

Figure 3. Structural changes observed by x-ray grazing incidence technique. Area ratio under the peak was plotted vs. film thickness for the (111)/(200) and (111)/(220) ratios.

Thin films with a large degree of (111) fiber texture have shown to exhibit larger times to failure [11, 12], due to the alignment parallel to the substrate surface of all grains, then the resulting grain boundaries are largely tilt boundaries. Moreover, tilt boundaries formed by steps and edges causes dislocations that are aligned normal to the film plane.

Film roughness was measured from the AFM images taken from samples immediately after preparation and also after eight months from preparation. During this time, films were stored in a closed and free humidity recipient. Figure 4 shows the results corresponding to these two periods. Film roughness as grown shows a near-linear increment with thickness. This is a typical behavior if we accept that diffusion is the dominant process during growth [13,14]

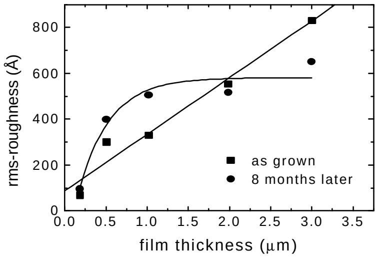

Figure 4. R_{rms} value calculated from the AFM images taken after preparation and eight months later. Different roughness is observed for both imaging conditions.

After eight months, films were measured but surface roughness found to have different behavior: as the thickness increases, the trend of the roughness is constant showing a smaller value compared with the as grown film. In this case, we want to highlight only the tendency of the roughness-curves obtained, due to the effect of the lapsed-time between both conditions; but it is not convenient to compare the absolute roughness values between them, since the obtained images were measured with different cantilevers. The roughness values presented in figure 4 are opposed apparently, because of the higher values measured eight months later.

4. Conclusions

We presented a study of Al thin films growth on silicon substrate prepared by free evaporation with different thickness. As their thickness increased, the surface morphology changes, appearing the different processes involved during film growth. Atomic force images of the films, were used for surface roughness calculation and their surface behavior changes depending on the lapsed-time after preparation. Roughness values decreases with time until reaching a constant value. However, when we measure the roughness as soon as the films are prepared, the roughness behavior shows a linear-increase with thickness. X-ray analysis by grazing incidence results, describe the preferred orientation as the thickness increases, and the surface stress after preparation. These results could be useful for thin films preparation for electronic applications.

Acknowledgments.

This work was supported by CONACyT (México) through project 28778-E.

References

[1]. J. R. Black, IEEE Trans. Electron. Dev. DE-**16,** 338 (1969).
[2]. M. J. Atardo, R. Rosemberg, J. Appl. Phys. **41,** 2381 (1970).
[3]. R. E. Hummel, R. T. DeHorff, H. Geiger, Phys. Chem. Solids **37,** 73 (1976).
[4]. Anon. MRS Bull. **17** (12) (1992); MRS Bull. **18,** (1) (1993).
[5]. W.R. LaFontaine, C.A. Paszkiet, M.A. Korhonen, Che-Yu Li, J. Mat. Res. **6,** 2084 (1991).
[6]. P. F. Fewster, N. L. Andrew, Thin Solid Films, **319,** 1 (1998).
[7]. J. Lai, T. Perazzo, Z. Shi, A. Majumdar, Sensors and Actuators A, **58,** 113 (1997).
[8]. K. K Nanda, S. N. Sarangi, S. N. Sahu, Appl. Surf. Sci. **133,** 293 (1998).
[9]. P. B. Barna, M. Adamik, Thin Solid Films, **317,** 27 (1998).
[10]. M. Avinum, N. Barel, W.D. Kaplan, M. Eizenberg, M. Naik, T. Guo, L.Y. Chen, R. Mosely, K. Littau, S. Zhou, L. Chen, Thin Solid Films, **320,** 67 (1998).
[11]. R. Rosemberg, J. Vac. Sci. Technol. **9,** 263 (1972).
[12]. R. E. Hummel, D. W. Malone, in Materials Reliability Issues in Microelectronics, Eds. J.R. Lloyd, F. G. Yost, and P.S. Ho, Mat. Res. Soc. Proc. **225**, Pittsburg, PA 1991, p.67.
[13]. L. Vazquez, R.C. Salvarezza, A.J. Arvia, Phys. Rev. Lett. **79,** 709 (1997).
[14]. A.I. Oliva, J.L. Sacedón, E. Anguiano, M. Aguilar, J.A. Aznarez, J.A. Mendez, Surf. Sci. Lett. **417,** L1139 (1998).