J. Phys. D: Appl. Phys. **31** (1998) L73–L77. Printed in the UK PII: S0022-3727(98)95515-2

RAPID COMMUNICATION

Morphology of thin silver film grown by dc sputtering on Si(001)

**S Kundu†, S Hazra†, S Banerjee†, M K Sanyal†, S K Mandal‡,
S Chaudhuri‡ and A K Pal‡**

† Surface Physics Division, Saha Institute of Nuclear Physics,
1/AF Bidhannagar, Calcutta 700 064, India
‡ Department of Materials Science, Indian Association for the Cultivation of
Science, Jadavpur, Calcutta 700 032, India

Received 2 July 1998, in final form 5 August 1998

Abstract. The morphology and growth mechanism of silver films approximately 150 Å in thickness on Si(001) substrates have been studied by atomic force microscopy and x-ray reflectivity. The thin films prepared by dc sputtering at room temperature are composed of islands of silver. The shape and size distribution of these islands are studied using these two complementary measurement techniques.

In recent years studies on the structures and properties of single and multilayer thin films with predetermined compositions are gaining importance as these systems exhibit some unique physical properties having potential applications in various electronic, magnetic and optical devices, such as the Schottky barrier [1], multilayered giant magnetoresistance materials [2], monochromators [3], etc. Surface/interface morphology play important roles in modifying the physical properties of the films. Study of thin films approximately 100 Å in thickness is important as this order of thickness is often used in the fabrication of the above mentioned materials. Understanding of the growth mechanism of a single thin film can provide an insight on how the growth of a multilayered system will proceed.

Extensive work has been done in understanding the growth mechanism of the Ag film on various substrates using different deposition processes [4]. Also, various probes have been used to study the growth of Ag film on silicon, such as, low energy electron diffraction, scanning tunnelling microscopy [5], transmission electron microscopy [6], Auger electron spectroscopy [7], x-ray reflectivity [8], etc. Different types of growth and morphology of Ag on Si(001) have been observed [4–9], for example, films having smooth surfaces and formation of 3D islands from an initially formed 2D layer depending on the thickness and deposition condition. In figure 1 we have shown schematically different types of possible morphology of Ag film as grown on the Si substrate and corresponding expected electron density profile (EDP, which is proportional to the mass density profile for a single material) for those systems.

In this work we have studied the growth of Ag thin films on Si single crystal deposited by a dc sputtering technique at room temperature. Atomic Force Microscopy (AFM) and x-ray reflectivity techniques are used to investigate the possible morphology of the Ag films. AFM has been used to see the top surface morphology of the film from which we observed the formation of islands. It gives us information regarding the average size and size distribution of the islands and also provides information regarding the shape of the island. On the other hand, x-reflectivity measurements [10–12] not only provide the EDP as a function of depth and the top surface roughness, but it also gives us unique information regarding the burried Ag-Si interface.

Ag films were grown on Si(001) substrates using a dc sputtering technique. Si substrates of electronics grade were cleaned in acetone and methanol in an ultrasonic bath. Then the substrates were subjected to vapour degreasing in isopropyl alcohol and were dried in a hot atmosphere of it. The sputtering chamber was evacuated to a base pressure of approximately 10^{-3} Pa. For sputtering, pure argon gas (99.99%) was introduced into the sputtering chamber through a needle valve at a constant pressure of 60 Pa. The Ag (99.999%) target, 25 mm in diameter was placed at a distance of 25 mm from the substrate. The substrate holder working as the anode was earthed and the cathode holding the sputter target was connected to the negative terminal and the voltage applied between them was 1 kV. Before the target sputtering starts the target surface was exposed to the plasma created between the target and the shutter to clean the surface of the target. Ag films were grown at room temperature for different durations of deposition. Two films designated by A and B, which were grown under identical conditions by sputtering for 30 s and 40 s, respectively are presented here.

X-ray scattering measurements were performed using a 18 kW rotating anode (FR 591, Enraf Nonius) x-ray source with Cu $K_{\alpha 1}$ line monochromatized by a Si(111) crystal. A

0022-3727/98/230073+05$19.50

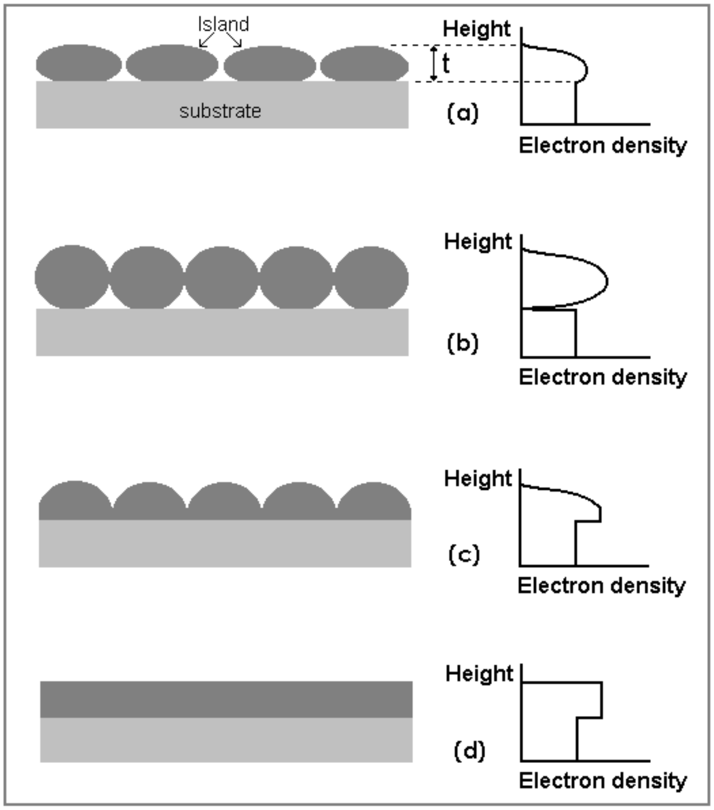

Figure 1. Schematic diagram of the possible morphology (side view) of the thin film of thickness, *t* and corresponding shape of EDP. (a) Ellipsoidal shaped loosely connected dewetted islands; (b) spherical shaped connected dewetted islands; (c) spherical shaped connected wetted islands; (d) wetted smooth film.

triple axes spectrometer (Optix microcontrole) was used to measure both specular and off-specular reflectivity. AFM studies were performed by scanning probe microscope (Autoprobe CP, Park Scientific) in ambient conditions. The scan was taken by 100 μ scanner using silicon nitride tip (radius of curvature $\sim$ 200 Å) in a constant force ($\sim$ 2 nN) contact mode. Initially the film quality was checked by performing large area scans. No inhomogeneity in the film was observed.

AFM images in different length scales have been taken to see the top surface morphology of the deposited Ag films. Four such AFM images of a small area scan are shown in figure 2. The AFM images show that both the films are composed of small islands as observed earlier by others [4]. The average sizes of these islands in the horizontal plane are $\sim$ 340 Å and $\sim$ 350 Å for films A and B respectively. In figure 3 we show the height distribution and bearing ratio (which is essentially an integral of the height histogram from the top surface i.e., a plot of the precentage of data points at or above a given height) of two AFM images of scan size 0.5 μ × 0.5 μ. We observed the height distribution to be symmetric. The rms roughness have been calculated from the height distribution. The average rms roughness obtained from several scans are 10 Å and 13 Å for films A and B, respectively. It can be noted that the above roughness obtained from the AFM image is the convolution of the shape and local roughness of the island. The local roughness ($\sim$ 3 Å) can be obtained from the scan over an individual island. The value of local roughness is very small compared to the value of the above mentioned average rms roughness, indicating that the average rms roughness is dominated by the shape of the islands. The bearing ratio, on the other hand, goes to zero beyond 60 Å and 90 Å for films A and B, respectively indicating that the islands are connected to each other (at least for the resolution dictated by the scan size and the aspect ratio of the tip) at this depth from the surface. This is true as the scan size was much larger than the size of the island. One can also obtain the average shape of these islands as a function of depth from the bearing ratio distribution.

In figure 4 x-ray specular reflectivity as a function of $q_z (= 4\pi \sin\theta/\lambda)$ are shown for both the films. The hump (or oscillation) around $q_z = 0.1$ Å^{-1} observed in the reflectivity curves is due to the total film thickness of Ag. The rapid fall of reflectivity after q_c (critical wave vector, indicated by an arrow in the figure 5) and the damping of the oscillation indicates high roughness at surface and interface of the film [11, 12]. The rounding of the x-ray reflectivity intensity near q_c is due to high x-ray absorption coefficient of Ag. We have also measured the off specular reflectivity and found it to be negligible (of the order of 10^{-3} times that of the specular reflectivity).

Parratt formalism [10] have been used to obtain the EDP as a function of depth from the diffuse subtracted reflectivity data. To fit the reflectivity data, the EDP of the film was divided into ten equal slices. The fit parameters

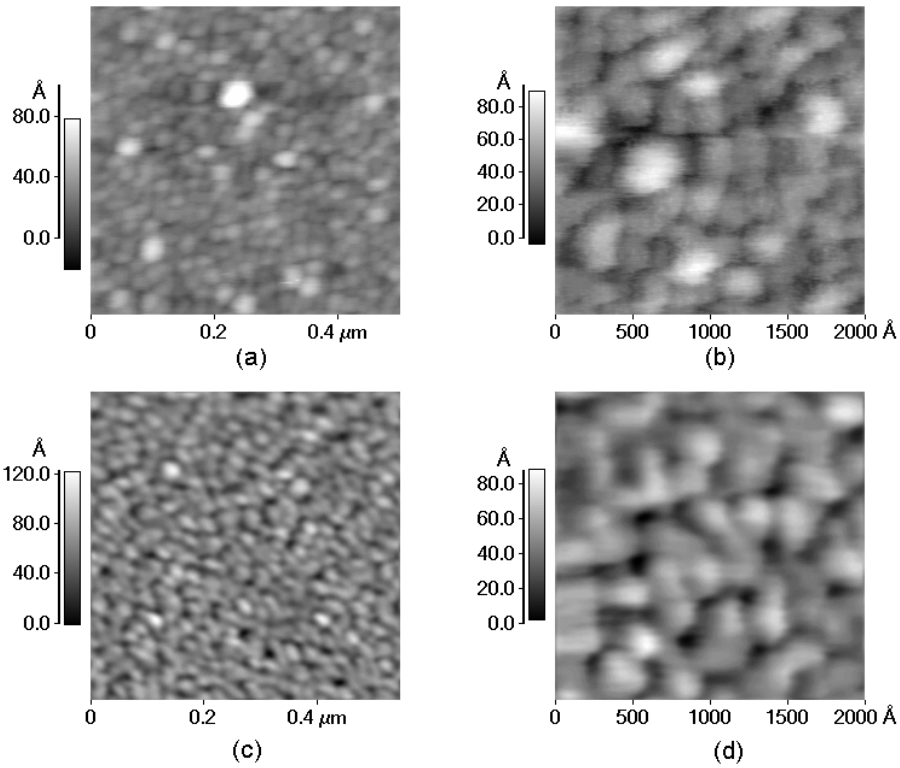

Figure 2. AFM images in two different scan sizes: (a) and (b) for film A and (c) and (d) for film B.

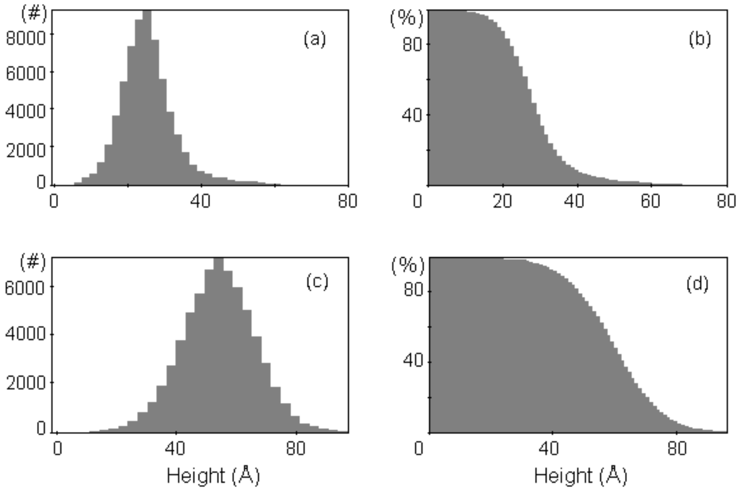

Figure 3. (a) and (c) height distribution [in number (#)] and (b) and (d) bearing ratio [in percent (%)] for two of the AFM images [(a) and (c)] of figure 2. The height zero in this case is define as the maximum depth that AFM tip can probe.

are the thickness of the film, electron density of each slice and roughness at the top surface and the film–substrate interface. The roughness at the surface and interface were incorporated assuming a roughness of the form of error function [11]. The roughness convoluted EDP obtained from the least square fitting of the reflectivity data are shown in the inset of figure 4. The total thickness (t, similar to that indicated in figure 1) for the films A and B are found to be (134 ± 2) Å and (158 ± 2) Å respectively. Both EDP show a maxima near the centre of the film and it decreases gradually to zero at the surface and to the value of the electron density of Si substrate beyond 134 Å and 158 Å for films A and B respectively. The maximum electron density observed for Ag are 60% and 70% that of the calculated bulk value (2.744 Å^{-3}) for films A and B respectively. Such reduction of density in nanocrystalline Ag was also observed earlier [13], which was attributed to the presence of voids at the grain boundaries as observed from positron annihilation measurements. Similarly, the low value of electron density obtained here can also be

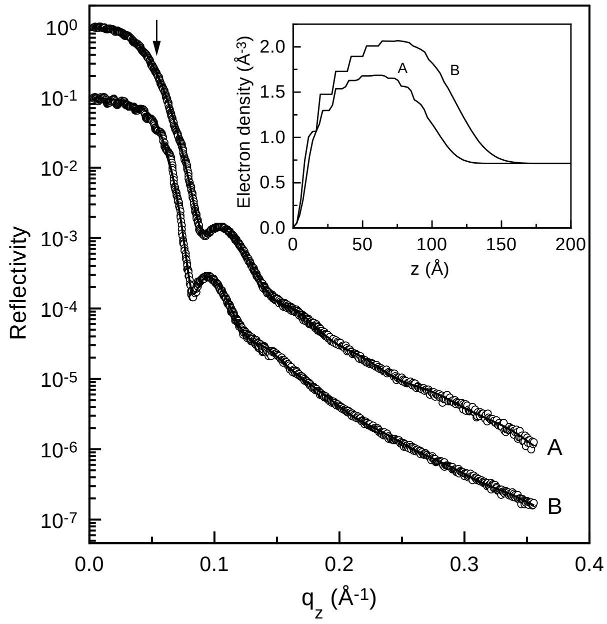

Figure 4. X-ray reflectivity for two films A and B; data for film B has been shifted down for clarity. Inset shows corresponding roughness convoluted EDP. z=0 in this case is define as the top surface of the film.

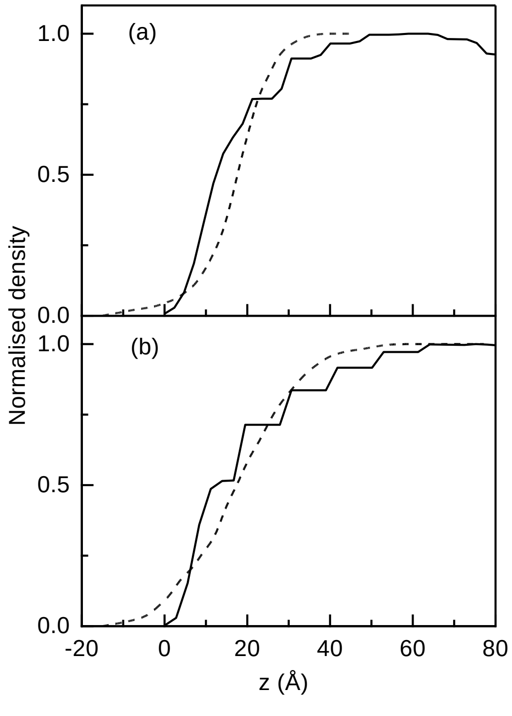

Figure 5. Normalised EDP (solid line) and modified bearing ratio (dashed line) shown for comparison. (a) for film A and (b) for film B.

attributed to the presence of the voids in the islands along with the low connectivity of the islands. As the islands grow with the deposition time the connectivity in xy plane increases and hence the electron density increases as we see in the case of film B.

In figure 5, normalized electron density and bearing ratio with the same convention of height as for electron density are plotted for both the films. For comparison we have shifted the bearing ratio to overlay on EDP and we observe both the profiles increase in the similar fashion. The value of bearing ratio at negative value of z is an artifact of the shifting, which indicates that the bearing ratio does not exactly follow the EDP. The difference between these two can be attributed to the tip artifact. However, here our main interest is to show that both the profiles are similar (although not exact) and such bearing ratio profile is arising from a particular shape and size of the islands as observed directly from the AFM images. Similarly, the same type of variation of electron density (in figure 4) on the other (film–substrate) side of the films can infer about the shape of the islands towards the burried interface, which can not be observed directly by AFM. The EDPs in figure 4 for the two Ag films are similar to that shown in figure 1(a), hence indicates formation of ellipsoidal-like islands of Ag. The size of the island in xy plane is mentioned above and the height of the islands formed is the thickness of the film. The shape of the island is however, dictated by the wetting properties and film-substrate interaction. It can be noted that the actual shape of the island can be obtained only after proper modeling taking into account of the various effects, such as the shape of the AFM tip, substrate roughness, etc., along with the transverse diffuse scattering data, which are in progress.

The growth of Ag thin film on Si(001) substrate deposited by dc sputtering technique have been studied by AFM and x-ray reflectivity measurements. Island formation of Ag was observed for film of thickness $\sim$ 150 Å from AFM images, while the average film thickness and EDP were obtained from the reflectivity measurements. The formation of ellipsoidal shaped islands deduced from these two complementary measurements have been attributed to the dewetting property of the Ag on Si substrate.

References

[1] Xu P, Miller P and Silcox J 1991 *Mater. Res. Soc. Symp. Proc.* **202** 19

[2] Gijs M A M and Bauer G E W 1997 *Adv. Phys.* **46** 285
Baibich M N, Broto J M, Fert A, Van Dau N, Petroff F, Etienne P, Creuzet G, Friederich A and Chazelas J 1988 *Phys. Rev. Lett.* **61** 2472

[3] Stearns M B, Chang C-H and Stearns D G 1992 *J. Appl. Phys.* **71** 187

[4] Doraiswamy N, Jayaram G and Marks L D 1995 *Phys. Rev.* B **51** 10167
Samsaver A, Hirschorn E S, Leibsle F M and Chiang T-C 1989 *Phys. Rev. Lett.* **63** 2830
Hanbucken M, Futamoto M and Venables J A 1984 *Surf. Sci.* **147** 433

[5] Folsch S, Meyer G, Winau D, Rieder K H, Hoegen M H, Schmidt T and Henzler M 1995 *Phys. Rev.* B **52** 13745
Lin X F, Wan K J and Nogami J 1994 *Phys. Rev.* B **49** 7385 Palasantzas G and Krim J 1994 *Phys. Rev. Lett.* **73** 3564
Winau D, Itoh H, Schimid A K and Ichinokawa T 1994 *Surf. Sci.* **303** 139

[6] LeGoues F K, Liehr M, Renier M and Krakow W 1988 *Phil. Mag.* B **57** 179

[7] Luo F C H, Hembree G G and Venables J A 1991 *Evolution of Thin Film and Microstructure* eds C V Thompson, J Y Tsao and D J Srolovitz (Pittsburg: Mater. Res. Soc.)
Hanbucken M and Neddermeyer H 1982 *Surf. Sci.* **114** 563
[8] Je J H, Kang T S and Noh D Y 1997 *J. Appl. Phys.* **81** 6716
Thompson C, Palasantzas G, Feng Y P, Sinha S K and Krim J 1994 *Phys. Rev.* B **49** 4902
[9] Glueckstein J C, Evans M M R and Nogami J 1996 *Phys. Rev.* B **54** R 11066
[10] Parratt L G 1954 *Phys. Rev.* **95** 359
[11] Piecuch M and Nevot L 1990 *Mater. Sci. Forum* **59&60** 93
[12] Sanyal M K, Datta A, Banerjee S, Srivastava A K, Arora B M, Kanakraju S and Mohan S 1997 *J. Synchrotron Rad.* **4** 185
Banerjee S, Sanyal M K, Datta A, Kanakraju A and Mohan S 1996 *Phys. Rev.* B **54** 16377
[13] Kizuka T, Nakagami Y, Ohata T, Kanazawa I, Ichinose H, Urakami H and Ishida Y 1994 *Phil. Mag.* A **69** 551