J.Serb.Chem.Soc. *66*(7)483–490(2001)
JSCS–2877

UDC 543.251
Original scientific paper

Surface roughness minimum: Ag thin layer deposited on a glass

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(Received 9 March 2001)

In this paper the results of an examination of the surface roughness and morphology dependence of silver thin films up to 100 nm thick deposited on a microscope glass on the deposition rate and on the deposition time are presented. It was found that, for a constant deposition rate, the surface roughness exhibits minimum at a certain layer thickness. This coincides with the turning point when the influence of the substrate surface on the deposition process becomes negligible, *i.e.,* to the change in the nature of the system substrate/deposit from Ag/glass to Ag/Ag. For a constant layer thickness, the surface roughness minimum, achieved at a certain deposition rate, coincides to the turning point when the average free path for vertical adatom mobility becomes zero.

Keywords: surface roughness, silver, glass.

INTRODUCTION

The growth of thin films by vapor deposition is often not perfect. Deviations from perfect growth are exclusively determined by kinetic parameters. The stochastic nature of the impinging flux, and possibly the nucleation process itself, induce a roughness, which is generally counterbalanced by a smoothing or ordering process due to the mobility of the deposited atoms.[1–3] There are two preferred directions in the mobility of adatoms: one perpendicular to the surface and the other one along the surface.[4] These directions exhibit asymmetry in the vertical and horizontal mobility of adatoms.[5] The change of the number of species in vertical and horizontal directions is a consequence of various processes influencing the orderliness of the thin layer. In a vertical direction, the process contributing to the orderliness of the thin layer is the vertical mobility of adatoms (Schwoebel effects).[6] In the horizontal direction, the change of the number of species on the surface depends only on the horizontal adatom mobility, which contributes to the orderliness of the thin layer. Both, the vertical and the horizontal mobility of adatoms, depend on the nature of the system deposit–substrate, the state of the substrate surface (surface defect density,[7] surface strain,[8] temperature) and the layer thickness.[9] They also influence the nucleation processes.[7,10]

The aim of the present work was to examine the surface roughness and morphology dependence of silver thin films deposited on a microscope glass on the deposition rate and on the deposition time with respect to the changes of the vertical mobility of the deposited adatoms.

EXPERIMENTAL

Silver was sputter deposited onto a highly polished microscope glass substrate (Glasco Products, Chicago, USA) previously cleaned in an ultrasonic bath with organic solvents and ultra pure water. The deposition was carried out in a commercial rf diode sputtering vacuum system in an argon atmosphere for various deposition times. The residual pressure in the chamber was 3×10^{-7} mbar, while the partial pressure of argon was 1×10^{-3} mbar. The substrate temperature was below 30 ºC. The chosen deposition rates, R_1, were: 0.09, 1.3, 4.12, 7.3, and 11.0 ML/s. For each deposition rate, the Ag layers were deposited with the thickness: 9, 20, 32, 40, 59 and 81 nm.

Analysis of the surface morphology of the deposited Ag thin layer was performed at room temperature using STM in air. The microscope used was a commercial NanoScope III (Digital Instruments, Inc.). The images were obtained in the constant-current mode using a W tip. The set-point current, i_t ranged from 7.1 to 8.5 nA, and the bias voltages, V_b from 16.4 to 18.7 mV. All the images were obtained immediately after the deposition by scanning over an area of (880 × 880) nm^2. The surface roughness was estimated using the subprogram packages of the NanoScope III as the average value out of 15 STM images taken from different points within the substrate. The thickness of the deposited layers was measured after imaging using a profilmeter, with the error less than 10 % for low layer thicknesses and less than 5 % for higher layer thicknesses.

RESULTS

The morphology of Ag thin layers of various thickness deposited on a microscope glass using various deposition rates is presented by the STM images (880 × 880) nm^2 in Figs. 1 and 2.

The morphology of the deposited Ag layers of various thickness (9, 60 and 81 nm), obtained using the same deposition rate of 1.3 ML/s is presented in Fig. 1. The change in morphology with increasing layer thickness is obvious. For a small layer thickness, Fig. 1a, the deposit consists of randomly distributed grains about 200–250 nm large. The almost hexagonal shape of the deposited Ag grains indicates a crystalline structure of the (111) orientation. For a higher layer thickness, (Fig. 1b), a quite different deposit morphology is obtained. The deposited Ag grains are much smaller in size (10 – 20 nm). With further increase in the layer thickness the deposit morphology changes again, which is illustrated in Fig. 1c, for a deposited layer 80 nm thick. The substrate surface is covered with a deposit consisting of about 150 – 200 nm large grains distributed orderly along one direction. The deposited Ag grains are homogeneous in size and the grain chape is clearly hexagonal.

Such a change in the deposit morphology can be explained by a change in the nature of the deposit/substrate system. For a small layer thickness, the deposit morphology is influenced by the interaction of Ag adatoms with the atoms of the substrate. The mobile Ag adatoms move over the substrate surface and coalesce creating grains, (see Fig. 1a). With increasing coverage with deposited Ag, the interaction of the Ag adatoms with the atoms of the substrate decreases. At a certain layer thickness, the influence of

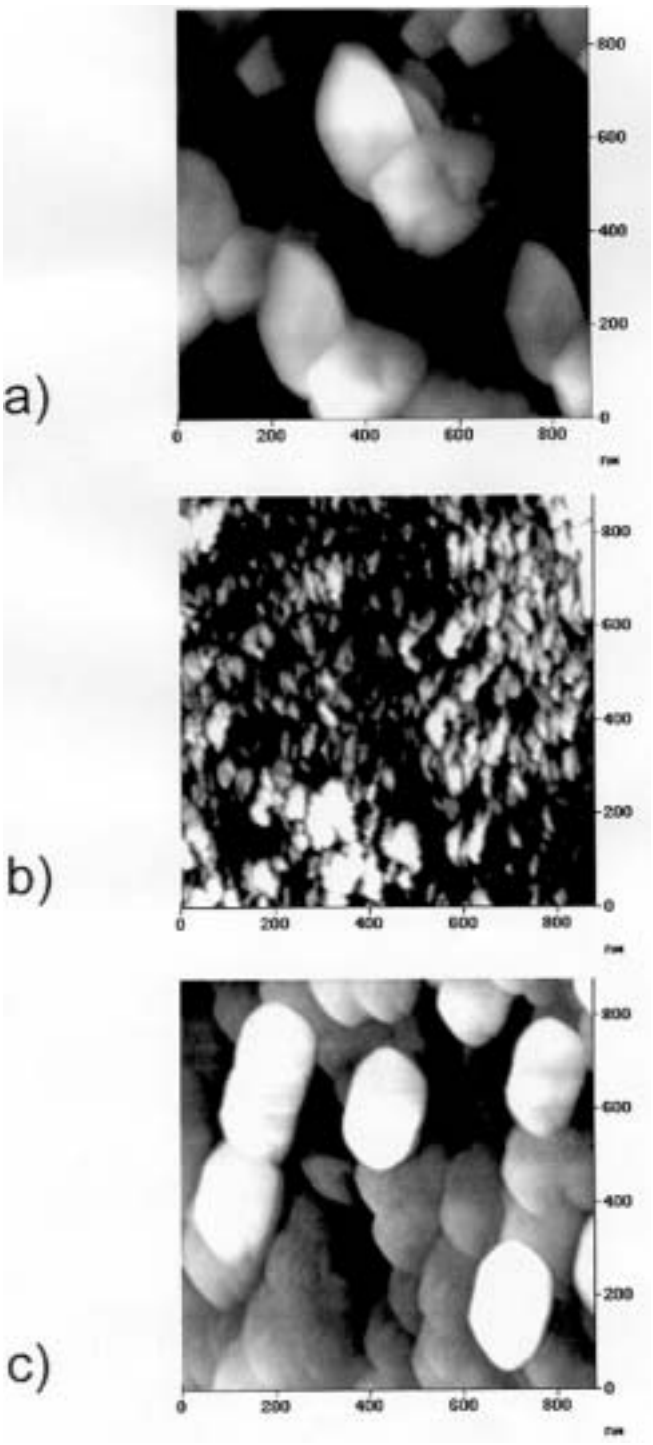

Fig. 1. STM images (800× 800) nm^2 of Ag deposited on a glass substrate at a deposition rate of R = 1.3 ML/s and layer thickness h: a) 9 nm; b) 60 nm; c) 81 nm. The white areas on the images correspond to the maximum height of 30 nm.

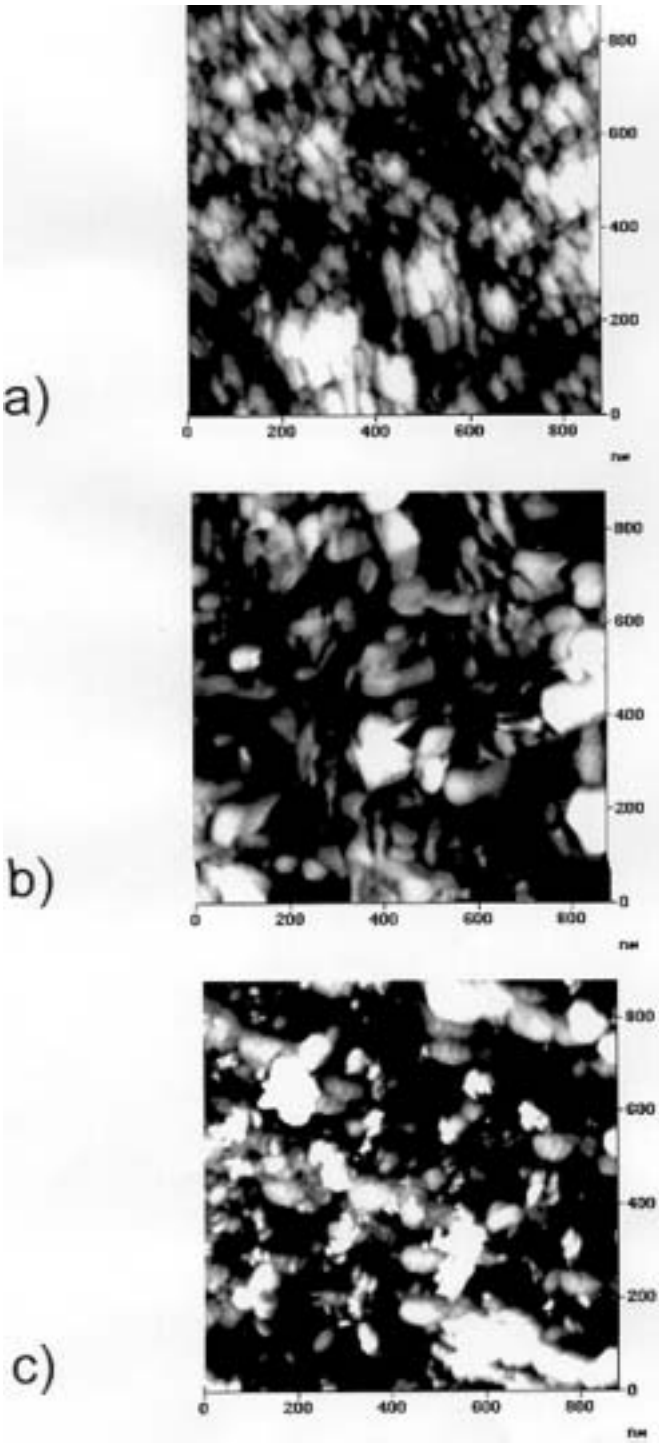

Fig. 2. STM images (800× 800) nm^2 of Ag deposited on a glass substrate to a layer thickness of $h = 32$ nm at the deposition rate, R of: a) 1.3 ML/s; b) 7.3 ML/s; c) 11.0 ML/s. The white areas on the images correspond to the maximum height of 30 nm.

the substrate on the new coming Ag adatoms becomes negligible and they behave as if they were arriving at a silver substrate, *i.e.*, the system deposit/substrate has changed from Ag/glass to Ag/Ag. The change in the mobility of Ag adatoms over the surface leads to a change in the surface morphology, (see Fig. 1b). With further increase in the layer thickness, the small Ag grains grow and coalesce which leads to the formation of larger grains and again to a change in the surface morphology, Fig. 1c.

The morphology of deposited Ag layers of the same thickness (32 nm) obtained at various deposition rates (1.3, 7.3 and 11 ML/s) is presented in Fig. 2. The change in morphology of the deposited Ag layer with increasing deposition rate is not reflected in a change of the lateral grain size, but to vertical inhomogeneity of the layer. For the low deposition rate (Fig. 2a), the distribution of the grains over the surface is homogeneous and the average grain size small (10 – 20 nm). It can be seen that the lowest deposition rate used corresponds to the deposition rate in Fig. 1, and the morphology of the deposited layer to the one shown in Fig. 1b. With increasing deposition rate (Fig. 2b), the morphology does not change significantly. The grains are a bit larger and higher indicating a similar lateral and lower vertical Ag adatoms mobility. At even higher deposition rates, Fig. 2c, the lateral dimensions of the grains are almost identical to the previous layers, but the existence of grains with a large vertical dimension can be noticed from the STM images (white areas). Such a structure corresponds to spongy layers. In this case, the change in the deposit morphology can be explained by the change in the average free path for Ag adatoms caused by the increase in the density new-arriving Ag adatoms with increasing deposition rate. The number of adatoms deposited per unit time and per unit surface area increases, leading to an increase of the frequency of Ag adatoms coalescence. As a consequence of the decreased average

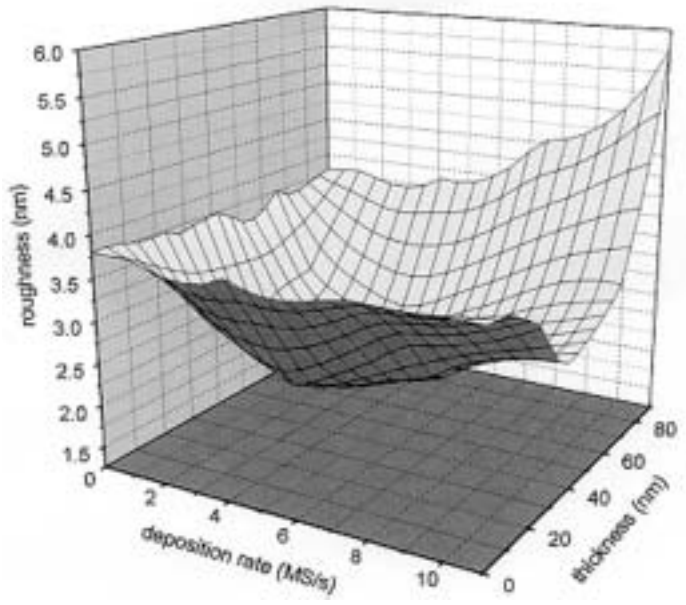

Fig. 3. The dependence of the average surface roughness on the deposition rate for various thicknesses of Ag layers deposited on a glass substrate.

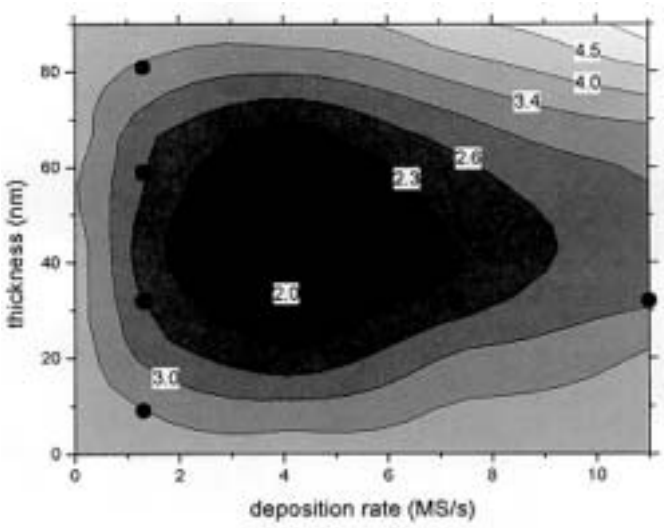

Fig. 4. The topographic dependence of the average surface roughness of Ag layers deposited on a glass substrtate as a function of the layer thickness and the deposition rate. The black points mark the places represented by STM images from Figs. 1 and 2.

free path on the substrate (glass) surface, the Ag adatoms coalesce one with another faster and the influence of the substrate surface is reduced.

The average surface roughness was calculated from the STM images for the deposits obtained using the deposition rates: 0.09, 1.3, 4.12, 7.3, and 11.0 ML/s and for layer thicknesses of: 9, 20, 32, 40, 59 and 81 nm for each deposition rate, as described in the Experimental. The dependence of the average surface roughness on the deposition rate and on the layer thickness is presented in the three coordinate system in Fig. 3. The complicated shape of the presented surface plot shows clearly the existence of a roughness minimum for particular values of the deposition rate and layer thickness. The horizontal projection of the surface roughness in the plane is given in Fig. 4. The curves on the plot correspond to the same values of surface roughness as given in Fig. 3. The area with the lowest roughness is shadowed by the darkest gray color, while the areas with higher surface roughness are shadowed by lighter tones of gray.

The surface roughness for low deposition rates (up to 0.1 ML/s) is not dependent on the layer thickness and has a constant value (in the interval of statistical error) equal to the substrate surface roughness. For higher deposition rates, the surface roughness exhibits a minimum with changing layer thickness. This minimum is most pronounced for deposition rates of 3–5 ML/s and is achieved at a layer thickness of about 45 nm. With further increase in the deposition rate, the value of the minimum decreases. Following this tendency, it can be concluded that for deposition rates higher than 11 ML/s, the surface roughness increases with the layer thickness.

The average surface roughness changes with the layer thickness in a similar manner. For a low layer thickness (up to 3.5 nm) it does not depend on the deposition

rate. With increasing layer thickness, the surface roughness exhibits a minimum with increasing deposition rate. The highest value of the minimum is reached at a layer thickness of 35 – 55 nm. This corresponds to the deposition of 150–300 monolayers of Ag on the glass. For layer thicknesses higher than 80 nm, the surface roughness of the layer increases with deposition rate. It can be noticed that the dependence of the average surface roughness and the surface morphology (with respect to the grain size) on the deposition rate are similar. This indicates to a direct relation between surface roughness and grain size for layer thicknesses up to 80 nm. This means that a better flattening of the surface is obtained with a larger number of smaller grains.

Thin layers with different surface roughness exhibit different electrical, optical and catalytic properties. According to the results presented above, it can be seen than Ag deposited on a glass substrate achieves the properties of a bulk Ag substrate at the layer thicknesses of about 40 nm. In order to obtain flat layers with good electrical and optical properties, the deposition should be performed at a deposition rate lower than 4 ML/s. On the other hand, rough Ag layers with a higher surface area should exhibit better catalytic properties and can be obtained using deposition rates higher than 4 ML/s.

И З В О Д

МИНИМУМ ПОВРШИНСКЕ ХРАПАВОСТИ: ТАНКИ СЛОЈ Ag ДЕПОНОВАН НА СТАКЛУ

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У овом раду су представљени резултати испитивања зависности површинске храпавости и морфологије танког слоја Ag, дебљине до 100 nm, депонованог на микроскопском стаклу, од брзине и времена депоновања. Нађено је да, за константну брзину депоновања, површинска храпавост показује минимум за извесну дебљину слоја. Минимум се поклапа са престанком утицаја подлоге на процес депоновања, односно са променом природе система депозид/подлога од Ag/стакло у Ag/Ag. За константну дебљину слоја, минимум површинске храпавости се постиже за извесну брзину депозиције, када средњи слободни пут за мобилност адатома постане једнак нули.

(Примљено 9. марта 2001)

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