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Regularities of Ultrathin Silver Films Formation on Cleaved KCl Facets

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The ultrathin silver layers formation regularities were studied. Silver films were obtained by DC magnetron sputtering onto KCl cleaved facets in an ultrapure inert environment. The technique of samples obtaining was based on reducing of the percolation threshold for silver condensates under Volmer-Weber nucleation conditions and exposing of a growth surface to plasma. The condensates structure was investigated by means of transmission electron microscopy. The experiments have shown that on the initial stage of film growth on cleaved KCl facets solid amorphous layer was formed. With increasing of layer thickness a transition to crystalline state was occurred.

Keywords: Silver, Nanostructures, Volmer-Weber conditions, Magnetron sputtering.

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1. INTRODUCTION

Formation of ultrathin solid metallic films with nanothickness is an important problem for many practical applications. Such films can be used in solar cells as transparent conductive layers as an alternative to ITO and TCO [1], layers with enhanced solar energy absorption [2], for creating metamaterials, various plasmonics elements and etc. [3].

As commonly known, growth of metallic films on dielectric substrates such as glass occurs sequentially through the following stages: nucleation of islands; their coalescence; formation of prolonged structures, when the coalescence becomes complicated; percolation; formation of channels and, finally, their closing up to form a solid layer. Such variant of films origination occurs in accordance to Volmer-Weber conditions [4,5]. That is why it is complicated to produce films, being both solid and thin as much as possible. Thus, it is important to find technological solution for lowering the thickness of a solid film. As a criterion of solid film formation, one can consider a percolation threshold or an equivalent film thickness, at which a film becomes conductive. For instance, the percolation threshold for gold and silver is typically close to and below 8-14 nm depending on deposition condition [6]. Besides, film properties, and in particular optical and electric properties, crucially differ from bulk material properties at thicknesses near and below the percolation threshold because of discontinuity of the whole structure.

The main aim of the given work is to reduce the film thickness corresponding to percolation for silver films while using Volmer-Weber nucleation conditions and exposing of a growth surface to plasma.

2. EXPERIMENTAL PROCEDURE

DC magnetron sputtering was used for obtaining Ag samples. An increased vapor pressure and relatively low discharge power were used to make quasi-equilibrium conditions. Cleaved KCl facets were used as substrates. Silver was deposited from both direct and back diffusive fluxes in high-purity argon at pressure 3-6 Pa. In order to prevent substrate heating the deposition mode was noncontinuous with sufficient interruptions. Deep cleaning of the working gas was carried to avoid the formation of impurity phases in the deposition process. Firstly, degassing was carried by working chamber pumping and then the chamber was cut off from the pumping system with further argon filling. On the next step titanium sputtering has been performed during more than 20 h. The titanium film was deposited on the chamber walls. It is well known that the titanium has good getter properties. Therefore the chemical active gases were adsorbed by this film. At the final stage of purification the total partial pressure of chemically active gases (mainly O_2, H_2 and N_2) was ~ $8 \times 10^{-8} - 10^{-7}$ Pa [7].

Prior to deposition, the substrates were exposed to plasma in order to create point defects in the form of Cl^- vacancies that was considered as a primary factor to lower the percolation threshold.

3. RESULTS AND DISCUSSION

Form fig.1a it follows that at the initial growth stage a continuous and very thin metallic layer was formed. It has disordered quasiamorphous structure – due to the fact that sputtered atoms have energy of a few eV and can penetrate up to two monoatomic layers deep.

We estimate thickness of this layer as ~1.5-3 nm. Besides, fig.1a depicts nucleation of crystals. Such kind of growth can be called as pseudomorphic growth of amorphous phase.

During further deposition amount and sizes of the crystals increase (fig.1b), while electron diffraction patterns show diffraction maxima of silver. Similar result is obtained when depositing back diffusive fluxes (fig.2a,b).

In this case crystalline state appears earlier. These results are obtained at substrate temperature range 35-40 0C, discharge current 30 mA and discharge voltage 170 V. By means of relatively low temperatures of

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the growth surface we have tried to reduce surface mobility of silver adatoms and prevent creation of crystals. Also we have tried to increase structural stability by depositing relatively weak vapor fluxes, using the plasma-condensate system and increasing the working gas pressure.

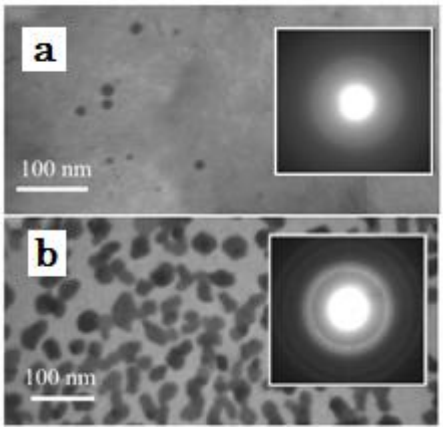

Fig 1 – TEM images of silver films growth stages deposited from direct diffusive flux (a - deposition time is 1 min, b - deposition time is 1.5 min)

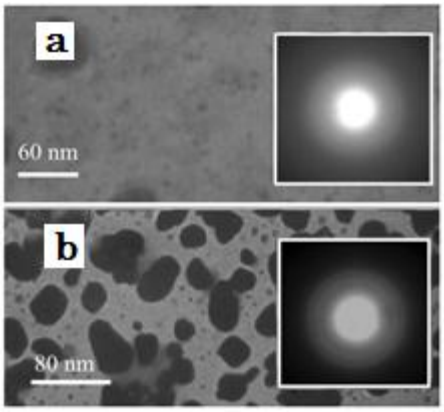

Fig 2 – TEM images of silver films growth stages deposited from back diffusive flux (a - deposition time is 1 min, b - deposition time is 2 min)

Under the same technological conditions Ag layers were fabricated on KCl substrates, preliminarily coated with a polymer (PC403 or ma-N405). Observed structure is approximately the same (fig. 3). However, one should remember that in this case in TEM we investigate two layers simultaneously – Ag and polymer. Hence, it does not seem possible to determine the Ag layer thickness. Also one can pay attention to the absence of Ag crystals on certain areas and well-defined faceting of the crystals.

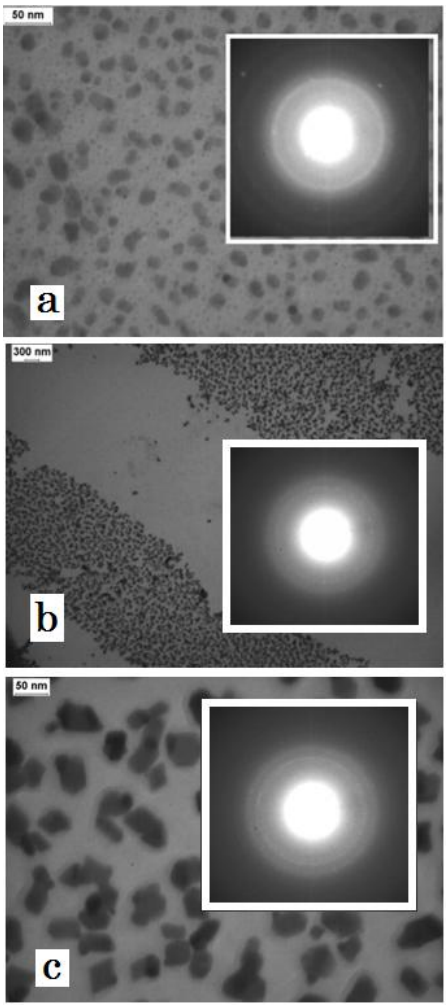

Fig 3 – TEM images of silver layers obtained on polymers (2 min; back diffusive fluxes (a); direct diffusive fluxes (b,c))

If to decrease deposition time in case of polymer, up to now we did not succeed to find essential structure changes. Probably, it is connected with more intense formation of crystals on polymer.

4. CONCLUSIONS

1. A solid amorphous layer is formed on the initial stage of film growth on cleaved KCl facets.
2. As thickness of the amorphous layer increases, a transition to crystalline state occurs because of increase in structural non-equilibrium.
3. As soon as crystalline nuclei have been formed, their growth becomes predominant.
4. In order to reduce the percolation threshold, it is necessary to create a system of active centers on a surface and deposit substance at low supersaturation.

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