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Insulator–metal transition in a conservative system: An evidence for mobility coalescence in island silver films

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Abstract. Aging, which manifests itself as an irreversible increase in electrical resistance in island metal films is of considerable interest from both academic as well as applications point of view. Aging is attributed to various causes, oxidation of islands and mobility of islands followed by coalescence (mobility coalescence) being the main contenders. The effect of parameters like substrate temperature, substrate cleaning, residual gases in the vacuum chamber, ultrasonic vibration of the substrate, suggest that the mobility coalescence is responsible for the aging in island metal films. Electron microscopy studies show evidence for mobility of islands at high substrate temperatures. The comparison of aging data of island silver films deposited on glass substrates in ultra high vacuum and high vacuum suggests that the oxidation of islands, as being responsible for aging in these films, can be ruled out. Further, under certain conditions of deposition, island silver films exhibit a dramatic and drastic fall in electrical resistance, marking the insulator–metal transition. This interesting transition observed in a conservative system – after the stoppage of deposition of the film – is a clear evidence for mobility coalescence of islands even at room temperature. The sudden fall in resistance is preceded by fluctuations in resistance with time and the fluctuations are attributed to the making and breaking of the percolation path in the film.

Keywords. Island silver films; mobility coalescence; insulator–metal transition; electrical resistance; aging.

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1. Introduction

Thin film growth on any substrate occurs in one of the two broad categories of growth modes, namely, layer by layer growth mode or Stranski–Krastanov growth mode and island growth mode or Volmer–Weber growth mode. Surface and interfacial energies determine the growth mode of a film. In general, vapor deposition of metal films on glass substrates grow by island mode. In the island mode, vapor atoms impinging on the substrate surface get adsorbed and are known as adatoms. These adatoms migrate on the substrate surface to form nuclei. When two nuclei touch each other they coalesce to form a larger cluster. As the deposition continues, at a particular surface coverage, a large scale coalescence (LSC) takes place forming a network structure leaving holes and channels in between. Secondary nucleation in the holes and channels fill up, to form a continuous film.

Therefore, by limiting the growth of a film to nucleation stage or by avoiding excessive coalescence, island films or discontinuous films consisting of an array of discrete islands with statistical distribution of island radii and separations, can be obtained. Although, such films have many attractive properties, they cannot be exploited yet for device applications due to their inherent temporal instability or aging even in vacuum. Aging manifests as an irreversible increase in their dc electrical resistance.

2. Aging studies

The expression for electrical resistance of an island film is exponentially dependent on the average inter-island spacing [1,2] and aging is attributed to the increase in average island spacing following the stoppage of deposition. The increase in the average island spacing can come about by island shape changes [3], oxidation of islands [4] and mobility of islands followed by coalescence [5]. Oxidation of islands model and mobility coalescence model are the main contenders in explaining the aging process in island metal films. Studies on the effect of residual gases on the aging of island silver and copper films through the quantification of the aging process indicated that oxidation might not be responsible for aging in these films [6,7]. The substrate temperature effect too supported this view [8].

As the deposited material in the film is increased, the average island size increases and average island spacing decreases resulting in lower resistance of the film. If mobility coalescence is operative, a reduced aging rate is expected for higher thickness or lower resistance films, as mobility is size dependent, larger islands being less mobile. If the deposition continues, a LSC would occur and a very small aging rate is expected due to the incorporation of small islands to the network structure.

Table 1 shows typical coalescence rates, defined as the change in tunneling length per minute calculated over fixed time interval [9], for silver films under different conditions of ultrasonic vibrations of the substrate. Here, NV refers to no vibration of the substrate either during deposition or during aging. VD is the condition wherein substrate is vibrated only during deposition. VA refers to the vibration only during aging [10]. It is clear from the table that larger islands formed due to vibration during deposition show lower coalescence rate as compared to NV condition. The films formed under similar conditions show higher coalescence rate when the films are subjected to vibration during aging (NV and VA). Further, lower initial resistance (resistance immediately after the deposition is stopped) films show lower coalescence rate as they contain larger islands. These observations strongly support mobility coalescence model.

Table 1. Coalescence rates for silver island films under different conditions of substrate vibrations.

	Coalescence rates (Å/min)		
Initial resistance	NV	VD	VA
2 MΩ/□	0.0656	0.0345	0.0889
10 MΩ/□	0.1284	0.0831	0.1566
20 MΩ/□	0.2209	0.1575	0.3179

The silver island films on glass substrates studied under a UHV of 2×10^{-8} Torr with an oxygen partial pressure of the order of 10^{-11} Torr, too show considerable aging for extended periods. Further, coalescence rates under UHV and HV of 2×10^{-6} Torr – HV obtained using an oil diffusion pump – are nearly the same [11]. These results further strengthen the mobility coalescence model.

3. Insulator–metal transition

It is well-known that after the occurrence of LSC, very little material is required to be deposited to form a semicontinuous film with a connecting path between the electrodes being established. This is the discontinuous – semicontinuous transition often termed as insulator–metal transition in the case of granular metal films. Island silver films deposited on glass substrates with an initial resistance of 2 MΩ/□, at a deposition rate of 0.2 Å/s exhibit a very interesting behavior and is shown in figure 1. The film shows a small aging rate for a few minutes of aging. Then the resistance falls drastically, after which the resistance remains almost steady. The aging is characteristic of island or discontinuous films while a steady decrease in resistance with time is the property of a continuous or a semicontinuous film. The behavior exhibited by this film clearly indicates an insulator–metal transition. The small aging rate observed before the transition indicates that the LSC stage has already been crossed for this film. At a very low deposition rate of 0.2 Å/s agglomeration would be much less. Therefore, one can expect LSC to occur at an earlier stage itself as the degree of agglomeration determines the thickness at which the film tends to become continuous [12]. However, insulator–metal transition occurring after the stoppage of deposition is a fascinating new result.

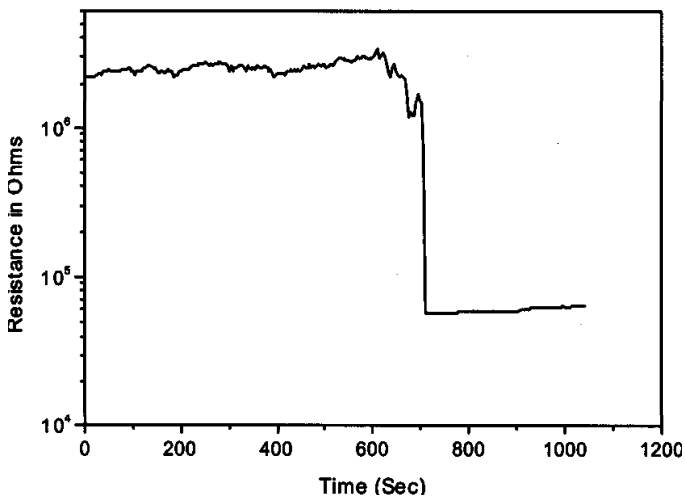

Figure 1. The variation of resistance with time after the stoppage of deposition for a silver film of thickness ∼ 100 Å.

The insulator–metal transition in a conservative system may be explained as follows: at very low deposition rates, not only LSC occurs at an early stage of deposition, but, due to reduced agglomeration, very little material is required to connect the large network to establish a percolation path between the electrodes. The incorporation of secondary nuclei into the network structure due to their mobility may result in a percolation path leading to an insulator–metal transition. This cannot be expected in a more agglomerated film deposited at higher deposition rates as more material is required to establish a percolation path and just the incorporation of secondary nuclei would not be sufficient for this purpose. As a consequence, insulator–metal transition after the stoppage of deposition is not observed for films deposited at higher rates [10].

The other interesting feature is that the transition is not a gradual decrease of resistance but a sudden one and is preceded by fluctuations in resistance with time. These fluctuations in resistance can, in principle, be due to the dynamic fluctuations in the average island spacing. But, such dynamic fluctuations in average island spacing due to the mobility of islands should always be present, not only in the vicinity of insulator–metal transition. Under the conditions that are not leading to such a transition, the variation of resistance with time was always found to be smooth. On the other hand, these fluctuations can be due to the making and breaking of the circuit or the conducting paths between the networks. The decrease in resistance is due to the filling up of the gaps between the networks by the secondary nuclei. The gaps filled by these secondary nuclei forms a weak electrical link between two large networks. The typical size of secondary nuclei can be from a few to a few tens of nanometers. The current applied by the resistance-measuring instrument used in the present studies (Keithley Electrometer model 617) is only of the order of microamperes. But still, the current density flowing through these weak links would be very large ($\sim 10^6$ A/cm^2 for the 10 nm cluster) and can cause catastrophic destruction of these weak links. This would give rise to the breaking of the conduction paths established, increasing the film resistance. At the onset of insulator–metal transition, a large number of parallel conducting paths between the electrodes are established and therefore, the current density gets distributed, avoiding the destruction of the weak links. Therefore, the film resistance remains steady after the transition.

4. Conclusion

The conclusions that can be drawn from the aging studies of silver island films are summarized as follows: (a) the effect of various parameters on the aging rates of silver island films suggest that the mobility of islands followed by coalescence is responsible for aging in these films; (b) an interesting insulator–metal transition is observed under certain conditions in silver films, long after the stoppage of deposition. This is attributed to the establishment of metallic conduction paths due to the incorporation of secondary nuclei to the network structure. The transition is preceded by large fluctuations in resistance; (c) the fluctuations in film resistance is attributed to the formation of conduction paths and their breakage due to the passage of high current density at the weak links in the path; and (d) the insulator–metal transition observed in a conservative system is a clear evidence for the mobility of islands.

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