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Aging and field-effect studies of Cu island films at near liquid-nitrogen temperatures

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We report the results of the investigation carried out on the aging and field effect of discontinuous copper films deposited on glass substrates held at around 125 K. Using the scheme adopted in our earlier investigations, it was found that even at this temperature there is considerable increase in resistance. However, the agglomeration rate is much less than for films deposited at room temperature. The *I-V* characteristics of the films revealed an irreversible change in resistance following removal of the field. On stabilization, heating of the films revealed a decrease in resistance with temperature followed by an irreversible resistance increase beyond a transition temperature which was less than room temperature.

The instability in the physical properties of discontinuous films is a serious problem in exploiting the attractive properties of these films. This instability is seen as an increase in the post-deposition resistance with time. The resistance increase has been attributed to various causes.[1–6] Electron microscopy studies[5,6] indicated that mobility coalescence leading to an increase in the average interisland spacing was responsible for the post-deposition resistance increase. At high fields, the nonlinearity in *I-V* characteristics of island films has been reported by researchers and interpreted physically.[7–9] Earlier investigations have been carried out in our laboratory on the influence of different parameters on the aging behavior of copper and silver island films.[10,11] This study was carried out to test the validity of the thermally activated mobility coalescence model and its applicability to explain the aging behavior in island copper films.

Island films of copper (purity 99.999%) were deposited onto clean glass substrates at a pressure of 1×10^{-6} Torr. The purity of the copper was checked using x-ray photoemission spectroscopy (XPS). The substrates were maintained at a temperature of nearly 125 K by fixing them to a liquid-nitrogen-cooled cryostat. The film dimensions were 1×1 cm^2 with a substrate-to-source distance of 20 cm. The film resistance during and after growth was monitored using a Keithley electrometer. During deposition of the films, the rise in substrate temperature was not more than 5 K. Following stabilization of the film resistance with time, *I-V* characteristics were studied and the change in film resistance on application of a high field was noted. The temperature variation of resistance was studied from 125 K to room temperature by attaching a heater to the cryostat.

The films were studied under two different conditions:

(1) under ambient conditions with the application of glow discharge for 5–7 min, of initial resistance $R_0 = 0.18$, 1.3, 7.4, 14.5, 40, and 60 MΩ/□;

(2) dehydrated condition obtained using P_2O_5 and anhydrous $CaCO_3$ moisture traps inside the vacuum chamber for 48 h before depositing the film. During this period a

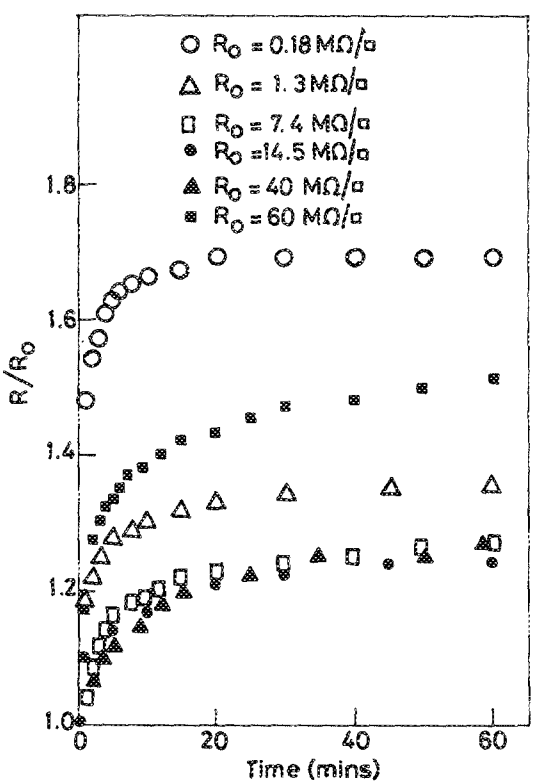

FIG. 1. Variation of normalized resistance with time for the films studied under condition (1).

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TABLE I. Agglomeration rate data at 125 K and at room temperature.

R_0(MΩ/□)	Agglomeration rate
At 125 K	
Condition 1	
0.18	0.0417
1.3	0.0363
7.4	0.0485
14.5	0.0344
40	0.0512
60	0.0644
Condition 2	
7	0.0568
13.2	0.0532
At room temperature[a]	
40.7	0.1308
61	0.2238

[a] See Ref. 10.

vacuum was maintained in the chamber and glow discharge was employed periodically. Films of $R_0 = 7$ and 13.2 MΩ/□ were studied in this condition.

One film of $R_0 = 10.7$ MΩ/□ was deposited at room temperature without using glow discharge under the undehydrated condition.

For all the films studied, the functional dependence of the film resistance on time was of the form $\ln(R) \propto \ln(t)$ with the constant of proportionality $d(\ln R)/d(\ln t)$ defined as the agglomeration rate calculated by a least-squares fit. The plot of normalized resistance versus time is shown in Fig. 1 for films studied in condition (1). Table I gives the agglomeration rates at 125 K for the two conditions and some typical values at room temperature for comparison. Once the film resistance stabilized, field-effect studies were carried out. The variation of current (I) with voltage (V) for the films of $R_0 = 1.3$, 7.4, 14.5, and 60 MΩ/□ studied in condition (1) is shown in Fig. 2. It can be seen that except for the film of $R_0 = 7.4$ MΩ/□, which is almost linear, all others show a nonlinear I-V behavior. There exists a nonlinear region marked between the vertical lines in the figure, before and after which it is nearly linear. A film of $R_0 = 0.18$ MΩ/□ studied under condition (1) showed metallic conduction and considerable Joule heating when kept at a voltage of 35 V. A temperature increase of nearly 35 K was observed. The I-V characteristics for the films studied under condition (2) were similar to those of condition (1). Table II gives the resistances before and after field-effect studies were carried out. Figure 3 shows the temperature variation of resistance for films of initial resistance 40 and 14.5 MΩ/□ of condition (1) and a film of $R_0 = 10.7$ MΩ/□ deposited under ambient conditions at room temperature. A transition temperature is seen below which the 40- and 14.5-MΩ/□ films show a reversible decrease in resistance with temperature and above which the resistance increases irreversibly. For the $R_0 = 40$ MΩ/□ film, heating and cooling cycles were repeated below the transition temperature as shown in the figure. The 10.7-MΩ/□ film cooled and heated below room temperature showed no transition.

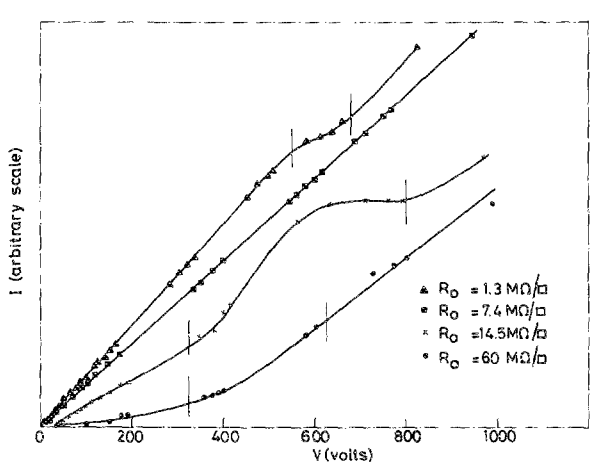

FIG. 2. I-V characteristics for films studied under condition 1.

The expression for the resistance of an island film based on the Neugebauer and Webb[7] model is exponentially dependent on the average interisland spacing. Damodara Das and Murali Sastry[10] concluded that mobility coalescence leading to an increase in the interisland spacing is responsible for the resistance increase in Cu and Ag films.

The diffusion coefficient of an island containing n atoms is[12]

$$D_n = D_0 \exp(-E_n/kT), \tag{1}$$

where E_n is the island-size-dependent activation energy for surface diffusion.

The mobility of islands on the substrate surface is a thermally activated process. One would expect a decrease in the agglomeration rate at near-liquid-nitrogen temperatures. Table I indicates that the agglomeration rate at 125 K is very much less than that at room temperature for films with nearly the same initial resistance. There is little difference in agglomeration rates for films of almost equal resistances studied in conditions (1) and (2), indicating that water vapor does not affect the aging process at 125 K.

From Fig. 2 it is obvious that the V^2 dependence of I at high fields as suggested by Milgram and Lu[9] does not hold. Resistance measurements after I-V characteristics studies revealed a change in resistance in all cases. Except for the $R_0 = 14.5$ MΩ/□ film, there was a reduction in resistance for the other films (Table II). This indicates that a structural change had taken place (the nonlinear region marked between the vertical lines being the region of structural change). In the $R_0 = 0.18$ MΩ/□ film Joule heating as well

TABLE II. Resistance before (R_1) and after (R_2) I-V measurements.

R_0(MΩ/□)	R_1(MΩ/□)	R_2(MΩ/□)
Condition 1		
1.3	1.75	1.18
7.4	9.38	7.2
14.5	18	23
60	94	36
0.18	0.307	0.007
Condition 2		
7	9.3	8.9
13.2	17.8	9.5

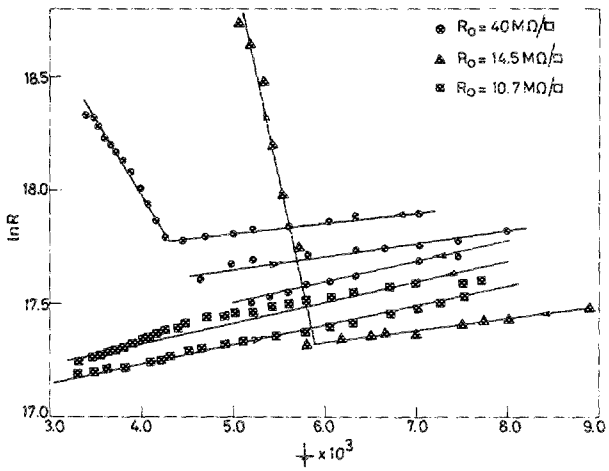

FIG. 3. ln R vs $1/T$ plots for two films in condition (1) and a film deposited at 300 K.

as the high field are responsible for the structural change, causing a decrease in resistance. Although the applied voltage was only 35 V, the fact that interisland spacing is very small results in a high microscopic field between the islands. Joule heating indicates that a metallic conduction path has been established between the electrodes.

At high fields, it is possible that polarization of the islands may induce strains large enough to break the islands. This would give rise to an irreversible fall in resistance as is observed for the films of $R_0 = 0.18$, 1.3, 7.4, and 60 MΩ/□ in condition (1) and $R_0 = 13.2$ and 7 MΩ/□ in condition (2). The increase in resistance upon application of the field in the case of the $R_0 = 14.5$ MΩ/□ film could be due to field-induced coalescence of the smaller islands resulting in an increase in the average interisland spacing and hence film resistance. Even though relatively high wattage power existed at high fields (0.1 W for the 10-MΩ/□ film at 1000 V), heating effects being responsible for the structural changes are ruled out because the film temperature was maintained constant at 125 K.

From the temperature response of resistance (Fig. 3) two distinct regions separated by a transition temperature are seen. A similar transition was observed earlier by us.[11] Below the transition temperature, repeated heating and cooling gives rise to almost the same slope in $\ln(R)$ vs $1/T$ curves (Fig. 3). This indicates that no structural changes are taking place. Near the transition temperature movement of islands gives rise to a small structural change and hence a shift in resistance. A similar transition was observed by Feldman,[13] who assumed structural stability since the films were annealed at 600 °C and measurements were made below this temperature. In our case, Feldman's interpretation does not hold due to the resistance increase being irreversible. The $R_0 = 10.7$ MΩ/□ film deposited at room temperature and subsequently cooled and heated below room temperature (Fig. 3) showed no transition temperature with a steady increase of resistance with falling temperature. This strengthens our model of activated coalescence leading to a resistance increase beyond the transition temperature.

At a pressure of the order of 1×10^{-6} Torr, water vapor forms 70%–90% of the rarified atmosphere.[14] If there is any gettering on the substrate, in all probability it is water vapor, and while heating one would expect an abrupt change in resistance when defrosting starts. This was not observed, indicating that gettering may be ruled out. This is supported by the fact that there is not much difference in agglomeration rates for the films of almost equal resistances studied under undehydrated and dehydrated conditions (Table I).

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