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Letter

The post-deposition resistance increase in discontinuous metal films

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In a recent paper[1], Nishiura and Kinbara have attempted to explain the resistance increase often observed immediately following deposition of a discontinuous metal film. The model proposed is based on shape changes of the metal islands whereby an island, originally in the form of a rotational ellipsoid, tends with time toward a spherical form. The resistance change arises from the resulting increase in the inter-island tunnelling distance. It is assumed in the model, however, that tunnelling takes place between the island extremities (*i.e.* across the vacuum gap), whereas it is widely accepted that tunnelling takes place through the substrate[2]. The latter view is supported by phenomena which are explained by substrate effects[3]. The purpose of this letter is to propose an alternative model, also based upon a changing island shape, which includes substrate-assisted conduction in the explanation of resistance increases. The revised model is also based upon a proposed mechanism which seems physically reasonable.

During the island nucleation and growth process, mobile adatoms on the substrate surface are absorbed by the metal islands. This process will presumably continue briefly after deposition ceases, but since free adatom lifetimes ($\sim \mu$s) [4] are much less than the time scale of the observed resistance increases (1 - 100 min) [1] adatom migration effects are ignored in this treatment. The essential point here is that the adatoms will reach the island and adhere at the circumference of the plane of contact between island and substrate. Transient island shape changes after deposition are therefore likely to be related to transitory effects at this island–substrate junction. It is assumed that the islands have the form of a spherical cap of constant volume at all times, but that the contact angle θ ($> \pi/2$) changes with time from an initial value θ_i toward the equilibrium value θ_e, where $\theta_i < \theta_e$ due to the adherence of adatoms. As θ increases, the tunnelling distance d to the next island also increases and the resistance R increases from its initial value R_i.

Varma *et al.* have evaluated the transient variation of contact angle for a spreading sessile drop[5] which is formally identical to the present model but

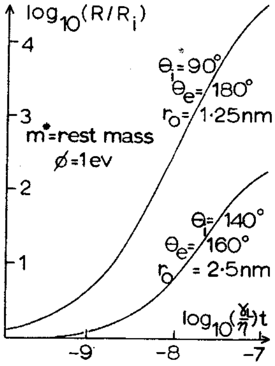

Fig. 1. Theoretical calculation of the resistance increase.

than $(2\gamma_L/\rho g)^{1/2}$ also holds for the islands of a discontinuous metal film. (γ_L is the island surface energy, ρ the island density and g gravitational acceleration.) The variation of θ with time is given by

$$\frac{\gamma_L}{\eta r_0}\ t = 3 \times 2^{2/3} \int_{\theta_i}^{\theta} \frac{(1 - \cos\theta)^2\ d\theta}{(2 - 3\cos\theta + \cos^3\theta)(\cos\theta - \cos\theta_e)}$$

where η is the island viscosity and $(4\pi/3)r_0^3$ is the island volume.

Film resistance is proportional to $\exp(4\pi d/h)\ (2m^*\phi)^{1/2}$ where h is Planck's constant, m^* the effective mass of the tunnelling electron and ϕ the tunnelling barrier[1,6]. As θ changes, the tunnelling path length change each island interface equals the variation in $r\sin\theta$, where

$$r = r_0\{4/(2 - 3\cos\theta + \cos^3\theta)\}^{1/3}$$

is the instantaneous radius of the spherical cap. Two calculated plots of $\log_{10}(R/R_i)$ are shown in Fig. 1 as functions of normalised time. The islar dimensions are of the order of those noted by Nishiura and Kinbara. Islar sizes tend to increase with average film thickness so the plot for the small island radius will correspond to that for a thinner film. (Note that the calculation assumes contact angle variation at one end of the tunnelling path only; the vertical scale should be doubled for tunnelling between identical islands.)

θ_e is expected to increase as island sizes decrease, due to the increase surface energy[7]. For the smaller islands, the adherent adatoms are expect to have a larger effect upon the contact angle, *i.e.* θ_i is expected to be low These two assumptions are necessary for the simple model described to predict the observation[1] of larger changes occurring earlier for smaller islands. Time has been left normalised since the quantitative effects of isla size on γ_L and η are unknown. η is expected to be smaller for smaller islands due to the "liquid-like behaviour"[8] caused by melting-point depre sion[9]. The most encouraging aspect of the model is the prediction that th majority of the resistance increase is completed over two decades of time, agreement with the results of Nishiura and Kinbara[1]. This prediction is ot affected by the numerical choice of any parameter. Comparing Fig. 1 ith Fig. 4 of Nishiura and Kinbara's paper[1] suggests that $\eta \sim 10^{11}$ J m^{-2} s vith $\gamma_L \sim 1$ J m^{-2} for Au). The value does not seem unreasonable[10] but annot be checked independently.

This letter has omitted any consideration of the effect the proposed nape changes will undoubtedly have upon the activation energy, particularly or small islands. Activation energies are difficult to calculate for non-ideal sland shapes but using $\{\mathrm{vol}/(4/3)\pi r^3\}\, e^2/4\pi\epsilon r$ as an estimate suggests that the mission leads to an error of less than 8% in the 2.5 nm curve (Fig. 1) at 00 K, but that the resistance change for the 1.25 nm island will be greater han that shown by a factor of 10. Island coalescence has also been disegarded.

As far as the model can be taken, it appears to offer a plausible explanation of the observations of Nishiura and Kinbara[1]. It cannot, however, xplain the resistance decreases sometimes observed[11,12], but neither can ther models[1,11]. No attempt has been made to fit the results precisely, ince there are too many unknowns involved. In particular, it is doubtful whether θ_i and η (for small islands) could ever be independently evaluated. Since precise calculations are not possible, thermal transients[1,10] have also been neglected.

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