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# The role of grain boundary sliding in solid-state dewetting of thin polycrystalline films

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We present experimental evidence that grain boundary sliding contributes to the mass transport associated with solid-state dewetting of a thin polycrystalline film of gold on sapphire. A model describing the retraction kinetics of the edge of polycrystalline thin film controlled by combined surface/interface diffusion and grain boundary sliding is proposed. The model predicts stepped film morphology in the vicinity of expanding hole, in good agreement with the experimental data.


*Keywords:* Thin films; Surface diffusion; Surface energy; Facets; Grain boundary sliding

Solid-state dewetting of thin metal films has attracted a great deal of attention recently as a promising method for the formation of ordered nanoparticle arrays [1–3]. This process is driven by the decrease in the total energy of all surfaces and interfaces in the system, while its kinetics is controlled by solid-state diffusion. Surface diffusion was considered as the main mass transport mechanism of solid-state dewetting [1,4], yet the importance of grain boundary (GB) [5] and interface [6] diffusion in the process has recently been demonstrated.

The role of GBs in solid-state dewetting of thin films (beyond being the sites of preferential hole nucleation) has rarely been considered. Müller and Spolenak [7] demonstrated that branched holes formed during solid-state dewetting of thin Au films on $SiN/SiO_2/Si$ substrates develop protrusions in the direction of high-angle GBs. Kovalenko et al. [5] explicitly took into account the contribution of GBs to the driving force of dewetting in their kinetic model.

It is well known that the GB sliding in polycrystalline thin films provides an important contribution to stress relaxation in the film [8] and to the overall plastic strain during plastic deformation of the film [9], and plays an important role in the formation of stress-relaxing hillocks and whiskers [10]. The capillary stresses arising at the growing hole edge are no different from the externally applied or internal stresses in the film, and they can also activate the mass-transporting GB sliding. To the best of our knowledge, the role of GB sliding in solid-state dewetting of thin films has not been considered in the literature. In this letter we present experimental evidence for GB sliding in the vicinity of expanding holes in the thin Au film. Based on these observations, we propose a new solid-state dewetting mechanism based on coupled surface/interface diffusion and GB sliding.

We deposited 25 nm thick Au film on c-cut sapphire substrate employing the magnetron sputtering technique. The films exhibited strong $\langle 111 \rangle$ out-of-plane texture and random in-plane grain orientations. Several consecutive heat treatments at 400 °C in air were performed (the total cumulative annealing time was 180 min). After each treatment, the surface topography of a single region of the film was studied by atomic force microscopy (AFM; Park Systems XE-70) in intermittent-contact mode. After the last treatment, hole edges were characterized by high-resolution scanning electron microscope (Zeiss Ultra plus HR-SEM). The average grain size in the as-received film determined from the AFM images was 35 nm.

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After the first treatment (15 min), small randomly distributed holes appeared in the film (see Fig. 1a). The holes expanded with each subsequent heat treatment, but no additional newly nucleated holes were observed. Thus the dewetting rate of the film is governed by the holes' expansion rate. Generally, the observed morphology of holes and grains was consistent with the morphologies reported by Müller and Spolenak [7]. Like in their study, we found that the branched holes expand in the direction of GBs, that the holes edges are faceted and that grains in the vicinity of growing holes are larger than their counterparts in the remaining film (see Fig. 1a and b).

However, the surface topography in the vicinity of the holes developing after long anneals was very different from the classical picture of surface diffusion-controlled hole growth [1,4]. Figure 1a and b presents 3-D AFM images of a single hole taken after annealings for 15 and 60 min (cumulative annealing time). After the longer anneal, a terrace-like topography of elevated grains develops in the hole rim region, with higher grains being closer to the hole edge. The top surface of these grains is flat rather than hemispherical, indicating that an important role is played by surface anisotropy in the dewetting process. The elevated grains can be also seen at some distance from the hole (Fig. 1b). The line topography profiles taken across the holes are shown in Figure 1c. In the case of a hole formed after the 60 min anneal, the following features can be identified in the profile: (i) narrow GB grooves with topography strongly affected by surface anisotropy [6,11]; (ii) steps at the GBs; and (iii) GB steps combined with the GB grooves, typical for the situation in which the GB grooving occurs simultaneously with the GB sliding [12]. The small lateral size of the GB grooves (compared with the overall lateral size of the elevated rim) indicates that surface diffusion alone cannot explain the observed expansion rate of the holes. Moreover, a detailed quantitative study of the mass balance in the vicinity of the holes demonstrated that only about half of the material rejected by the growing hole is accumulated in the nearby ridge [13]. The rest is distributed in the more remote regions of the film, causing formation of elevated grains at some distance from the ridges. This again indicates that some alternative (to the film upper surface) diffusion paths contribute to dewetting. After annealing for 180 min, the holes become interconnected, exposing a large fraction of the substrate area (Fig. 2). The characteristic features observed in the AFM images for shorter annealing times can be also recognized in Figure 2: a terraced morphology of the elevated hole rim, with the GB steps indicative of GB sliding.

We propose a one-dimensional dewetting model of polycrystalline thin film with rectangular faceted grains (see Fig. 3a). The terraced morphology of the hole rim with well-defined GB steps implies that shape-changing diffusion on the upper surface of the film is slow. In the simple one-dimensional model considered here, the only alternative diffusion path allowing mass transport along the substrate away from the growing hole is diffusion along the film–substrate interface. Thus, we assume that Au atoms diffuse along the hole edge (this type of diffusion determines the rate of edge movement), with subsequent diffusion along the interface away from the growing hole and accretion at the interface. As a result, the width of the first grain decreases, and the height of the near-hole grains increases.

We employed the concept of weighted mean curvature to describe the diffusion along the vertical edge of the first near-hole grain [14–17]. This resulted in the following expressions for the surface diffusion flux at the interface, $j_{y\to 0}$, and for the edge migration rate:

$$j_{y\to 0} = \frac{3D_s v_s \Omega}{h_0 kT}\left(\frac{\Delta\gamma_x}{h_0} - \sigma_0\right) \quad (1)$$

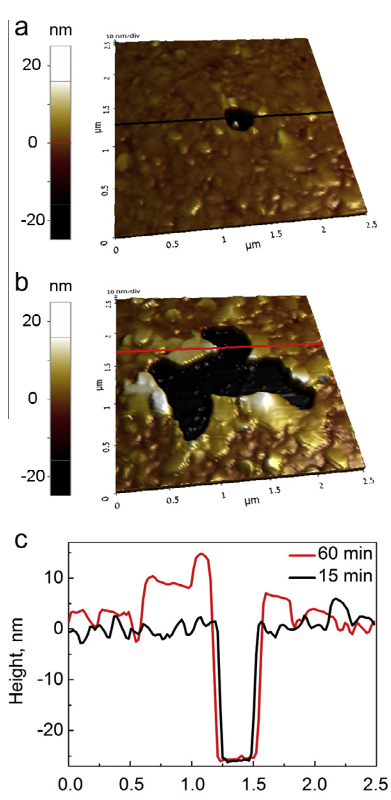


**Figure 1.** Three-dimensional AFM images of a single hole in thin Au film after annealing for (a) 15 min and (b) 60 min, and (c) the corresponding line topography profiles taken along the black and red lines. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

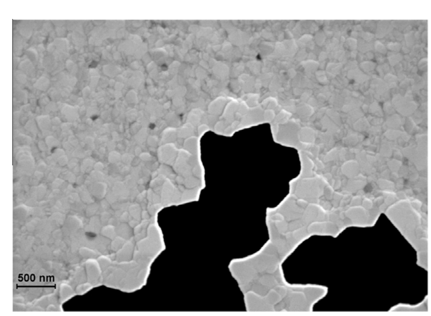


**Figure 2.** HR-SEM image of a hole formed after annealing for 180 min. A terraced grain morphology is clearly visible in the vicinity of the hole.