Scripta Materialia 134 (2017) 115–118

Contents lists available at ScienceDirect

Scripta Materialia

journal homepage: www.elsevier.com/locate/scriptamat

Regular Article

Grain boundaries effects on hole morphology and growth during solid state dewetting of thin films

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ARTICLE INFO

Article history:
Received 4 January 2017
Accepted 26 February 2017
Available online xxxx

Keywords:
Dewetting
Surface diffusion
Grain boundaries
Simulation
Thin films

ABSTRACT

We probe the effects of grain boundaries on thin film dewetting numerically for initially circular and elongated holes in single and bi-crystalline systems. We find that grain boundaries play a critical role in determining hole morphologies, in particular in the formation of highly branched hole morphologies, since while elongated holes in single crystals tend to become rounder over time, circular holes in bi-crystals destabilize, leading to accelerated dewetting along the grain boundaries.

1. Introduction

Upon heating, thin films deposited on a foreign substrate lose their stability, and transform via dewetting into an array of isolated islands [1]. In the solid state, the dewetting process is controlled by surface diffusion, with possible contributions from alternative short-circuit diffusion paths [2,3]. Typically several stages can be distinguished: (i) formation and nucleation of holes, (ii) expansion of the holes, (iii) break-up of percolating clusters of the film material and formation of isolated particles/islands. This article focuses on the second stage of the process within the context of thin polycrystalline films. This stage is important since for a given density of nucleating holes, the morphology of the growing holes determines the average size and spacing of the particles which are seen during the final stage of dewetting [4].

Hole expansion dynamics in thin polycrystalline films are qualitatively different than in thin heteropitaxial single crystalline films. Holes formed in anisotropic single crystalline thin films exhibit well-defined geometric shapes reflecting the intrinsic surface anisotropy and atomic structure of the film material [5,6]. The holes formed in continuous polycrystalline thin films often exhibit irregular, fractal-like branched shapes [7,8], and hole expansion is accompanied by grain growth in the immediate vicinity of the hole [9,10].

Several hypotheses have been put forward to explain the appearance of unusually shaped branched dewetting holes. Kwon et al. proposed that a Rayleigh-type instability of the ridge surrounding the hole can lead to branching, resulting in dendrite-like hole morphologies [8,11]. Such morphologies have also been observed during dewetting of some single crystalline semiconductor thin films [12]. Kosinova et al. suggested that anisotropy in the surface diffusion coefficient along the hole edge can lead to branching [13]. They performed dewetting heat treatments in different annealing ambients and demonstrated that an increase in the fraction of faceted surfaces in the forming gas atmosphere can result in a decrease in the extent of hole branching. Recently, Müller and Spolenak observed that during the dewetting of a polycrystalline Au film deposited on silica, the hole branches protrude in the direction of large angle grain boundaries, and they proposed that grain boundary grooving-like processes plays a crucial role in developing branched hole morphologies [9].

While the suggestions above sound plausible and the contribution of grain boundaries to the overall thermodynamic driving force for dewetting has been considered in the past [2], to the best of our knowledge no quantitative treatments of the effect of grain boundaries on the morphology of growing dewetting holes are available in the literature. In the present work we breach this gap by demonstrating that a single grain boundary can induce a shape

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http://dx.doi.org/10.1016/j.scriptamat.2017.02.046
1359-6462/

instability in an initially circular dewetting hole. This result reflects part of an ongoing study of the dynamics of wetting and dewetting, with and without grain boundaries, based on careful numerical simulations [14]. While numerous simulations of wetting and dewetting have been undertaken [15–17], they failed to address the influence of grain boundaries on the dewetting process.

2. A numerical model

To explore hole evolution and the morphology resultant upon dewetting, we follow the evolution of a thin film containing a single hole, which is initially either circular or elongated. The thin film and hole are located within a bounding cylinder: $(x/r_x)^2 + (y/r_y)^2 = 1$, $z \geq 0$. To compare dewetting in single and polycrystalline thin films, similar geometries are considered with and without grain boundaries, see Fig. 1 and Fig. S1, Supplementary materials.

The thin film is assumed to evolve by surface diffusion, which for simplicity is assumed to be isotropic. Along the moving contact line where the thin film meets the substrate, the contact angle, θ_c, is prescribed in accordance with the energies of the film surface, the substrate, and the film-substrate surface. The thin film is assumed to normally intersect the bounding cylinder. The system is mass conservative with no mass flux through the bounding surfaces or at the contact line. In systems containing a grain boundary, the dihedral angle, θ_d, satisfies $\theta_d = \pi - 2\arcsin(m/2)$ with $m = \gamma_{\text{grain}}/\gamma_{\text{exterior}}$, where γ_{grain} (γ_{exterior}) denotes the free energy of the grain boundary (the exterior surface).

Mirror symmetry is assumed along the x-z and z-y planes, see Fig. 1, which enables numerical simulation of only one quarter of the domain. The grain boundary initially lies within the x-z plane, and remains within the x-z plane by mirror symmetry. All our simulations are three dimensional. In the initially circular holes, the initial hole was a quarter circle with initial radius $x_c = y_c$, and in the initially elongated holes, the initial hole consisted of a rectangular region with a quarter circle attached at one end, yielding a hole initially satisfying $x_c > y_c$, where x_c (y_c) denotes the distance of the contact curve from the z-axis along the x-axis (y-axis). The projection of the initial thin film configuration on the x-z and y-z planes consisted of a part of circle with radius $l_z/(1-\cos\theta_c)$ smoothly attached to a linear segment with uniform height l_z; the projection of the initial thin film configuration on the y-z plane was similarly defined, and the initial profile was smoothly defined in between. See Figs. S1–S2, Supplementary materials. Surface diffusion may be expressed as

$$V_n = -\mathscr{B}\,\triangle_s H, \tag{2.1}$$

where V_n is the normal velocity of the evolving exterior surface, H denotes its mean curvature, $\triangle_s$ is the Laplace - Beltrami operator (the surface Laplacian), and $\mathscr{B}$ is the Mullins' coefficient [18]. The boundary conditions are summarized in the Supplementary materials. Our model is fully nonlinear; no small slope approximation [18] is made.

3. A dimensionless parametric formulation of the model

To obtain a dimensionless formulation, we rescale all lengths in the system by $\bar{l} = l_z$, where l_z denotes the initial height of the thin film far from the contact line, and we rescale time by $\bar{t} = l_z^4/\mathscr{B}$. After introduction of the rescaled variables, the governing equations outlined above remain valid upon rescaling the Mullins' coefficient $\mathscr{B}$ to unity in Eq. (2.1).

In our numerical simulations, the evolving film surface is assumed to be describable as a regular oriented parametric 2D surface in 3D, namely as

$$X(\alpha,\beta,t) = (x(\alpha,\beta,t), y(\alpha,\beta,t), z(\alpha,\beta,t)), \qquad 0 \leq \alpha,\beta \leq 1, \qquad t \in [0,\tilde{t}]. \tag{3.1}$$

This assumption appears to be quite mild, if the time $\tilde{t}$ is chosen to precede any possible break down of the model due, say to break up of the film into pieces or hole nucleation. The mean curvature of the evolving surface, H, can be expressed explicitly via the parametric description given in Eq. (3.1), as can $\triangle_s$, the surface Laplacian, ∇_s, the surface gradient, and $\vec{n}$, the unit normal to the (oriented) evolving surface. This allows the normal velocity V_n to be expressed as $X_t \cdot \vec{n}$, and Eq. (2.1) can then be expressed in dimensionless parametric form as

$$X_t \cdot \vec{n} = -\triangle_s H, \quad 0 \leq \alpha,\beta \leq 1, \quad t \in [0,\tilde{t}]. \tag{3.2}$$

Noting that Eq. (3.2) prescribes only the normal motion of the evolving surface, we fix the tangential motion by requiring uniform grid spacings along the exterior surface. This condition may be formulated as

$$X_\alpha \cdot X_{\alpha\alpha} = 0, \quad X_\beta \cdot X_{\beta\beta} = 0, \quad 0 \leq \alpha,\beta \leq 1, \quad t \in [0,\tilde{t}]. \tag{3.3}$$

The angle and mass flux conditions described in Section 2 can be expressed by writing the various unit exterior normals to the thin film which are tangent along its intersection in terms of Eq. (3.1), and the persistence conditions stating that the film remains attached along the contact line, the bounding cylinder, and the x-z and y-z planes. The considerations outlined above yield a coupled system of equations, whose numerical solution is adapted from Derkach et al.[19] and outlined in the Supplementary materials. See also [14,20].

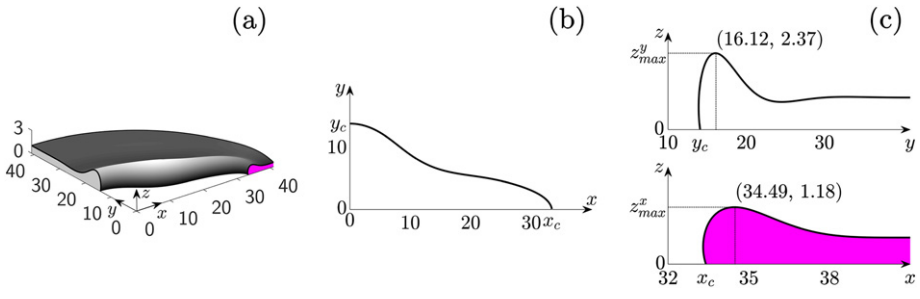

Fig. 1. In a), the profile of a thin dewetting film at (dimensionless) time $t = 222$ (see definition in Section 3) for an initially circular hole in a bi-crystalline system with a grain boundary (purple) in the x-z plane. Here $m = 0.3$, $\theta_c = 120^\circ$, and $x_c = y_c = 1$, $r_x - x_c = r_y - y_c = 40$, initially, see Section 2. In b), the contact line in the x-y plane; its intersection with the x-axis (y-axis) is marked as x_c (y_c). In c), the intersection of the thin film with the y-z and x-z planes, with grain boundary in purple. The maximum heights, $z^x_{\max}$, $z^y_{\max}$, are indicated. See Movie M1, Supplementary materials. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)