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Power-law scaling regimes for solid-state dewetting of thin films

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ABSTRACT

The capillary force drives the edges of solid thin films to retract. The distance a film edge has retracted over time is usually fitted to a power law. However, experiments and numerical simulations suggest that edge retraction does not follow a power-law. In this work, a simple geometric model for edge retraction is presented that reproduces the retraction distance *versus* time scalings of simulations for both isotropic and highly-anisotropic films, and is consistent with experiments. The earliest time at which a power-law fit becomes a reasonable approximation is calculated as a function of substrate–film contact angle.

Thin films are the fundamental building blocks for many micro- and nano-scale devices and systems. However, they are unstable against capillary forces due to their high surface-area-to-volume ratio. Capillarity (*i.e.*, surface tension) drives a process in thin films known as solid-state dewetting [1], which occurs in the solid state primarily *via* surface self-diffusion, though other transport mechanisms are possible [2].

The main feature of dewetting is retraction of the film's edges and the formation of thick "rims" of material along the retracting edges. The rim volume primarily comes from the volume of film that has been consumed by the retraction process [1,3]. Retraction is facilitated by a mass flux from the receding triple line (the intersection of the film/vapor, vapor/substrate, and substrate/film interfaces) towards the advancing side of the rim [4]. The flux from the bulk film towards the rim is extremely small, and may usually be neglected [5].

Brandon and Bradshaw developed a simple model for edge retraction that provides two important scaling laws [3]. First, the model predicts that the radius of a growing hole in a thin film will increase with time to the 2/5 power. Second, it predicts that the height of the rim will increase with time to the 1/5 power.

The B&B (Brandon and Bradshaw) model has two major limitations: first, it was developed for a contact angle of 90° only, and second, the cross-section of the rim was taken to be a semi-circle. Because of the first assumption, the effect of film–substrate contact angle on the scaling is unknown. The second assumption makes the model valid only in the limit of long retraction times, when the rim is much taller than the film height. Other phenomena such as pinch-off [6,7] or fingering instabilities [8,9,10] typically occur on thin film edges, which prevent the system from reaching the long-time limit of edge retraction in many cases.

Experiments do not agree with the scaling predicted by B&B [1]. For single-crystal nickel thin films, the exponent in the best power-law fit has been reported as 0.4 and 0.56 [11], and 0.38–0.43 $\pm$ 0.1 [12], for varying crystallographic directions. For single-crystal silicon, the exponent is reported to be 0.42–0.58 for different film thicknesses and orientations [13], and has been fitted by an exponent varying between 1/2 and 2/5 [14].

Numerical simulations of edge retraction also show that the retraction distance does not follow a power law. Both isotropic and fully-faceted films initially retract linearly in time, then the exponent in the power law gradually decreases, approaching 2/5 in the long-time limit [7,4]. Kinetic Monte-Carlo simulations give an initial retraction rate proportional to $t^{1/2}$, and approaching $t^{2/5}$ in the long-time limit [14].

In this work, we identify the underlying physics describing edge retraction which are consistent with experiments and numerical simulations. We present an analytical model, based on Brandon and Bradshaw's approach, which overcomes the limitations of the original model. Our model captures the transition from linear retraction to 2/5 power-law behavior and offers a physical explanation for this phenomenology. The retraction rate and earliest time for $t^{2/5}$ retraction are also provided as a function of film–substrate contact angle.

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The velocity of an isotropic surface evolving by capillary-driven surface diffusion was given by Mullins as [2]

$$u_n = B\left(\frac{\partial^2 K}{\partial\sigma_1^2} + \frac{\partial^2 K}{\partial\sigma_2^2}\right), \tag{1}$$

where $B = \frac{D_s\gamma\Omega^2\nu}{kT}$, D_s is the surface self-diffusivity, γ is the surface energy, Ω is the volume per atom, ν is the density of mobile surface atoms, k is Boltzmann's constant, T is temperature, and K is the mean curvature of the surface as a function of orthogonal arc length coordinates σ_1 and σ_2. This equation can be made dimensionless to have the form

$$v_n = \left(\frac{\partial^2\kappa}{\partial s_1^2} + \frac{\partial^2\kappa}{\partial s_2^2}\right), \tag{2}$$

where v_n is the dimensionless velocity and $v_n = u_n H^4/B$, H is the film thickness, s_1 and s_2 are the dimensionless arc length coordinates, $s_i = \sigma_i/H$, and κ is the dimensionless mean curvature, $\kappa = HK$.

The following five assumptions made in the B&B model are preserved in our model to simplify the rim geometry: *i*) The film is taken to be isotropic. *ii*) The film edge profile is identical everywhere along the triple line, so there is no dependence on the arc length coordinate parallel to the triple line s_2, and it can be ignored. *iii*) When the film is cross-sectioned normal to the triple line, the rim profile is a circular arc. *iv*) The film behind the rim has uniform thickness, *i.e.*, there is no valley ahead of the retracting rim. The discontinuity in slope where the rim meets the film is artificial and is therefore ignored; and *v*) There is no mass flow between the flat film and the rim.

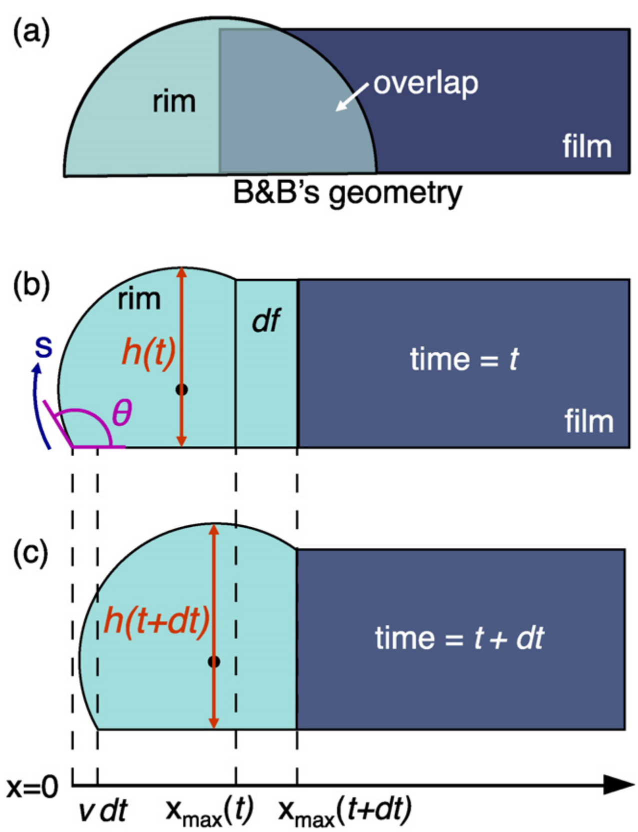

Fig. 1. (a) The geometry assumed by Brandon and Bradshaw [3] treats the rim as a semi-circle, with overlap between the film and rim, violating mass conservation. (b, c) The cross-sectional profile of the edge of the film is shown at time = t (b), and at time = $t+dt$ (c). It is assumed that retraction proceeds at velocity v, which is a function of the rim height $h(t)$, for a short amount of time dt. The new film edge geometry can be found by assuming that the new rim area (light shading in (c)) is the sum of the old rim area plus the area df (light shading in (b)). The x-axis is drawn below the figures, and the positions used in the model are indicated. All length scales are normalized to the film thickness, and the contact angle θ and arc length coordinate s are shown in (b).

To overcome the limitations of the original B&B model, we introduce two augmentations: we allow any contact angle, and perform a more accurate treatment of the rim volume over time. While B&B take the cross-section of the rim to be a semi-circle, here it is treated as a circle that is cut along two perpendicular chords (see Fig. 1): the horizontal cut ensures that the rim meets the substrate at the equilibrium contact angle θ (which is no longer constrained to be 90°), and the vertical cut ensures a flush match between the rim and bulk film, so that volume is conserved (which was not the case in the original B&B model).

All lengths in this analysis are normalized to the film thickness H, and time is normalized to H^4B^{-1}, so that all quantities are dimensionless. The height of the rim, h, is related to the radius of curvature of the rim, r, and the contact angle, θ, by

$$r = \frac{h}{1-\cos\theta}. \tag{3}$$

To compute the velocity of surface motion using Eq. (2), the second derivative of curvature along the film profile is needed. We assume that the curvature as a function of arc length is parabolic near the triple line, *i.e.*, we use a second-order accurate approximation of the curvature, similar to B&B's and Danielson's approach [3,13]. In general, the second derivative of a parabola, $k(s)$, of best fit to three distinct points (s_i, k_i) is

$$\frac{\partial^2 k}{\partial s^2} \approx \frac{2(s_1k_2 + s_2k_3 + s_3k_1 - s_1k_3 - s_2k_1 - s_3k_2)}{(s_1-s_2)(s_2-s_3)(s_3-s_1)}. \tag{4}$$

Three points are selected along the s coordinate, $(s_i, k_i) = (0, k_{s=0})$, $(\Delta s, k_{s=\Delta s})$, $(2\Delta s, k_{s=2\Delta s})$, where Δs is taken to be the arc length from the triple line to where the rim meets the bulk film,

$$\Delta s = r\left(\theta + \arcsin\frac{x_{max}(t) - r\sin\theta}{r}\right), \tag{5}$$

and the value of $x_{\max}(t)$ is indicated in Fig. 1. Note that Δs is not an infinitesimal quantity, and changes with time. The curvature at the triple line, $k_{s=0}$, is equal to the curvature of the rim, $1/r$. At arc distance Δs and $2\Delta s$ from the triple line, the curvature ($k_{s=\Delta s}$ and $k_{s=2\Delta s}$) is that of the flat film, 0. Substitution into Eq. (2) using Eqs. (3) and (5), and projecting the normal motion into the plane of the substrate (*i.e.*, dividing by $\sin\theta$), yields

$$v_{\text{retr}} = \frac{\csc\theta\cos\theta - 1^3}{h^3\left(\theta + \arcsin\left(\frac{1}{h}\sin\left(\frac{\theta}{2}\right)\sqrt{2(h-1)(1+h+(h-1)\cos\theta)}\right)\right)^2}. \tag{6}$$

The rim height at a future time, $h(t+dt)$, is computed using conservation of mass within the rim. The old rim, with height $h(t)$, will incorporate material from the flat film with a cross-sectional area df, as illustrated in Fig. 1. The cross-sectional area of the rim is found by integrating the curve that describes it from $x_{\min}$ to $x_{\max}$,

$$\text{rim profile}(x) = \sqrt{r^2 - (r\sin\theta + x_{min} - x)^2} - r\cos\theta, \tag{7}$$

where $x_{\min} = 0$ at time t and $x_{\min} = v_{retr}dt$ at time $t+dt$. The additional volume in the rim where $x < x_{\min}$ if $\theta > 90^\circ$ is also integrated and added. The area of flat film that is incorporated into the rim, df, is simply $(x_{\max}(t+dt) - x_{\max}(t))$ (the film thickness is 1).

The cross-sectional area of the film is conserved, giving the equation

$$\text{rim area}(t+dt) - \text{rim area}(t) - df = 0. \tag{8}$$