Inhibition of Rayleigh-Plateau instability on a unidirectionally patterned substrate

G. Boussinot[1] and Efim A. Brener[2]

[1]*Access e.V., Intzestr. 5, 52072 Aachen, Germany*
[2]*Peter Grünberg Institut, Forschungszentrum Jülich, D-52425 Jülich, Germany*

A fundamental process of surface energy minimization is the decay of a wire into separate droplets initiated by the Rayleigh-Plateau instability. Here we study the linear stability of a wire deposited on a unidirectionally patterned substrate with the wire being aligned with the pattern. We show that the wire is stable when a criterion that involves its width and the local geometry of the substrate at the triple line is fulfilled. We present this criterion for an arbitrary shape of the substrate and then give explicit examples. Our result is rationalized using a correspondence between the Rayleigh-Plateau instability and the spinodal decomposition. This work provides a theoretical tool for an appropriate design of the substrate's pattern in order to achieve stable wires of, in principle, arbitrary widths.

I. INTRODUCTION

Recently, the deposition of technologically relevant materials on solid substrates that are patterned at the microscale or the nanoscale has received a large attention. The inhomogeneity of the substrate allows the deposited material to present interesting properties such as a reduced dislocation density [1], ordered arrays of emitting quantum dots or wires [2,3] or of monodispersed metallic particles [4], tunable adsorption isotherms in the capillary condensation regime [5–7], microchannels decorated with a wider and thicker bulged region [8], superhydrophobicity or superhydrophilicity [9]. These properties may be apprehended by the study of the wetting behavior of the deposited material, which is mainly a consequence of the minimization of surface energy at the length scales of interest.

The decomposition of a rod into droplets is probably one of the most fundamental dynamical processes of surface energy minimization. In opposition to a metastable flat film, decaying through the nucleation of dry patches (holes), the rod is linearly unstable and decays through the so-called Rayleigh-Plateau instability. After the experimental observations of Plateau [10], Rayleigh described the instability of a rotationally invariant liquid jet through a theory for low-viscosity fluids [11]. Since these seminal works, a huge amount of literature on the breakup of a liquid rod has appeared and we refer to Ref. [12] for a review.

In an equivalent manner, the decay of a solid rod into droplets may also occur. While for the liquid the transport mechanism is the fluid flow, it is commonly accepted that the evolution of the surface of a solid surrounded by the vapor below the melting temperature is driven by surface diffusion. In this respect, the evolution of a cylindrical rod by surface diffusion was discussed theoretically by Nichols and Mullins [13]. Since the original work by Mullins [14] on surface diffusion, the technology related to the deposition of a solid on a solid substrate has emerged and pattern formation processes such as Asaro-Tiller-Grinfeld instability, quantum dots formation, or dewetting dynamics are commonly described using Mullins' equation for surface diffusion [15–19]. Closer to our current interest, the decomposition of solid Cu or Au nanowires into droplets has been reported [20,21] and

explained using the Nichols-Mullins theory. However, the latter theory does not take into account the contact angle that exists at the deposited solid-substrate-vapor triple line. In Ref. [22], McCallum *et al.* studied the influence of a flat substrate on the Rayleigh-Plateau instability of a solid wire evolving via surface diffusion. Although the substrate is found to have a stabilizing effect, i.e., restricting the range of unstable wavelengths, the wire is found to be linearly unstable.

In this article, we consider a wire lying on a unidirectionally patterned substrate and being aligned with the pattern. We perturb the wire with sinusoidal modulations of its height along its axis (varicose). Using the surface diffusion equation, we show that the wire may be linearly stable. More precisely, we search for the neutral mode that yields vanishing surface diffusion fluxes (the mode that neither grows nor decays), providing a critical wavelength above which the wire is linearly unstable. For some condition that involves the width of the wire and the local geometry of the substrate at the triple line, no solution is found for the neutral mode and the wire is then linearly stable.

The equilibrium and stability of the deposited material on a patterned substrate was studied theoretically some time ago on a general level (see Refs. [23,24] and for a recent review [25]). Here, the unidirectional pattern of the substrate that we consider simplifies the problem drastically, allowing us to explicitly assess the influence on the stability of the wire of the few parameters describing the substrate's pattern. The linear stability of the wire is related to the capillary filling phenomenon [9] or to the above-mentioned capillary condensation. Indeed, when the contact angle at the triple line is smaller than $\pi/2$, the surface energy of the wire-substrate interface is effectively negative and the system may efficiently decrease its surface energy by a spreading of the deposited material on the patterned substrate.

After describing the unperturbed wire in Sec. II A, we perform the linear stability analysis of the perturbed wire in Sec. II B and find the stability criterion for an arbitrary pattern of the substrate. In Sec. III, we illustrate our theory using explicit examples for the pattern. In Sec. IV, we discuss our results using energetic arguments and finally conclude in Sec. V.

II. MODEL

A. Equilibrium state

We consider a substrate whose height is a function that does not depend on one of the two coordinates in the (x,y) plane, say y. We thus write the height of the substrate $h_s(x)$. Moreover, we assume that the substrate's shape is symmetric with respect to $x = 0$, i.e., $h_s(-x) = h_s(x)$. A symmetric equilibrium wire that possesses a curvature only in the x direction is then represented by a height

$$h_{\text{eq}}(x) = r - x^2/(2\rho). \tag{1}$$

The two quantities r and ρ are related to the position of the triple line $x_0 > 0$ by the two equations

$$h_{\text{eq}}(x_0) = h_s(x_0),$$
$$h'_{\text{eq}}(x_0) = h'_s(x_0) - \varphi,$$

which describe the contact position and the contact angle, respectively (see Fig. 1). This yields

$$r = x_0^2/(2\rho) + h_s(x_0), \tag{2}$$

$$-x_0/\rho = h'_s(x_0) - \varphi. \tag{3}$$

In this description of the equilibrium wire, we have used a small slope approximation that relies on the assumption of a small contact angle $\varphi \ll 1$. The circular shape that holds for an arbitrary contact angle is thus replaced by a parabolic one. This avoids the consideration of different cases due to a square root $\sqrt{\rho^2 - x^2}$ in the function describing the height for the circular shape. In particular, it allows us to have one single set of formulas [Eqs. (2) and (3)] valid for arbitrary x_0. Moreover, we assume isotropic wire-vapor, wire-substrate, and substrate-vapor surface energies in order to highlight the role of the substrate's pattern.

B. Linear stability analysis

Depending on x_0, this one-dimensional wire may be in an unstable equilibrium state. For the unstable situation, surface diffusion at the wire-vapor interface is assumed to allow for a decrease in the surface energy of the system through sinusoidal modulations of the wire's height along the y direction (varicose perturbation modes). The axis of symmetry of the unperturbed wire remains in this case the symmetry axis of the perturbed
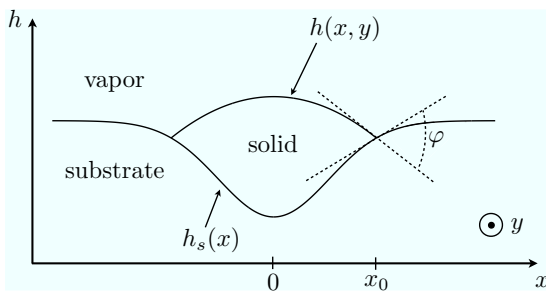

FIG. 1. Cross section of the wire on the unidirectionally patterned substrate. At the triple line $x = x_0$, the wire presents the equilibrium contact angle φ.

wire. The height of the wire is now a function of x and y, and is written as

$$h(x,y) = h_{\text{eq}}(x) + \delta h(x,y), \tag{4}$$

where

$$\delta h(x,y) = \epsilon \cos(ky) f(x). \tag{5}$$

Here k is the wave number of the perturbation, ϵ is the amplitude of the perturbation that will be kept only to the first power in the linear stability analysis, and $f(x) = f(-x)$ is a symmetric function that has to be determined. Neglecting bulk diffusion (see the discussion in Ref. [14]), the surface diffusion mechanism leads to a normal velocity of the wire-vapor interface $\dot{h}$:

$$\dot{h} = -D\nabla^2\nabla^2 h, \tag{6}$$

where D is the surface diffusion coefficient (m^4 s^{-1}) [14]. We are searching for the wave number k^* for which the perturbation neither grows nor decays, i.e., for which $\dot{h} = 0$. This means that the perturbation itself obeys the bi-Laplacian equation

$$\nabla^2\nabla^2\delta h(x,y) = 0. \tag{7}$$

In addition, since we assume surface diffusion only at the wire-vapor interface, the conservation of mass in the system implies a no-flux boundary condition at the triple line

$$\frac{\partial}{\partial x}\left(\frac{\partial^2 h}{\partial x^2} + \frac{\partial^2 h}{\partial y^2}\right) = 0, \tag{8}$$

yielding

$$f'''(x_0) - k^2 f'(x_0) = 0. \tag{9}$$

The symmetric function $f(x)$ that allows Eq. (7) to be fulfilled and that obeys the boundary condition (9) is

$$f(x) = \cosh(kx). \tag{10}$$

The critical wave number k^* is then determined using the boundary conditions for the contact with the substrate and the contact angle at the new position of the triple line $x_0 + \delta x(y)$, where $\delta x \sim \epsilon \cos(ky)$:

$$h(x_0 + \delta x, y) = h_s(x_0 + \delta x), \tag{11}$$

$$\partial_x h(x_0 + \delta x, y) = h'_s(x_0 + \delta x) - \varphi. \tag{12}$$

Here, we have neglected the triple line tension that may correct the contact angle equation at large curvatures of the triple line [26]. Together with Eqs. (1) and (3), the last two equations yield

$$x_0 f'(x_0)/f(x_0) = A_s(x_0), \tag{13}$$

where

$$A_s(x_0) = 1 + x_0 h''_s(x_0)/\varphi - h'_s(x_0)/\varphi. \tag{14}$$

The critical wave number k^* thus obeys

$$F(k^* x_0) = A_s(x_0), \tag{15}$$

where

$$F(z) = z\tanh(z). \tag{16}$$

$F(z)$ is a positive and even function. It is parabolic near the origin, i.e., $F(z) \simeq z^2$ for $|z| \ll 1$, and it rapidly reaches the linear behavior $F(z) \simeq |z|$ for $|z| \gg 1$. Therefore k^* exists only if $A_s(x_0) > 0$. Then, the wire is unstable for $k < k^*$ and stable for $k > k^*$. Let us note that when $0 < A_s(x_0) \ll 1$, we have $k^*x_0 \ll 1$, i.e., the unstable wavelengths of the wire are much larger than its width. On the other hand, when $A_s(x_0) < 0$, k^* does not exist and the wire is stable for all k. This stability is entirely due to the pattern of the substrate. Especially, it is not due to any anisotropy of surface energy, a property that may indeed have a stabilizing influence [27,28]. Furthermore, the stability of the wire depends only on its width x_0 and on the local geometry of the substrate at the triple line through $h'_s(x_0)$ and $h''_s(x_0)$. This demonstrates the possibility of the deposition of stable wires using an appropriate design of the substrate's pattern.

A few remarks are in order. First, let us note that the antisymmetric modes [for which one replaces cosh by sinh in Eq. (10)] are more stable than the symmetric ones and are thus irrelevant here. Second, the conclusions drawn from the above analysis could as well be performed by an energetic calculation. Indeed, for $k < k^*$, the energy per unit volume of the perturbed wire is smaller than the energy per unit volume of the unperturbed one, the latter being then linearly unstable. Conversely, for $k > k^*$, it is larger and the unperturbed wire is linearly stable. Finally, we would like to mention that our analysis reproduces the known results for a flat substrate. Indeed, setting $A_s(x_0) = 1$, we recover $k^*x_0 \simeq 1.200$, which is found as the small contact angle limit of the theory presented in Ref. [22].

III. EXAMPLES

Let us illustrate the behavior of $A_s(x_0)$ for a few given substrate shapes.

A. Gaussian groove

First, let us consider a Gaussian groove with

$$h_s(x) = -h_0 \exp[-x^2/(2\sigma^2)]. \quad (17)$$

Inserting h_s into Eq. (14) we obtain

$$A_s(x_0) = 1 - \left[h_0 x_0^3/(\varphi\sigma^4)\right]\exp\left[-x_0^2/(2\sigma^2)\right] < 1.$$

For $h_0/(\varphi\sigma) > (e/3)^{3/2}$, A_s is negative in some range around $x_0 = \sqrt{3}\sigma$ and the wire is stable in this range. We illustrate this in Fig. 2(a) by plotting $A_s(x_0)$ as a function of x_0/σ for $h_0/(\varphi\sigma) = 0.5$ and $h_0/(\varphi\sigma) = 1.5$. For $h_0/(\varphi\sigma) = 0.5$, A_s is positive for all x_0 and therefore the wire is unstable for all x_0. For $h_0/(\varphi\sigma) = 1.5$, the situation is different and the wire is stable in the range of x_0 (represented by the horizontal arrow) where $A_s < 0$.

B. Sinusoidal substrate

Second, let us consider a substrate with a sinusoidal pattern:

$$h_s(x) = h_0 \cos(k_s x).$$

Using this substrate shape in Eq. (14) gives

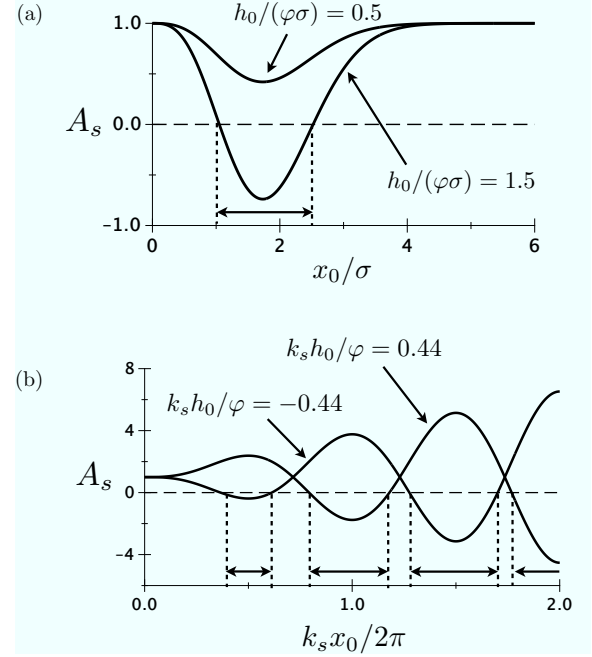$$A_s(x_0) = 1 + (k_s h_0/\varphi)[\sin(k_s x_0) - k_s x_0 \cos(k_s x_0)].$$

FIG. 2. Function A_s as a function of (a) x_0/σ for a Gaussian groove of depth h_0 and (b) $k_s x_0$ for a sinusoidal pattern of amplitude $|h_0|$ (the sign of h_0 describes the possibility for the wire to be centered either on the top of a hill or on the bottom of a valley of the substrate; see text for precisions). The values of x_0 for which A_s is negative correspond to stable situations for the wire. The corresponding ranges are represented by horizontal arrows.

The equilibrium state of the wire may be centered on the top of a hill ($h_0 > 0$) or on the bottom of a valley ($h_0 < 0$). The function $A_s(x_0)$ is an oscillating function whose amplitude increases with x_0. In Fig. 2(b), we present $A_s(x_0)$ as a function of $k_s x_0/2\pi$ for $k_s h_0/\varphi = 0.44$ and $k_s h_0/\varphi = -0.44$. Alternatively, the valley-centered and the hill-centered wires present an infinite succession of bands of x_0 where $A_s < 0$, i.e., where they are stable. An interesting effect following from our analysis concerns the fact that for an arbitrarily small amplitude h_0, there exist bands of stable x_0 as soon as $k_s x_0 \gg \varphi/(k_s h_0)$. Besides this interesting result, two remarks are in order for this case of a sinusoidal pattern. First, there is an upper bound on the amplitude h_0 in order for our analysis to remain correct at arbitrary x_0. For a given x_0, we indeed have $h_{eq}(x) > h_s(x)$ for all $x < x_0$ only if h_0 is small enough. A sufficient (but not necessary) condition for this is $k_s|h_0|/\varphi < 1/2$ [note the choice of h_0 in Fig. 2(b)]. Second, it should be mentioned that the alternation of stable bands for the valley-centered wire and the hill-centered one has energetic reasons. Indeed, it can be shown that for a given area S of the cross section of the wire, the two equilibrium states (valley- or hill-centered) do not have the same surface energy. Then, when one increases S, the minimum of energy changes from one equilibrium state to the other alternatively. However, a precise description of this phenomenon is beyond the scope of this paper, and here we just aim at showing that for a sinusoidal pattern, the wire possesses an infinite succession of bands of stable widths x_0, whatever the amplitude h_0 of the sinusoid.

C. V-shaped substrate

We focus now on a V-shaped pattern of the substrate, i.e.,

$$h_s(x) = \varphi_0 |x| \tag{18}$$

with $\varphi_0 > 0$. This gives, using Eq. (14),

$$A_s(x_0) = 1 - \varphi_0/\varphi. \tag{19}$$

The wire is thus stable, i.e., $A_s < 0$, if $\varphi_0/\varphi > 1$. We recover here the usual criterion for the well-known capillary filling phenomenon or the capillary condensation for which the material is adsorbed on the substrate although it is not stable thermodynamically ($\rho < 0$).

We would like also to mention the case where the V shape ends with a sharp edge at $|x| = x_s > 0$, i.e.,

$$h_s(|x| < x_s) = \varphi_0(|x| - x_s), \quad h_s(|x| > x_s) = 0, \tag{20}$$

with $\varphi_0 > 0$. At $x = x_s$, h_s' possesses a discontinuity and we may regularize it with the help of a microscopic length scale $a \ll \varphi_0 x_s$, which yields $h_s''(x_s) = -\varphi_0/a$. This implies that

$$A_s(x_s) = 1 - \varphi_0/\varphi - \varphi_0 x_s/(\varphi a) < 0, \tag{21}$$

and $-A_s(x_s) \gg 1$. We thus find here that when the triple line is in the so-called pinned state (when the contact angle is not properly defined without the introduction of the microscopic length scale a) the wire is stable against the Rayleigh-Plateau instability.

D. Chemically patterned substrate

One may, in principle, introduce a space-dependent contact angle $\varphi(x)$, which is typical for the so-called chemically patterned substrates. Then A_s possesses an additional contribution $-x_0 \varphi'(x_0)/\varphi(x_0)$, i.e.,

$$A_s(x_0) = 1 + \frac{x_0[h_s''(x_0) - \varphi'(x_0)]}{\varphi(x_0)} - \frac{h_s'(x_0)}{\varphi(x_0)}. \tag{22}$$

Thus, when one introduces a stripe with $\varphi(|x| < x_s) = \varphi_1$ on a more hydrophobic flat substrate with $\varphi(|x| > x_s) = \varphi_2 > \varphi_1$, then $\varphi'(x_s)$ is positive and large (the same regularization as above is possible) and thus the wire is stable when the triple line is pinned at x_s, i.e., $A_s(x_s) < 0$. This is precisely what was observed in the experiments presented in Ref. [8] before the transition to a bulged state.

IV. DISCUSSION

A. Energetics

We now analyze the Rayleigh-Plateau stability criterion $A_s(x_0) < 0$ where A_s is given by Eq. (14) using energetic arguments. The surface energy per unit length of the equilibrium wire $E(x_0)$ consists of two contributions:

$$E(x_0) = \gamma_1 L_1(x_0) + \gamma_2 L_2(x_0), \tag{23}$$

where L_1 and L_2 are the lengths of the wire-vapor and wire-substrate interfaces, respectively, given by

$$L_1(x_0) = 2 \int_0^{x_0} dx [1 + h_{\text{eq}}'^2(x)/2], \tag{24}$$

and

$$L_2(x_0) = 2 \int_0^{x_0} dx [1 + h_s'^2(x)/2]. \tag{25}$$

γ_1 is the energy density of the wire-vapor interface and γ_2 is the difference between the energy densities of the wire-substrate and substrate-vapor interfaces (γ_2 is negative for $\varphi < \pi/2$). According to Young's law, we have $\gamma_2 = -\gamma_1 \cos \varphi \simeq -(1 - \varphi^2/2)\gamma_1$. We may also calculate the cross section area S of the wire given by

$$S(x_0) = 2 \int_0^{x_0} dx [h_{\text{eq}}(x) - h_s(x)]. \tag{26}$$

One may then show that

$$dE/dS = (dE/dx_0)(dS/dx_0)^{-1} = \gamma_1/\rho, \tag{27}$$

and since

$$d^2E/dS^2 = \gamma_1(dS/dx_0)^{-1}d(1/\rho)/dx_0, \tag{28}$$

that

$$\frac{d^2E}{dS^2} = -\frac{\gamma_1}{2x_0^3} \frac{A_s(x_0)}{1 - A_s(x_0)/3}. \tag{29}$$

The transition that we describe here from an unstable wire ($A_s > 0$) to a stable wire ($A_s < 0$) thus corresponds to a change of sign of d^2E/dS^2. More precisely, our analysis reproduces the usual criterion: if the second derivative of the energy with respect to the conserved quantity is negative ($A_s > 0$), spatial fluctuations are amplified (spinodal decomposition), while if it is positive ($A_s < 0$) the fluctuations are damped.

B. Ground state

Let us now make a remark that concerns the ground state of the system. An obvious attractor for the late stages of the temporal evolution that follows the spinodal decomposition is a coarsened state that develops through an Ostwald ripening regime. Here it means that after the Rayleigh-Plateau instability, the wire breaks up into separate droplets, and the subsequent evolution consists of the growth of the large droplets at the expense of the small ones, leaving at the end a single coarse droplet. If the wire is stable against Rayleigh-Plateau instability, the ground state of the system is determined by the comparison of the energy of the wire and the one of the coarsened state. At this point, one should note that while the wire extends indefinitely in the y direction, the coarse droplet is localized. On the one hand, the wire's energy per unit volume is its energy per unit length E divided by its cross section area S. On the other hand, if R is the linear dimension of the coarse droplet, its energy scales as R^2, its volume as R^3, and its energy per unit volume as $1/R$. Thus for infinite volumes the energy per unit volume of the coarse droplet vanishes. Therefore when the energy per unit length E of the wire is positive, the latter is metastable and thermally activated finite amplitude fluctuations will cause its decay into droplets (nucleation) with a subsequent Ostwald regime. Conversely, when E is negative, the coarse droplet is not the ground state of the system. However, a bit of care should be taken in order to apprehend the ground state in this

case. Let us consider a linear groove, for example, the Gaussian groove $h_s(x) = -h_0 \exp[-x^2/(2\sigma^2)]$. For large enough h_0, E presents a maximum at some x_0 and a minimum at a larger x_0 (these two extrema verify $dE/dS = \gamma_1/\rho = 0$). Then above a critical h_0, the value $E_{\min}$ of E at this minimum is negative. Let us denote by $S_{\min}$ the cross-section area of this wire having the minimum energy per unit length $E_{\min}$. This wire is stable with $A_s < 0$ and corresponds to the ground state of the system when the volume of deposited material per unit length of the substrate is $S_{\min}$. A wire with S slightly larger than $S_{\min}$ remains stable against the Rayleigh-Plateau mechanism, its energy per unit length E is larger than $E_{\min}$, and its energy per unit volume is E/S. Let us now consider a system with a volume of deposited material per unit length of the substrate being also S and consisting of a wire with $S_{\min}$ coexisting with a coarse droplet. Due to the argument given above concerning the energetics of a coarse droplet, the energy per unit volume of this system is $E_{\min}/S < E/S$. Therefore the configuration with a wire of cross-section area $S_{\min}$ coexisting with a coarse droplet represents the ground state of the system when the volume of deposited material per unit length of the substrate is larger than $S_{\min}$.

This transition from a ground state presenting a single wire to a ground state presenting a wire coexisting with a coarse droplet is actually an interesting point to investigate further. Here, this transition is described for infinite volumes, i.e., for an infinitely long wire and an infinitely large droplet. For a finite length of the wire, it is believed that the transition should occur in a rather similar way as the bifurcation toward a bulged state presented in Ref. [8]. All these scenarios where the deposited material tends to spread on the substrate due to energetic reasons correspond to the so-called hemiwicking phenomenon [9] (in opposition to simple wicking that describes the spreading in a pore and for which no interface with the vapor is produced).

Let us finally note that in the case of a wire of finite length, another mechanism for the decay of the wire into droplets may exist. Indeed, the end of the wire is subjected to a retraction that favors necking, and a so-called edge-driven instability was evidenced for a nickel wire on a flat substrate in Ref. [29]. Investigating this issue for the type of patterned substrate that is studied in the present work is also an interesting perspective.

V. CONCLUSION

We have studied analytically the linear stability against the Rayleigh-Plateau mechanism of a wire aligned with the unidirectional pattern of a substrate. We use the neutral modes for surface diffusion (the eigenmodes that make the surface diffusion fluxes vanish) to find a criterion for stability. This criterion involves the wire's width and the local geometry of the substrate at the triple line. This criterion is found for an arbitrary shape of the substrate and we illustrate our analysis using explicit examples for the substrate's pattern. In a Gaussian groove, there exists a range of stable widths of the wire for large enough depth-to-width ratios of the groove. For a groove presenting a sharp edge, we show that the wire is stable at widths where the triple line is pinned and discuss the link to the experiment in Ref. [8]. For a sinusoidal pattern, there exists an infinite succession of bands of stable widths. We finally show that energetic arguments allow an analogy between the Rayleigh-Plateau instability described here and the spinodal decomposition process.

Our results shed light on the relation between the width of the wire and the geometry of the substrate in order to achieve stable configurations. In principle this relation allows one to produce stable wires of arbitrary width by an appropriate design of the substrate's pattern.

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