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# Nonlinear evolution of a morphological instability in a strained epitaxial film

Jean-Noël Aqua,\* Thomas Frisch,† and Alberto Verga‡

*Institut de Recherche sur les Phénomènes Hors Équilibre, UMR 6594, Aix-Marseille Université, France*

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A strained epitaxial film deposited on a deformable substrate undergoes a morphological instability relaxing the elastic energy by surface diffusion. The nonlinear dynamical equation of such films with wetting interactions are derived and solved numerically in two and three dimensions. Nonlocal nonlinearities together with wetting effects are crucial to regularize the instability leading to an island morphology. The island chemical potential decreases with its volume and the system consistently experiences a non-interrupted coarsening evolution described by power laws with a marked dimension dependence.

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The evolution of semiconductor thin films is under active scrutiny due to its importance for both fundamental science and technological applications. Indeed, different instabilities lead to self-organized nanostructures<sup>1</sup> potentially useful e.g. for quantum dots, wires or specifically confining electronic devices.<sup>2</sup> A notorious experimental example is Si/Ge films on Si substrates which exhibit a variety of structures such as pre-pyramids, pyramids, domes and huts.<sup>3,4</sup> However, the basic mechanisms ruling the evolution of heteroepitaxial films are not fully apprehended and are still under debate.<sup>5,6,7</sup>

Heteroepitaxial films experience an elastic stress due to the film-substrate misfit which is relaxed by surface diffusion during annealing. The resulting morphological instability driven by the interplay between elastic and isotropic surface energies<sup>8</sup> is similar to the Asaro-Tiller-Grinfeld (ATG) instability in solid-liquid interfaces,<sup>9</sup> observed in helium and various solid interfaces.<sup>10,11</sup> Linear analysis leads to exponentially growing long wavelength perturbations either for an infinitely thick film<sup>8</sup> or a film on an elastic<sup>12</sup> or rigid<sup>13</sup> substrate. The late stage evolution given by the first elastic nonlinearities (arising from geometry within linear elasticity) then reveals finite-time singularities<sup>13,14,15</sup> enforced by elastic stress concentration at cusps. The latter account well for experiments in thick films<sup>10</sup> where dislocations can finally occur but not for thin films in the Stranski-Krastanov mode where islands separated by a wetting layer coarsen under annealing.<sup>3,4</sup>

To describe more precisely these systems, the effects of a wetting potential  $\mu_w$  between the film and the substrate was considered<sup>16</sup> and discloses a critical film thickness above which the instability grows.<sup>16</sup> Island equilibrium shapes could then be derived thanks to the existence of a wetting layer.<sup>17</sup> Thence, the dynamical behavior of thin wetting films was investigated under different frameworks. With a regularizing wetting potential which diverges as  $1/h^2$  for small film height  $h$  and a linear in  $h$  elastic energy, steady-state solutions were displayed in Ref. 5 whereas a power-law coarsening evolution was recently obtained using numerical simulations.<sup>7</sup> Using a non-diverging wetting potential, which is however still leading at small- $h$ , it was found in Ref. 6 that elas-

tic nonlinearities enforce non-coarsening islands whereas finite-time singularities appear when truncating the elastic energy at linear order.<sup>6</sup> Moreover, considering a regular  $\mu_w$  but an infinitely rigid substrate, nonlinear blow-up solutions arose again in Ref. 18 unless strong enough anisotropy is accounted for. Finally, island coarsening was shown to terminate in Ref. 6 whereas power-law dynamics was found in Ref. 7. In the present article we clarify the debate and show that a consistent description of the morphological instability evolution arises when the nonlinear relaxation of the elastic energy is included together with a regular wetting potential, e.g. exponentially decaying as first introduced in Ref. 16 and consistent<sup>19</sup> with ab initio calculations.<sup>20</sup> We show that the combination of elastic nonlinearities and even *small* wetting effects regularizes the ATG singularity and leads to regularly evolving islands. The resulting fully nonlinear film dynamics is then characterized by a non-interrupted coarsening with power-law exponents different from Ref. 7 for the surface roughness and number of islands.

We consider specifically a three-dimensional (3D) dislocation free film deposited on a substrate with slightly different lattice parameters and a priori different isotropic elastic properties. During annealing, the film shape  $h(x, y, t)$  evolves by surface diffusion (no external flux nor evaporation). The boundary at  $z = h(x, y, t)$  is free while the film-substrate interface at  $z = 0$  is coherent. In the reference state, the film is flat and the elastic energy density is  $\mathcal{E}^0 = E_f (a_f - a_s)^2 / a_s^2 (1 - \nu_f)$  where  $a_\alpha$ ,  $E_\alpha$  and  $\nu_\alpha$  are the lattice parameter, Young modulus and Poisson ratio of the solid  $\alpha$  with  $\alpha = f$  for the film and  $s$  for the substrate. The film dynamics is then given by,<sup>8</sup>

$$\frac{\partial h}{\partial t} = D \sqrt{1 + |\nabla h|^2} \nabla_S^2 \mu, \quad (1)$$

with  $D$ , a constant related to surface diffusion, and  $\nabla_S$ , the surface gradient. Both elastic  $\mathcal{F}^{\text{El}}$  and surface  $\mathcal{F}^{\text{S}} = \int d\mathbf{r} \gamma(h) \sqrt{1 + |\nabla h|^2}$  free energies contribute to the surface chemical potential  $\mu = \delta(\mathcal{F}^{\text{El}} + \mathcal{F}^{\text{S}}) / \delta h$  reading

$$\mu = \mathcal{E}[h] + \gamma(h) \kappa(h) + \gamma'(h) / \sqrt{1 + |\nabla h|^2}, \quad (2)$$

with  $\mathcal{E}[h]$ , the elastic energy density computed at  $z =$

$h(x, y, t)$ ,  $\gamma$ , the isotropic surface energy and  $\kappa$ , the free surface mean curvature, see e.g. Ref. 5. To account for wetting,<sup>16</sup> the surface energy  $\gamma$  is supposed to be a function of  $h$  (so that  $\mu_w = \gamma'(h)$ ), extrapolating from the bulk value  $\gamma_f$  when  $h \rightarrow \infty$ , to some upper value when  $h \rightarrow 0$ . We write  $\gamma(h) = \gamma_f [1 + c_w f(h/\delta)]$  with the characteristic length  $\delta$ , the strength  $c_w > 0$  and some function  $f$  going to zero at infinity. In the following, we use  $f(\xi) = \exp(-\xi)$  when a specific form is needed in order to mimic ab initio calculations.<sup>20</sup> Finally, we set the length unit as  $l_0 = \mathcal{E}^0/\gamma_f$ , the characteristic length of the instability with the corresponding time unit  $t_0 = l_0^4/D\gamma_f$ .

To compute elastic energies, we use the isotropic continuum framework where stresses  $\sigma_{pq}^\alpha$  are proportional to strains  $e_{pq}^\alpha$  in the solid  $\alpha$ ,

$$\sigma_{pq}^\alpha = \frac{E^\alpha}{1 + \nu^\alpha} \left[ e_{pq}^\alpha + \frac{\nu^\alpha}{1 - 2\nu^\alpha} e_{nn}^\alpha \delta_{pq} \right], \quad (3)$$

with summation over repeated latin indices,  $\delta_{pq}$ , the Kronecker symbol,  $n, p, q = x, y, z$  and  $e_{pq}^\alpha = \frac{1}{2}(\partial_q u_p^\alpha + \partial_p u_q^\alpha) - \eta^\alpha \delta_{pq}(\delta_{p1} + \delta_{p2})$  where  $\mathbf{u}$  is the displacement with respect to the reference state commensurate with the substrate so that  $\eta^f = a^f/a^s - 1$  and  $\eta^s = 0$ . Since the system is supposed to be at mechanical equilibrium, it satisfies  $\partial_q \sigma_{pq}^\alpha = 0$  with the following boundary conditions:  $\mathbf{u}^s \rightarrow \mathbf{0}$  when  $z \rightarrow -\infty$  and is continuous at  $z=0$ ; at  $z=0$ ,  $\sigma_{pz}^\alpha$  is continuous while at the free surface  $z=h(x, y, t)$  with the outward normal  $\mathbf{n}$ ,  $\sigma_{pq}^f n_q = 0$  as the wetting surface stress is negligible in usual films.<sup>7</sup> Elasticity is solved within the thin film approximation, following Ref. 5, assuming the thickness  $h$  to be an order  $\epsilon$  smaller than the characteristic length  $l_0$ . In the film, we consider the rescaled variable  $Z = z/\epsilon$  and derive here  $\mathbf{u}$  as an expansion up to  $\epsilon^3$ ,  $\mathbf{u} = \sum_{n=0}^3 \epsilon^n \mathbf{u}^{(n)}(x, y, Z)$ . In the plane substrate however, elasticity is solved as usually using Fourier transforms with respect to  $\mathbf{r} = \{x, y\}$ ,  $\mathcal{F}[h] = (2\pi)^{-2} \int d\mathbf{r} e^{i\mathbf{k}\cdot\mathbf{r}} h(\mathbf{r})$ . After consideration of the boundary conditions, we compute the elastic energy density  $\mathcal{E}[h] = \frac{1}{2} e_{pq}^\alpha \sigma_{pq}^\alpha$  up to  $\epsilon^2$ , the first nonlinear term which requires  $\epsilon^3$  terms in  $\mathbf{u}$ . Eventually, we obtain the central result of this article describing the 3D film dynamics,

$$\begin{aligned} \frac{\partial h}{\partial t} = \Delta \left\{ - \left[ 1 + c_w f \left( \frac{h}{\delta} \right) \right] \Delta h + \frac{c_w}{\delta} \frac{f'(h/\delta)}{\sqrt{1 + |\nabla h|^2}} \right. \\ \left. - \omega_1 \mathcal{H}_{ii}(h) + \omega_2 \left( 2h\Delta h + |\nabla h|^2 \right) \right. \\ \left. + \omega_2^* \left( 2\mathcal{H}_{ij} [h \theta_{ijkl} \mathcal{H}_{kl}(h)] + \mathcal{H}_{ij}(h) \theta_{ijkl} \mathcal{H}_{kl}(h) \right) \right\}, \quad (4) \end{aligned}$$

with  $i, j, k, l = x, y$ . In (4), we use the notation  $\theta_{ijij} = 1$ ,  $\theta_{xxyy} = \theta_{yyxx} = \nu_f$ ,  $\theta_{xyyx} = \theta_{yxyx} = -\nu_f$ , and  $\theta_{ijkl} = 0$  otherwise, and define the functionals

$$\mathcal{H}_{ij}[h] = \mathcal{F}^{-1} \{ (k_i k_j / k) \mathcal{F}[h] \}, \quad (5)$$

with  $k = |\mathbf{k}|$ . The different elastic constants are  $\omega_1 = 2E_f(1 - \nu_s^2)/E_s(1 - \nu_f)$ ,  $\omega_2 = (1 + \nu_f)/(1 - \nu_f) + E_f(1 - 2\nu_s)(1 + \nu_s)/E_s(1 - \nu_f)$  and  $\omega_2^* = 2E_f^2(1 - \nu_s^2)^2/E_s^2(1 - \nu_f)^2(1 + \nu_f)$ , which match  $2(1 + \nu_f)$  in the case of equal film and substrate elastic properties,  $\nu_s = \nu_f$  and  $E_s = E_f$ . In fact, up to order  $h^2$ , Eq. (4) corresponds to the elastic free energy

$$\mathcal{F}^{\text{El}} = \int d\mathbf{r} h(\mathbf{r}) \left[ -\frac{1}{2} \omega_1 \mathcal{H}_{ii}(h) - \omega_2 |\nabla h|^2 + \omega_2^* \mathcal{H}_{ij}(h) \theta_{ijkl} \mathcal{H}_{kl}(h) \right]. \quad (6)$$

In two dimensions (2D), Eq. (4) reduces to

$$\begin{aligned} \frac{\partial h}{\partial t} = \frac{\partial^2}{\partial x^2} \left\{ - \left[ 1 + c_w f \left( \frac{h}{\delta} \right) \right] h_{xx} + \frac{c_w}{\delta} \frac{f'(h/\delta)}{\sqrt{1 + h_x^2}} \right. \\ \left. - \omega_1 \mathcal{H}(h_x) + \omega_2 (2hh_{xx} + h_x^2) \right. \\ \left. + \omega_2^* \left( 2\mathcal{H} \{ [h\mathcal{H}(h_x)]_x \} + [\mathcal{H}(h_x)]^2 \right) \right\}, \quad (7) \end{aligned}$$

where  $x$ -indices denote  $x$ -derivatives and  $\mathcal{H}$  is the Hilbert transform acting in Fourier space as  $\mathcal{H}[h_x] = \mathcal{F}^{-1} \{ |k| \mathcal{F}[h_x] \}$ . For equal film and substrate elastic properties and without wetting effects, we retrieve the results of Ref. 15 describing a 2D semi-infinite film.

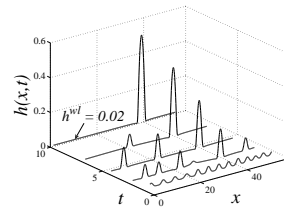


FIG. 1: Space-time evolution of a 2D film according to (7) with  $h_0 = 0.1$ . Surface diffusion induces a non-interrupted coarsening until only one island is left surrounded by a wetting layer with height  $h^{wl}$ .

We now investigate the dynamics predicted by Eqs. (4) and (7). In the linear regime, considering small perturbations of amplitude  $\exp[\sigma(\mathbf{k})t + i\mathbf{k}\cdot\mathbf{r}]$  around a flat film of height  $h_0$ , we find  $\sigma(\mathbf{k}) = -ak^2 + \omega_1 k^3 - bk^4$  with  $a = 1 + c_w f(h_0/\delta)$  and  $b = c_w f''(h_0/\delta)/\delta^2$ . Hence, when  $f''(\xi)$  is decreasing and positive, there exists some critical height  $h_c$  below which  $\sigma(\mathbf{k}) < 0$  everywhere so that the film is linearly stable thanks to the wetting interactions. However, for  $h_0 > h_c$ ,  $\sigma(\mathbf{k}) > 0$  for  $k_{min} < k < k_{max}$  with  $k_{min} > 0$ , so that the film is linearly unstable consistently with Ref. 16. For small wetting length  $\delta$  and exponential wetting potential, one gets  $h_c \simeq -\omega_2 \delta \ln(\omega_1 \delta^2 / 4c_w)$ .

To analyze the following nonlinear evolution of this instability, we performed numerical simulations using a pseudo-spectral method in a periodic box of length  $L$ . To be specific, we selected parameters depicting a  $\text{Si}_{0.8}\text{Ge}_{0.2}$

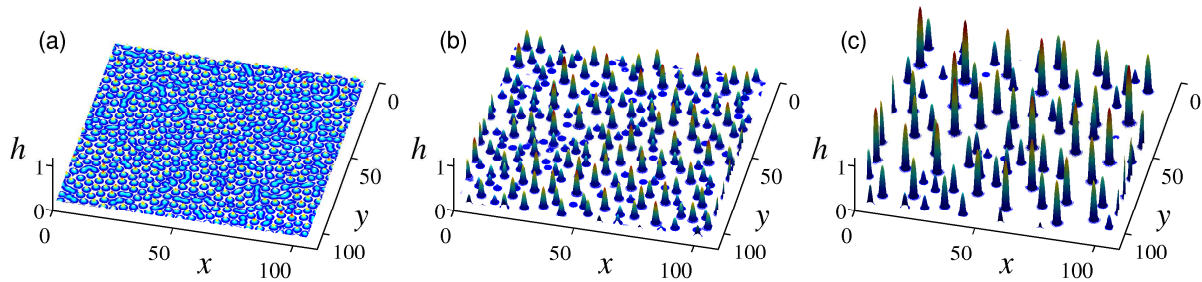


FIG. 2: Space-time evolution obtained by Eq. (4) with  $t=3.9$  (a), 8.3 (b) and 13.9 (c).

film on a Si substrate with  $\nu_f=0.278$ ,  $\omega_1=2.44$ ,  $\omega_2=2.52$  and  $\omega_2^*=2.34$ , leading to  $l_0=200$  nm and, with the value of the diffusion parameter  $D$  given in Ref. 12,  $t_0=8$  hours at  $750^\circ\text{C}$ , see Ref. 4. In fact, thanks to space and time rescaling, only  $\nu_f$  and  $\omega_2^*/\omega_2$  are relevant parameters for characterizing Eqs. (4) and (7). The wetting potential is described in an indicative way by  $c_w=0.05$  and  $\delta=0.005$ . The initial condition is a flat film perturbed by a small noise with a mean initial height  $h_0$ . As shown in Figs. 1 and 2, a film with  $h_0 > h_c$  is first destabilized by the morphological instability which generates surface undulations according to the linear growth. This stage is then quickly replaced by a nonlinear one which in fact does not display any singularity. Instead, islands emerge and grow without moving and with a decreasing aspect ratio, surrounded by a wetting layer smaller than  $h_c$  which allows surface transport and the subsequent islands ripening. Studying parity in  $h$  of the different terms in (6), one concludes following Ref. 15 that the last nonlinear nonlocal term drives the surface towards smooth peaks and deepening valleys which would lead to singularities. However, the wetting effects included in  $\gamma(h)$  enforce here a higher energetic cost for small  $h$  and thus stabilize the system. In fact, *both* nonlocal nonlinearity and wetting are crucial for regularizing the dynamics of the instability which we now characterize by its final state and time dependence.

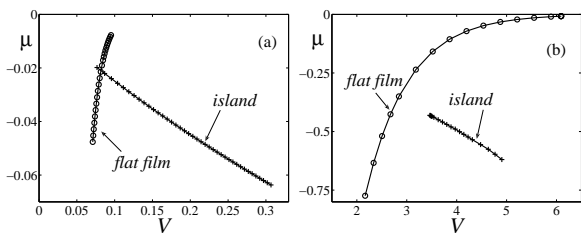


FIG. 3: Equilibrium surface chemical potential (2) as function of the flat film volume  $V=V^f$  when  $h < h_c$ , and of the island volume  $V$  when  $h > h_c$  in 2D (a), 3D (b).

Within the present model, the system evolves continuously in both 2D and 3D towards an equilibrium state characterized when  $h_0 > h_c$  by a single stable island in equilibrium with a wetting layer of height  $h^{wl}$ , see e.g.

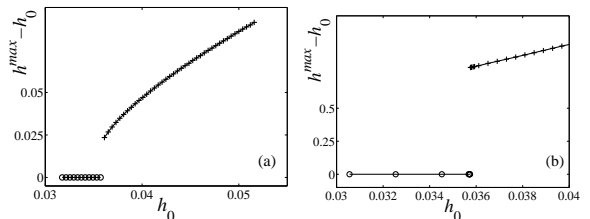


FIG. 4: Maximal height of an equilibrium island as function of the initial height in 2D (a), 3D (b).

Fig. 1, whereas when  $h_0 < h_c$ , the final stage is a flat film of height  $h^{wl}=h_0$ . The equilibrium properties ( $h^{wl}$ , island volume  $V$  measured above  $h^{wl}$ , etc.) depend only on the homogeneous chemical potential (2) and on the sign of  $h_0 - h_c$  for large enough  $L$ . Computing  $\mu$  and  $V$  at equilibrium as parametric functions of the film volume  $V^f$ , we find that when  $h < h_c$ ,  $\mu=\gamma'(h)$  increases with  $V^f$  until  $V_c^f=L^2h_c$ , whereas when  $h > h_c$ ,  $\mu$  depends only on  $V$  and is monotonously decreasing in both 2D and 3D similarly to Ref. 17, see Fig. 3. Hence, in a regime of well-separated islands, bigger ones should always grow by surface diffusion at the expense of smaller ones. We also compute the equilibrium maximum height  $h^{max}$  as function of the initial height  $h_0$ , see Fig. 4. The system undergoes a discontinuous bifurcation as the difference  $h^{max}-h_0$  jumps at the transition height  $h_c$  which agrees within a few percents with the linear estimate  $h_c \simeq 0.036$  corresponding to 7 nm. This first-order like transition also shown in the  $\mu(V)$  plot of Fig. 3 is at stake in similar instabilities.<sup>21</sup>

Finally, to describe quantitatively the dynamics of the island growth, we compute the surface roughness  $w(t) = (\langle h^2 \rangle - \langle h \rangle^2)^{1/2}$ , number of islands  $N(t)$  and the island surface coverage  $\theta(t)$ .<sup>25</sup> Both 2D and 3D simulations reveal a non-interrupted coarsening with power-law behavior  $w(t) \sim t^\beta$ ,  $N(t) \sim 1/t^\zeta$  and  $\theta(t) \sim 1/t^\gamma$ , see Figs. 5, 6 and 7. For 2D systems, we find  $\beta=0.26$ ,  $\zeta=0.59$  and  $\gamma=0.43$  over nearly three decades. Similarly, over the last time-decade of the 3D simulations, we find  $\beta=0.7$ ,  $\zeta=1.3$  and  $\gamma=0.8$  which are noticeably departing from the 2D values illustrating the difference between diffusion process over a one or two dimensional

surface.

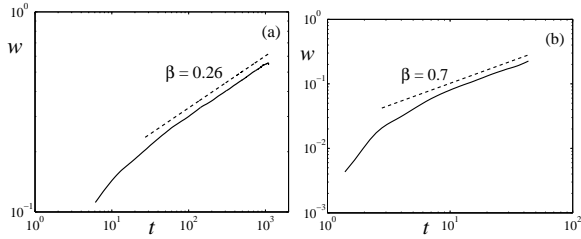


FIG. 5: Roughness as function of time with  $L = 6700$  in 2D (a) and  $L = 209$  in 3D (b).

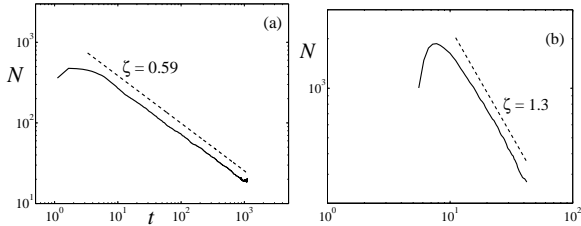


FIG. 6: Evolution of the number of islands with the same parameters as in Fig. 5.

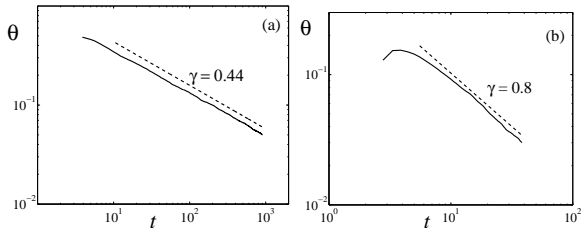


FIG. 7: Evolution of the surface coverage with the same parameters as in Fig. 5.

Significant discrepancies can be noted with the dynam-

ical behavior found in Ref. 6 (coarsening termination) and Ref. 7 (coarsening with different exponents). However, both studies regularize the ATG singularity with diverging<sup>7</sup> or dominant<sup>6</sup> wetting potentials. Moreover, the analysis in Ref. 6 dismisses the film and substrate interface and applies in fact to a semi-infinite elastic film, whereas Ref. 7 did not consider nonlinear elastic effects. Hence, one expects the adequately nonlinear equation (4) with regular wetting potential to give a reliable nonlinear behavior. Note that finite-element simulations concerning a similar system were performed in Ref. 22 but did not explore the island coarsening behavior.

In summary, we derived a nonlinear model describing the stress driven morphological instability of a thin film on a deformable substrate with a priori different elastic constants and which accounts for wetting interactions. The combination of *both* wetting and *nonlocal* nonlinearities is essential for regularizing the finite-time singularity at stake in the bulk instability and numerical simulations reveal a regular evolution towards an equilibrium state. The latter stage consists of a single island with a chemical potential monotonously decreasing with its volume. Consistently, the system undergoes a *non-interrupted coarsening* in both two and three dimensions characterized by power-law behavior with time which significantly depend on the system dimensionality. Further experiments on the number of islands of annealing films in the prepyramid regime of the Stranski-Krastanov mode<sup>4</sup> would be of great interest. Note that we did not account here for more complex effects regarding surface dynamics such as alloying<sup>23</sup> or anisotropy<sup>24</sup>, the influence on coarsening of which is left for future work within nonlinear analysis.

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\* Electronic address: jnaqua@irphe.univ-mrs.fr; École Centrale Marseille, France

† Electronic address: frisch@irphe.univ-mrs.fr

‡ Electronic address: verga@irphe.univ-mrs.fr

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