Supplementary Material

An appropriate numerical initialisation of the liquid sheet is critical to trigger droplet shedding. Consequently, to demonstrate the robustness of our study, we have performed a range of simulations varying the initial perturbation, ϵ, and the initial ratio of the film thickness to the radius of the rim, e.

The initial instability, ϵ

First, we turn our attention to the breakup time of liquids and the selection of ϵ. According to Driessen *et al.* [2], in the inviscid limit, the maximum growth rate of a surface perturbation is inversely proportional to the capillary breakup time, $t_{cap} = (\rho a_o^3/\sigma)^{1/2}$. However, when viscosity plays a role, the R-P instability needs to account for a nonlinear term which depends on the Oh number. Consequently, the (dimensionless) growth rate of the viscous-mediated R-P instability is given by

$$\omega t_{cap} = \sqrt{\frac{1}{2}(k^2 - k^4) + \frac{9}{4}Oh^2k^4} - \frac{3}{2}Ohk^2,$$

where, for each value of Oh, the fastest growth rate is given by $k_{max} = \left(2 + 3\sqrt{2}Oh\right)^{-1/2}$, [2]. Under the assumption that the R-P instability is triggered at k_{max}, the breakup time t_B is given by:

$$t_B = t_0 + \frac{1}{\omega_{max}(Oh)} log\left(\frac{1}{\epsilon}\right) =,$$

where δ is the initial amplitude of the perturbation (at $t = t_0$), and the relative initial disturbance is given by $\epsilon = \delta/a_0$. Under our conditions, as clearly seen in Figure 1(a), a large ϵ is required for the breakup dynamics to be driven by the surface perturbation.

Figure 1(a-c) shows the effect of the initial perturbation, ϵ, on the dynamics of the liquid sheet in the absence of surfactants when $Oh = 0.0833$ and $e = 0.2$ at $t = 188$. Here, the three-dimensional representation of the interface shows that, in the absence of ϵ, the rim is stable and does not lead to perturbations on its surface. Therefore, it is evident the need of an initial ϵ to trigger the growth of the nonlinear modes. We observe that the addition of a perturbation leads to a capillary singularity. As expected, the larger the ϵ is, the shorter the capillary breakup time. In addition, we monitored the temporal evolution of the ligament's tip position and the kinetic energy as a function of ϵ, see figure 1b and c. We observe similar trends regardless of the value of the initial perturbation.

Figures 1(d-f) show the effect of the initial perturbation, ϵ, on the sheet dynamics in the presence of surfactants when $\beta_s = 0.3$. The three-dimensional representation of the interface shows that in the presence of ε the R-P instability triggers droplet detachment for $\epsilon > 0.1$ (see figure 1d). In the absence of an initial ϵ, there is an uniform recoiling, and the only effect observed is the convection of surfactant from the rim towards the sheet, resulting in the reopening of the sheet. Similar trends are observed for the ligament's tip position, and the kinetic energy, regardless of the initial perturbation amplitude (see figure 1e,f).

The initial ratio of the film thickness to the radius of the rim, e

Next, we turn our attention towards the variation of the ratio of the film thickness to the radius of the rim, e, for surfactant-free cases (see figure 2).

Sweeping across a range of e is a delicate job as this requires adjusting the overall size of the domain, and consequently affecting the computing costs. In simulations, as demonstrated by Fullana & Zaleski [3], the break up of a liquid rim, due to a surface instability, depends on the size of the domain, i.e. in short domains (in the work of Fullana & Zaleski [3] (1999), the total length of the sheet was 40 times its thickness) every unstable mode eventually becomes linearly stable but, for a sufficiently large spatial domain, the surface remains unstable and the dynamics are driven by the perturbation. In our study, the total length of the sheet is ~ 305 times the thickness of the sheet.

Figure 2 illustrates the effect of varying the initial ratio of the film thickness to the radius of the rim, e, on the rim dynamics. Figure 2(a-c) shows the variation of e, for surfactant-free cases. We observe that this parameter affects heavily the dynamics of the tip location and the kinetic energy. Figure 2(d-f) shows the effect of the initial radius rim, e, on the sheet dynamics in the presence of surfactants when $\beta_s = 0.3$.

Figure 2 shows a three-dimensional representation of the interface where the R-P instability triggers droplet detachment for $\beta_s > 0.3$. In the absence of an initial ϵ, there is an uniform recoiling, and the only effect observed is

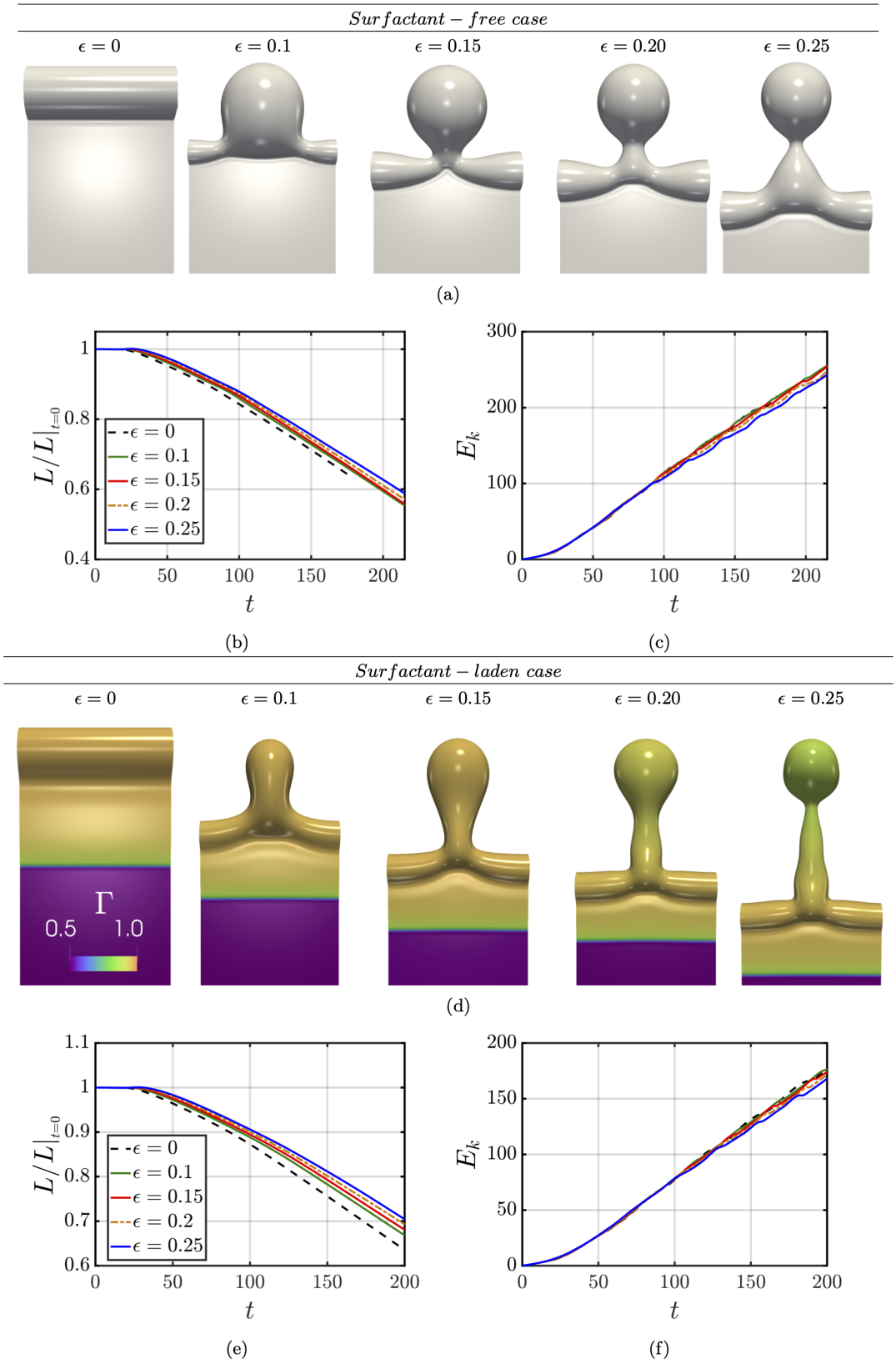

FIG. 1: Effect of the initial amplitude of the perturbation, ε, for the surfactant-free and surfactant-laden cases corresponding to panels (a-c) and (d-f), respectively. Three-dimensional representation of the interface at $t = 188$, the temporal evolution of the ligament's tip position, the kinetic energy are shown in panels (a)-(d), (b)-(e) and (c)-(f), respectively. Here, $Oh = 0.0833$ and $e = 0.2$; for the surfactant-laden case: $\beta_s = 0.3$ and $\Gamma = \Gamma_\infty/2$

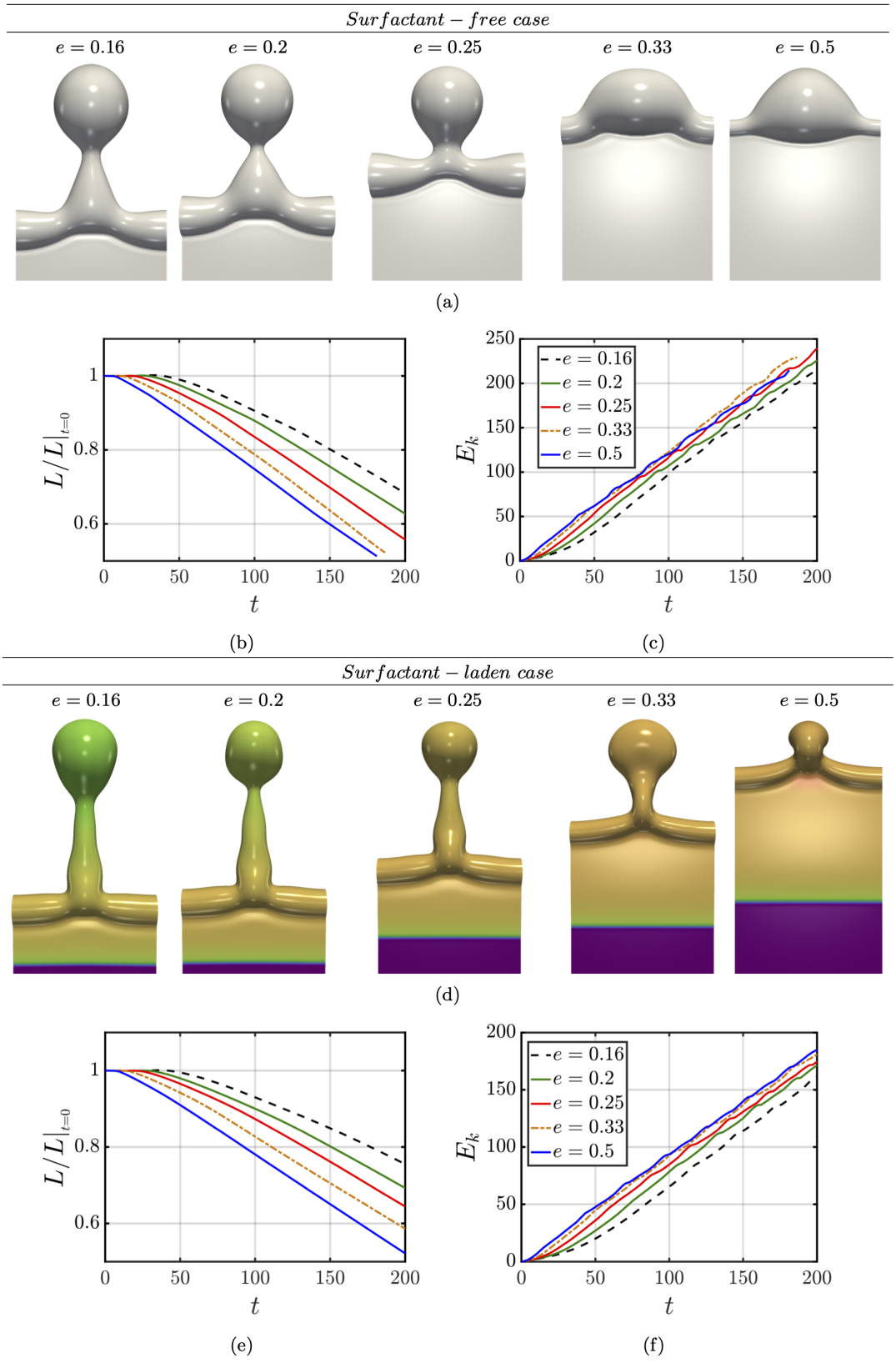

FIG. 2: Effect of the parameter e for the surfactant-free and surfactant-laden cases corresponding to panels (a-c) and (d-f), respectively. Three-dimensional representation of the interface at $t = 188$, the temporal evolution of the ligament's tip position, the kinetic energy are shown in panels (a)-(d), (b)-(e) and (c)-(f), respectively. Here, $Oh = 0.0833$ and $\varepsilon = 0.25$; for the surfactant-laden case: $\beta_s = 0.3$ and $\Gamma = \Gamma_\infty/2$ (same legend as figure 1d).

the convection of surfactant from the rim towards the sheet, resulting in the reopening of the sheet. Similar trends, regardless of the initial perturbation amplitude, are observed for the ligament's tip position and the kinetic energy (see figure 2e,f).

In addition, our selection of $e = 0.2$ is justified by the work by Agbaglah *et al.* [1], that presented 3D numerical simulations on retracting sheets leading to droplet detachment. In fact, their results have guided this study. Agbaglah *et al.* [1] chose an initial value of $e = 0.2$, so that the viscous effects on the rim could be minimized; allowing for inertia and surface tension to dominate the rim dynamics.

[1] Agbaglah, G., Josserand, C. & Zaleski, S. 2013 Longitudinal instability of a liquid rim. *Phys. Fluids* **25** (2), 022103.
[2] Driessen, T., Jeurissen, R., Wijshoff, H., Toschi, F. & Lohse, D. 2013 Stability of viscous long liquid filaments. *Phys. Fluids* **25** (6), 062109.
[3] Fullana, J. M. & Zaleski, S. 1999 Stability of a growing end rim in a liquid sheet of uniform thickness. *Phys. Fluids* **11**, 952.