Monitoring photochemical reactions using Marangoni flows
Supporting information

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I. INTERMITTENT VS CONTINUOUS IRRADIATION

In the experiments, a chopper was used to facilitate the recording of interferometric images without blinding the camera. The duty cycle was 50% and the effective illumination frequency was 4 Hz (see inset in Fig. 1). Measurable deformations were only obtained after at least a few seconds, which by far exceeds the chopper time scale of 0.25 s. For reasons of numerical efficiency, we therefore implemented continuous illumination with half the relevant reaction rate $j_{\mathrm{bulk}}/2$ in the numerical simulations. This has no observable effect on the deformation dynamics as shown in Fig. 1, where we compare a simulation with continuous illumination at $j_{\mathrm{bulk}}/2$ with two simulations with intermittent illumination at two frequencies of 2 and 4 Hz, respectively.

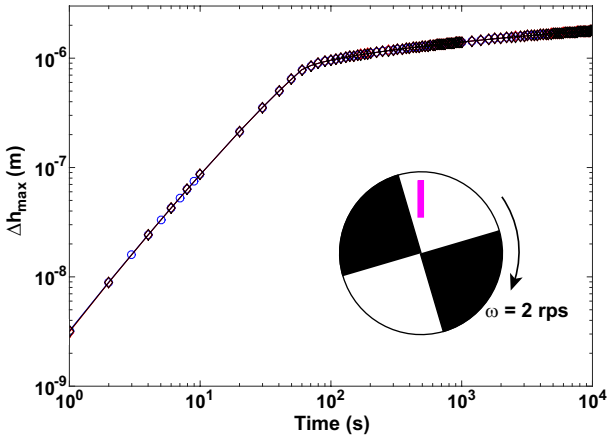

FIG. 1. Comparison of a simulation with continuous illumination at $j_{\mathrm{bulk}}/2$ (blue line and circles) with two simulations at j_{bulk} with intermittent illumination at frequencies of 2 and 4 Hz, respectively (red and black lines and diamonds). The inset represents a sketch of the chopper blade, which is rotating at 2 rps.

II. VIDEO OF A TYPICAL EXPERIMENT

The video *Muller_et_al_video_typical_experiment.avi* presents a typical experiment for an illumination time $t_{\mathrm{UV}} = 20\,\mathrm{s}$ and an initial layer thickness $h_0 = 4.8\,\mu\mathrm{m}$. The video was recorded with a frame rate of 1 frame/s and represents a duration of the experiment of 5 min.

III. PARAMETER STUDY OF THE SURFACE-DOMINATED REACTION MODEL

Analogous to Fig. 6 of the manuscript, we performed numerical simulations of the film thickness modulation Δh_{max} as a function of $J_{\mathrm{surf}}(\partial\gamma/\partial C)$, D and w_x for the surface-dominated reaction model. The results are qualitatively very similar to those for the bulk-dominated reaction model and are presented in Fig. 2.

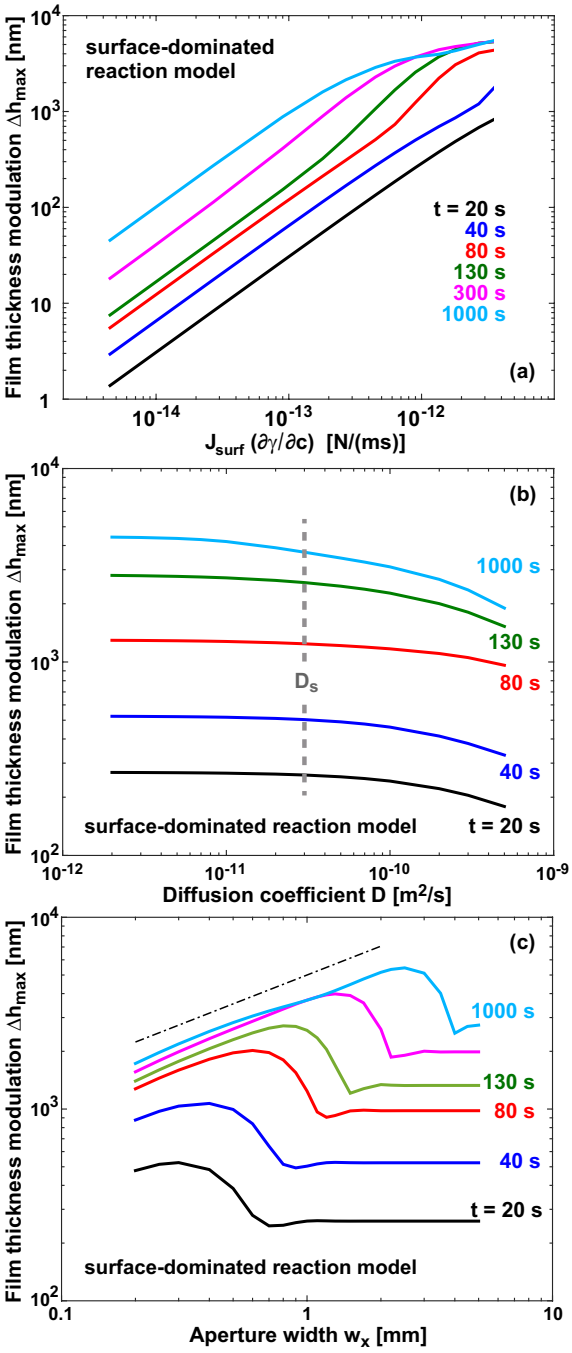

FIG. 2. (a) Numerical simulations of $\Delta h_{\max}$ as a function of the rate of increase of surface tension $J_{\mathrm{surf}}(\partial\gamma/\partial C)$ at different times t = 20, 40, 80, 130, 300 and 1000 s. (b) $\Delta h_{\max}$ as a function of the diffusion coefficient D at different times t = 20, 40, 80, 130 and 1000 s. The dashed vertical line indicates the self-diffusion coefficient of squalane. (c) $\Delta h_{\max}$ as a function of aperture width w_x at different times t = 20, 40, 80, 130, 300 and 1000 s. The parameters that were not varied in (a,b,c) were $h_0 = 3\,\mu$m, $w_x = 1$ mm, $w_y = \infty$, $\Delta w = 0.2$ mm, $t_{\mathrm{UV}} = 60$ s, $D = 3 \cdot 10^{-11}\mathrm{m}^2/\mathrm{s}$, $J_{\mathrm{surf}}(\partial\gamma/\partial C) = 1.8 \cdot 10^{-12}\mathrm{N}/(\mathrm{m\,s})$. The surface-dominated reaction model was used.