.

Supplemental Material

for "Early azimuthal instability during drop impacts"

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Drop free-fall oscillations and impact velocity:

4.4. *Drop impact velocity and aspect ratio vs release height*

The drop shape and impact velocity U was characterized in a separate set of experiments using the same liquid. Following Thoraval *et al.* (2013) we thereby measured the aspect ratio and modelled the impact velocity using the expression

$$U = V_T \sqrt{1 - \exp\left(-\frac{2g\,(H - D - h_0)}{V_T^2}\right)}, \tag{4.1}$$

where $V_T = 8.52$ m/s corresponds to the terminal velocity of the drop. Gravity is $g = 9.81$ m/s^2 and $h_0 = 1.93$ mm is a fitting parameter accounting for the pinch-off dynamics. Here H is the measured distance from the nozzle tip to the undisturbed pool surface, whereas the adjusted height is defined as $H = h - D - h_0$. Figure S1 shows the fit to the experimental data, showing that air drag slows down the free-fall velocity.

We define the aspect ratio of the drop as

$$\alpha = \frac{D_V}{D_H},$$

where D_V and D_H are the instantaneous vertical and horizontal diameters. Following again Thoraval *et al.* (2013) we can estimate the aspect ratio of the drop following the pinch-off from the nozzle. The drop is much larger than the capillary length $\ell_c = \sqrt{\sigma/(\rho g)}$, which for water is 2.7 mm. The axisymmetric oscillation of a drop can be accurately modeled by inviscid potential theory in terms of Legendre polynomials, see Lamb 1975 (section 275),

$$R(t,\theta) = R_o[1 + a\,cos(\omega t + \phi)\,P_2(cos\,\theta)], \tag{4.2}$$

where $P_2(x) = (3x^2 - 1)/2$ is the first non-trivial mode of oscillation. θ is the polar angle in the spherical coordinates. a is the amplitude of the oscillations ω is the oscillation angular frequency and ϕ a phase angle. From this disturbance shape we can calculate the aspect ratio of the vertical to horizontal axes, as

$$\alpha = \frac{D_V}{D_H} = \frac{1 + a\,cos(\omega t + \phi)}{1 - (a/2)cos(\omega t + \phi)}. \tag{4.3}$$

This aspect ratio is plotted in Fig. S2 with the best-fit values of a, ω and ϕ. The curve is plotted vs. falling height and is stretched by the free-fall acceleration. The amplitude is also damped by the viscous dissipation within the liquid.

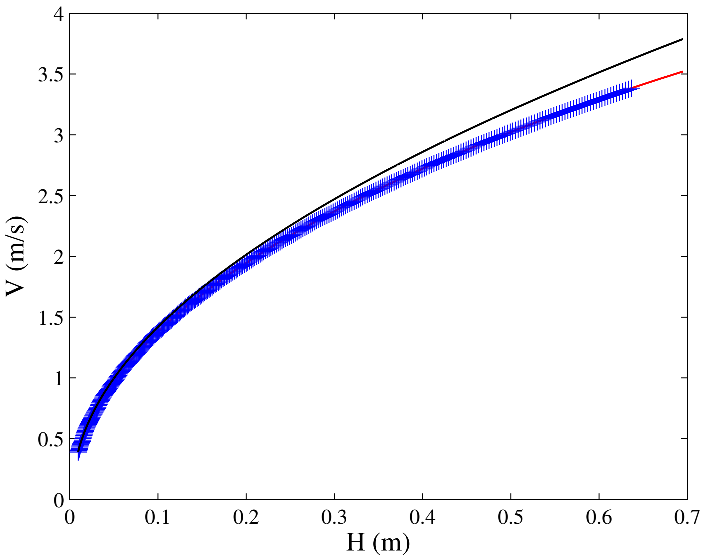

FIGURE S1. The impact velocity as function of release height H, measured from high-speed video, blue symbols. The red line corresponds to Eq. (4.1), whereas the solid black line is simply $\sqrt{2g(H-D)}$.

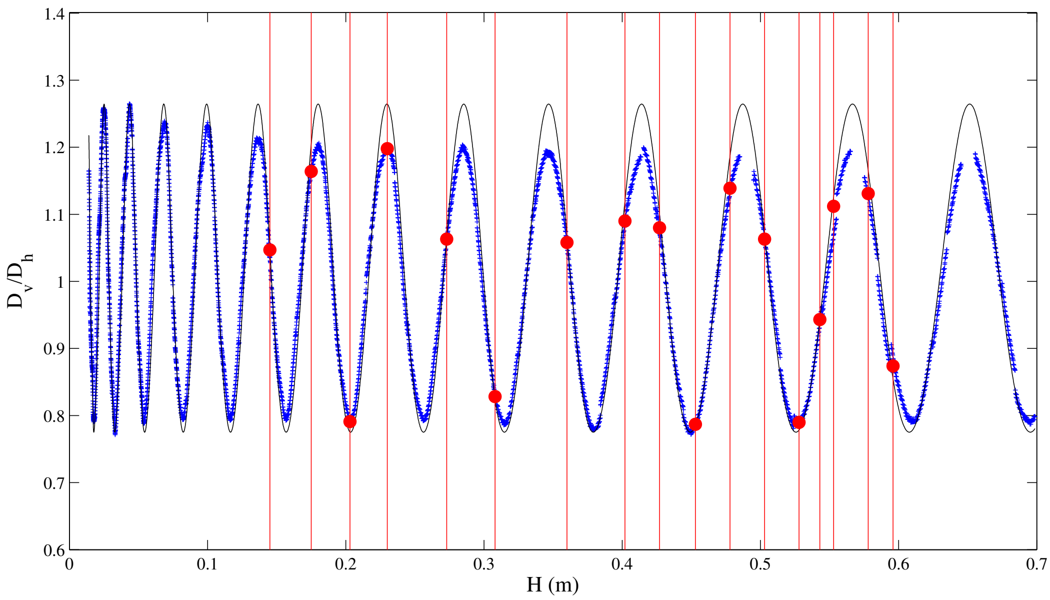

FIGURE S2. The aspect ratio of the drop vs impact height. The vertical red lines mark the impact heights used in the experiments and the red dots are the corresponding aspect ratios. The solid curve is the theoretical inviscid oscillation from Eq. (4.3), following Thoraval *et al.* (2013). The blue symbols are the experimentally measured aspect ratios, during the free-fall of the drop.

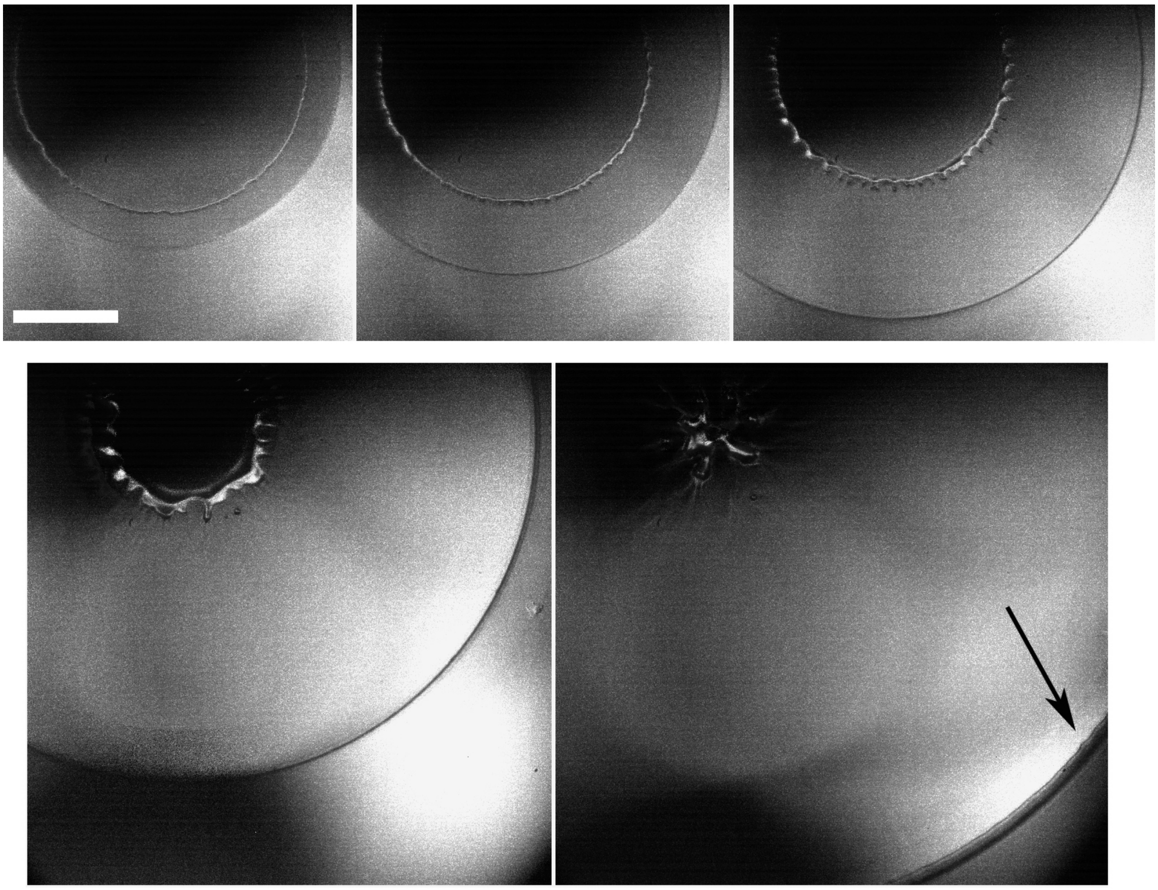

FIGURE S3. The smooth neck for the lowest impact velocity of $U = 1.64$ m/s, $Re_\alpha = 6,510$ and $We_\alpha = 163$. Times shown at $t = 2$, 5, 11, 29 & 65 μs from impact. Scale bar is 200 μm long. The arrow points at a weak undulation.

4.5. *Smooth neck shape for low impact velocity*

Figure S3 shows that the edge of the neck between drop and pool remains smooth for low values of the impact Re and We. The arrow in the last panel, points at weak undulations in the neck surface, which restabilize at larger radii. Thoraval *et al.* (2013) noticed similar weak undulations for Re as low as 3,610.

Supplemental References

LAMB, H., *Hydrodynamics, 6th edn.* Dover 1975.

THORAVAL, M.-J., TAKEHARA, K., ETOH, T. G. & THORODDSEN, S. T. 2013 Drop impact entrapment of bubble rings, *J. Fluid Mech.*, **724**, 234-258.