Double contact during drop impact on a solid under reduced air

Supplemental Material

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Effects of Gravity

The importance of gravity can be determined by comparing its influence with droplet inertia. The appropriate non-dimensional parameter for this is (the reciprocal of), the Froude number $Fr = V/\sqrt{gR}$. In the cushioning region itself, scaling arguments imply the first influence of gravity occurs (in the droplet momentum equation), when $St^2/3/Fr^2 \sim 1$. This gives a condition on the impact velocity $V > (g^3 R \mu^2 / \rho^2)^{1/8}$, for impacts in which gravity is negligible over the cushioning time scale. In our experiments, this equates to $V \gg 0.01 \text{ m/s}$ and hence gravity is not relevant in our experiments, which always have much higher impact velocities. Determination of the Bond number $\Delta \rho g R^2/\sigma$, which measures the importance of gravity relative to surface tension, is not instructive, as surface tension is unimportant in our experiments.

We conclude that gravity only determines the magnitude of $V$ and has no effect on the very rapid dynamics during the early contact studied herein, where neither gravity nor surface tension play a role.

Effects of Evaporation

The possible effects of evaporation can emerge in two ways. First, evaporation can alter the properties of the drop liquid and surrounding atmosphere on timescales of minutes. Secondly, evaporation can modify the thin air layer during the cushioning on timescales of micro-seconds. The change in liquid properties can affect the volume of the drop, lower temperature due to cooling can change surface tension and viscosity, and increase in the water content of the surrounding atmosphere. Changes in the volume of the drop would affect the radius of the drop; however, since we measure the radius of the drop immediately prior to impact any changes of the radius prior to this will not have any bearing. A decrease in the temperature of the drop could increase the surface tension and viscosity of the drop; however, as can be seen in Figure 6, we are in a regime where surface tension is not important and the liquid viscosity, remains low and does not enter into our scaling analysis. This leaves only an increase in the water content of the surrounding atmosphere, which could decrease the viscosity of the air. Our experiments were not designed to control for humidity and thus the water content in the air has random variations across all of the experiments. Keep in mind that the vacuum chamber is not fully dry between each experiment. It contains a small wet container, which catches a few drops before each experimental realization. Furthermore, splashed droplets are also present at its bottom. These water sources will contribute to the vapor content within the reduced-pressure air, which evaporate for about 10 minutes before a typical impact. This should diminish any effects of vaporization on the surface of the pendent drop.

Regarding the second point: the very short cushioning time scale gives limited time for evaporation from the droplet interface. The Hertz-Knudsen equation quantifies the net evaporative flux from the droplet interface $j$, which is proportional to

$$j \propto \left( \sigma_e \frac{p_s(T_1)}{\sqrt{T_1}} - \sigma_e \frac{p_e}{\sqrt{T_g}} \right). \tag{1}$$

Here $p_s$ is the saturation vapour pressure, $p_e$ is the partial pressure of water vapour in the gas, $T_1$ and $T_g$ are the liquid and gas temperatures, while $\sigma_e$ and $\sigma_c$ are evaporation and condensation coefficients [1]. For isothermal gas compression, all the terms remain constant except $p_e$, which increases as gas cushions the droplet impact. For adiabatic gas compression heat is not transferred from the gas to its surroundings over the short cushioning time scale, and so while both $p_e$ and $T_g$ increase, $p_s(T_1)$ and $T_1$ remain constant. In both cases, gas cushioning of the droplet reduces $j$, and may produce condensation of water vapour from the gas. Water vapour condensation cannot significantly alter the volume trapped beneath the droplet, because the partial pressure of water vapour (even at saturation), is small compared to the total gas pressure.

The possible effects of evaporation were also addressed for the study of Xu et al. [3], which investigated the splashing under vacuum of more volatile ethanol drops, in a comment [4] and reply [5].
Supplemental Figures

Figure 1 shows a comparison of the experimental data with the scaling derived in equation (2) of the main text. Figure 2 shows predicted values of $H^*$ from [2] covering our experimental parameter space. This figure also shows where the lower limit for the interferometry method that we use lies. Figure 3 shows that double contact can also occur for impacts on to a thin layer of water. Figure 4 compares the effects of the nanometer scale surface roughness on the entrained air discs at high values of the compressibility factor $\epsilon^{-1}$. More details are provided in the captions of each of the figures.

FIG. 1. The initial radial size of the air-disc $L_0$, showing the scaling from equation (2) in the main text, using isothermal compression ($\gamma = 1$) in (a) versus adiabatic compression of the gas ($\gamma = 1.4$) in (b). The lines have a slope of 1. We note that the theory used to develop equation (2) is considering a compressible, rarefied gas and thus only drop impacts that have $Kn \gtrsim 1$ are expected to follow this scaling, which roughly corresponds to the point of bifurcation and further left. In the upper branch of the bifurcation, there are impacts that the theory would predict should fall on the lower line, but do not. Solid symbols indicate the second contact, or outer edge of the band of micro-bubbles.
FIG. 2. Predicted values of $H^*$ from Mandre et al. [2], assuming isothermal compression (solid black line). The smallest predicted value of $H^*$, in our parameter range, is a mere 1.7 nm. This refers to the $H$ at the centerline where the air-layer is thickest. We must conclude that van der Waals forces can come into play during these impact conditions, when the slip due to rarefied-gas effects thins locally the air disc. The interferometry technique used in this paper cannot accurately resolve air disk thicknesses smaller than 160 nm.
FIG. 3. Drop with $R_b = 4.4$ mm impacting a thin layer of water at $V = 5.0 \text{ m/s}$ in a reduced ambient pressure of 5 kPa. The same double contact phenomenon which occurs on a dry substrate also occurs on a very thin film of water. However, in this instance after the torus of air has contracted into a ring of bubbles the radius of the ring of bubbles continues to expand. The scale bar is 100 $\mu$m long.

FIG. 4. Comparison of the effect of surface roughness at high values of the compressibility factor. (a) Impact onto Fisher glass with $R_a = 1.2$ nm. A cloud of micro-bubbles is formed in place of the air disc. The microbubbles do not contract into one central bubble $P = 3.25$ kPa, $V = 3.45 \text{ m/s}$, $R_b = 3.49$ mm, $\epsilon^{-1} = 317$. (b) Impact onto a freshly cleaved mica sheet. In this case, contact is made around a ring and the central air disc contracts into a tiny bubble. $P = 2.94$ kPa, $V = 3.45 \text{ m/s}$, $R_b = 3.70$ mm, $\epsilon^{-1} = 356$. Scale bar is 100 $\mu$m long.