Drop splashing is independent of substrate wetting

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A liquid drop impacting a dry solid surface with sufficient kinetic energy will splash, breaking apart into numerous secondary droplets. This phenomenon shows many similarities to forced wetting, including the entrainment of air at the contact line. Because of these similarities and the fact that forced wetting has been shown to depend on the wetting properties of the surface, existing theories predict splashing to depend on wetting properties as well. However, using high-speed interference imaging we observe that at high capillary numbers wetting properties have no effect on splashing for various liquid-surface combinations. Additionally, by fully resolving the Navier-Stokes equations at length and time scales inaccessible to experiments, we find that the shape and motion of the air-liquid interface at the contact line at the edge of the droplet are independent of wettabiltiy. We use these findings to evaluate existing theories and to compare splashing with forced wetting.

I. INTRODUCTION

At first glance, one might suppose that the same physics should describe a solid plunging into a liquid and a liquid drop impacting a solid: both scenarios revolve around a liquid-gas-solid contact line that is forced to move at large velocities. In the former case, exceeding a critical contact line velocity leads to the destabilization of the contact line and the entrainment of gas bubbles in the liquid. This phenomenon, called dewetting or wetting failure, is also observed in drop impact.

It has therefore been suggested that the onset of contact line instability can serve as an onset criterion for drop splashing. However, contrary to intuition, in this work we find that splashing is independent of the wetting properties of the surface. This result helps us to further our understanding of the many processes that rely on splashing droplets, including erosion, coating, cleaning, cooling, high-throughput drug screening, and different printing technologies.

The wetting of a solid by a liquid depends on many parameters, including viscosity, surface tension, contact line velocity, impurities in the liquid, and roughness and heterogeneities of the substrate. In addition, there is the complication that for a moving contact line the classical fluid-mechanics assumption of a no-slip condition on the wall breaks down, and that due to strong local curvature at a contact line the observed contact angle is not necessarily the same as the microscopic contact angle. For real substrates, which show contact angle hysteresis due to surfaces roughness and chemical heterogeneity, the situation is even more complicated; while it is known from experiments that surface roughness can either enhance or reduce splashing depending on its characteristic length scale, in general its effect on wetting and contact angle hysteresis is poorly understood.

For smooth surfaces experiments on forced wetting have typically focused on the relationship between the velocity of the edge of the liquid/gas interface and the observed dynamic contact angle. Typically, in the steady state case when the contact angle is plotted as a function of the non-dimensionalized edge velocity, i.e. the capillary number, a single curve is found. For low capillary numbers and a completely wetting surface it has been experimentally well established that the contact line moves according to the Tanner-Voinov law. For partly wetting liquids with a sufficiently high viscosity the data can be described by a variation of the same law without the assumption of small slopes. For forced wetting at larger capillary numbers the contact line eventually becomes unstable, and this is a topic of active research.

Models of forced wetting use the wetting properties of the surface as a boundary condition to determine the stability threshold for the contact line. Indeed, the wettability of the object that is plunged into a liquid has been found to have a strong influence on wetting failure. In contrast to recent conclusions based on simulations, we present experimental results which show that for rapidly moving contact lines, for a wide array of liquids, the surface wettability has negligible effect on splashing. We also describe simulations that are able to resolve contact line behavior at high resolution. These simulations reveal that the contact line motion previously associated with splashing on completely wetting surfaces is nearly identical for a completely non-wetting surface. Before dewetting, the rapidly moving contact line in both the wetting and the non-wetting case shows a microscopic contact angle of 180°. This suggests that the assumption of a fixed microscopic contact angle is inapplicable to contact lines that are forced to move at high speeds, as is the case in splashing, and challenges theories based on this assumption.

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strates only affects the contact angle and does not change the spreading dynamics. Fig. 2 shows that drops spread at the same rate regardless of \( \theta_0 \). Air was used as the ambient gas, whose pressure \( P \) was controlled in a vacuum chamber (5kPa \( \leq P \leq 101\text{kPa} \)). Impacts were recorded with high-speed cameras (Vision Research) at rates up to 130 000 fps either from the side as in Fig. 1(a-c), or with interference imaging (d-f). The latter method measures the interference between light reflected from the bottom surface of the spreading liquid and the top surface of the substrate. Wherever the liquid is in contact with the substrate, there is no reflection of light from that surface and thus no light entering the camera. Wherever the two are separated, an interference pattern is created, as seen in Fig. 1(e-f). Since this method is particularly sensitive to the presence of the air gap, it allows us to measure precisely when the contact line begins to entrain air.

A typical splash is presented in the left column of Fig. 1 Fig. 1(a) shows that a drop does not splash immediately. Instead the liquid spreads radially outward in the form of a lamella. The simultaneous interference image shows that the lamella fully wets the substrate. Beginning at time \( t_{\text{sheet}} \) after impact (where we define \( t_{\text{sheet}} \) as the time when the thin sheet first starts to appear at the front of the expanding lamella) one can observe an interference pattern at the liquid-air-solid contact line, as in Fig. 1(c), indicating the presence of a gas film that is of order a micron thick that separates the spreading liquid from the substrate. The time \( t_{\text{sheet}} \) is the start of the formation of a thin sheet of liquid, as can be seen in Fig. 1(b). The thin-sheet grows and ultimately breaks up into the secondary droplets that form the splash (\( t = 2.1\text{ms}, \text{Fig. 1(c,f)} \)).

The thin-sheet creation time depends on a number of parameters. Most importantly, \( t_{\text{sheet}} \) is delayed as the ambient gas pressure is reduced. However, if the pressure is decreased below a threshold pressure \( P_{\text{sheet}} \), instead of being further delayed, the thin sheet will fail to appear and the splash will have been completely suppressed. We quantify the effect of wetting on splashing by measuring the dependence of both \( t_{\text{sheet}} \) and \( P_{\text{sheet}} \) on the surface properties.

II. METHODS

A. Experiments

The experiments were conducted with either silicone oil (PDMS, Clearco Products) of viscosity \( \mu = 9.4 \) and 32cP or a mixture of water and glycerol (\( \mu = 32\text{cP} \)). The drops with diameter \( D = 3.3 \pm 0.1\text{mm} \) were produced at a nozzle with a syringe pump and were accelerated by gravity to an impact velocity of \( V = 3.4 \pm 0.1\text{ms}^{-1} \). This resulted in the non-dimensional numbers and ratios shown in Tab. 1. The silicone oil drops impacted glass slides (Fisherbrand Microscope Slides) that were left untreated to provide a completely wetting surface, with contact angle \( \theta_0 = 0^\circ \), or glass slides covered with an oleophobic coating (Fussode Coat, \( \theta_0 = 42 \pm 2^\circ \)). Similarly, the water-glycerol drops impacted either clean glass (\( \theta_0 = 36 \pm 3^\circ \)), glass coated with indium tin oxide (\( \theta_0 = 79 \pm 4^\circ \)) or a hydrophobic coating (RainX, \( \theta_0 = 90 \pm 3^\circ \)). A wetting substrate was achieved for the water-glycerol by pre-wetting the glass slide with the same mixture. A liquid drop fully wets such a prepared slide (\( \theta_0 = 0^\circ \)), however the coating is thin enough not to change the splashing behavior. The changing of sub-

\[
\alpha = \begin{cases} 
0 & \text{in gas phase} \\
(0,1) & \text{on interface} \\
1 & \text{in liquid phase}
\end{cases}
\]
The continuity equation:

\[ \vec{v}_\text{avg} = \nabla \cdot (\rho \vec{v}) = 0, \]

where \( \vec{v} \) is the phase averaged velocity, and \( \vec{v}_{lg} \) is a velocity field suitable to compress the interface. This equation is equivalent to a material derivative, but rewritten to minimize numerical diffusion.

The phase parameter is used to calculate the phase averaged density, \( \rho \), velocity, \( \vec{v} \), and viscosity, \( \mu \), which are used in the momentum balance:

\[
\frac{\partial \rho \vec{v}}{\partial t} + \nabla \cdot (\rho \vec{v} \otimes \vec{v}) = -\nabla p + \nabla \cdot (\mu \nabla \vec{v}) + \rho \vec{g} - \vec{f},
\]

and the continuity equation:

\[ \nabla \cdot \vec{v} = 0. \]

In the above equations \( t \) is time, \( p \) is pressure, \( g \) is gravity, \( \vec{f} \) is any body force, like the surface tension force, and \( \otimes \) is the dyadic product. To complete the VOF model, an expression is needed to calculate the surface tension force \( \vec{f}_{st} \), and a model is needed for the contact line. The surface tension force is calculated using the expression:

\[ \vec{f}_{st} = \sigma_{st} \kappa \nabla \alpha, \]

where \( \sigma_{st} \) is the surface tension coefficient, and \( \kappa \) is the curvature of the interface.

The effect of varying the Young’s angle \( \theta_0 \) from 0° to 180° is calculated directly through the generalized Navier boundary condition at the impact wall. With this boundary condition the dynamic contact angle \( \theta \) is allowed to vary freely, but a restoring line-tension force is applied at the contact line whenever the dynamic angle deviates from \( \theta_0 \). This restoring force is an additional source term in the Navier-Stokes equations, and has the following form:

\[ \vec{f}_l = -\frac{\sigma}{h} \cos \theta_0 \nabla_{2D} \alpha \]

In the above equation \( \sigma \) is the surface tension coefficient, \( h \) is the height of the local grid cell, and \( \nabla_{2D} \alpha \) is the gradient of \( \alpha \) on the wall. This force is applied on the liquid-gas interface in the first grid cells adjacent to the wall and is balanced by the surface tension force when \( \theta \) is equal to \( \theta_0 \). Away from the contact line the used implementation of the generalized Navier boundary condition reduces to the Navier-slip boundary condition. Using this slip boundary condition gives a good approximation for the thin film behavior at the wall. Because the model used can accommodate only one value for the slip length, a value of \( \lambda = 1nm \) is chosen to be able to describe the contact line accurately. However, in practice the effective slip length is on the order of the mesh size of 10nm. This results in the gas film potentially closing faster in our simulations than if the slip length were truly 1nm.

The simulations are performed for ethanol in air using the VOF solver of the OpenFOAM Finite Volume toolbox at up to 10nm resolution at the wall. More information on the boundary conditions at the contact line can be found in Ref. More information on the equations, initial conditions, and a comparison with experiments can be found in Ref. In this paper it is shown that the scaling of the gas film height as a function of impact velocity is consistent with theory and experiments, and multiple experimental observations are reproduced, including the formation of the central air bubble, liquid sheet formation, and contact line instability.

To reduce memory requirements, we consider an ethanol (\( \mu = 1.1cP \)) droplet with a diameter of 300μm, as opposed to the 3nm more viscous droplets used in the experiments. This results in the non-dimensional numbers shown in Tab. As can be seen in this table the non-dimensional numbers for the whole droplet are quite different between the experiments and simulations. However, the focus of this work is on the capillary number at the contact line/edge of the droplet, which, as is shown below, is of the same order between simulations and experiments. In addition, as the splashing threshold has been shown to scale across a wide range of parameters, comparisons with experiments should not be compromised.
Table I. An overview of the non-dimensional numbers and ratios of liquid and gas properties under conditions of our experiments.

| Material                | Re  | We  | $\rho_l/\rho_g$ | $\nu_l/\nu_g$ |
|-------------------------|-----|-----|----------------|---------------|
| Silicone oil (9.4 cP)   | 1106| 1759| 0.6852         | 927           |
| Silicone oil (32 cP)    | 335 | 1762| 2.262          | 956           |
| Water & glycerol (32 cP)| 418 | 686 | 1.825          | 1191          |
| Ethanol                 | 986 | 264 | 0.1028         | 789           |

Figure 3. Time of thin-sheet creation vs. ambient gas pressure for droplet impacts on glass slides with different wetting angles. Image (a) shows the impact of 32cP water-glycerol drops on glass slides with $\theta_0 = 0^\circ$ ($\bigcirc$), 36$^\circ$ ($\bigcirc$), 79$^\circ$ ($\square$), and 90$^\circ$ ($\triangle$). $t_{\text{sheet}}$ is independent of wetting. Image (b) shows the impact of of 9.4cP silicone oil drops on glass slides with $\theta_0 = 0^\circ$ ($\bigcirc$) and 42$^\circ$ ($\square$), and of 32cP silicone oil drops on glass slides with $\theta_0$ of 0$^\circ$ ($\triangle$) and 42$^\circ$ ($\square$). The small differences in $t_{\text{sheet}}$ with wetting properties are within error.

III. RESULTS

A. Experiments

Varying the surface wettability does not affect $t_{\text{sheet}}$, as shown for water-glycerol drops in Fig. 3 (a). Notably, the onset of thin-sheet creation is surprisingly independent of changes in surface properties. Not only does a hydrophobic coating fail to change $t_{\text{sheet}}$, but even coating the glass slide with a thin layer of water-glycerol yields the same result. Similarly, no noticeable effect of wetting can be seen for silicone oil drops in Fig. 3 (b), where the substrate was changed from fully wetting with $\theta_0 = 0^\circ$ to partially wetting with $\theta_0 = 42^\circ$.

Fig. 4 compares the threshold pressure for the different substrates. As the ambient pressure is decreased, $t_{\text{sheet}}$ is delayed and the resulting thin-sheet is diminished. Consequently, below a pressure $P_{\text{splash}}$, the thin-sheet is too small to break apart into secondary droplets and splashing is suppressed. If the ambient pressure is further decreased below $P_{\text{sheet}}$, the thin-sheet is never formed. We find that both $P_{\text{sheet}}$ and $P_{\text{splash}}$ are independent of $\theta_0$. This result is similar to the velocity threshold in Ref. 20, which was also independent of $\theta_0$ for $\theta_0 < 90^\circ$.

B. Simulations

The simulation results shown in Fig. 5 compare the shape of the air-liquid interface for wetting and non-wetting surfaces at different stages of impact. The interface has a thickness of about 20nm, which is two times the highest resolution of the simulations. To facilitate comparisons with experiments, velocity is non-dimensionalized as the capillary number $Ca = \mu V/\sigma$. At all times for which $Ca \geq 1$ the interface is the same on both surfaces and, within the resolution of the simulations, the microscopic contact angle in both cases is 180$^\circ$. The observed gas film is about 20nm thick.

Only for times at which $Ca < 1$ do the interfaces begin to look different; the contact angle on the non-wetting surface remains 180$^\circ$, while the contact angle on the wetting surface decreases and would converge to its equilib-
rium Young’s angle, $\theta_0 = 0^\circ$, if the simulation were to run long enough until the drop is stationary.

A consequence of the lower $\theta$ in the wetting case at low $Ca$ is a thicker gas film at the contact line compared to the non-wetting surface, as can be observed most clearly in Fig. 5(e). Independent of the wetting properties, the gas film thickness is greatest when the air-liquid interface becomes parallel to the substrate (cf. Figs. 5(c-e)). The lower $\theta$ at the contact line of a wetting substrate requires a longer arc length to satisfy this condition, which results in a thicker gas film. Additionally, a greater separation between the liquid sheet and the substrate stabilizes the contact line. At early times, the gas film in front of the contact line periodically collapses and the liquid touches down on the substrate, entraining air bubbles (Fig. 5(d)). The contact line on the non-wetting surface never stabilizes and bubbles are entrained throughout the simulation. In contrast, these touch-down events are no longer observed at late times on the wetting surface.

A quantitative description of the differences in contact line behavior for splashing on wetting versus non-wetting surfaces is provided in Fig. 5(a) by plotting the microscopic contact angle as function of the capillary number for wetting and non-wetting surfaces. In agreement with Fig. 5 for $Ca > 1$ both wetting and non-wetting surfaces show the same contact angle of $180^\circ$. When the contact line slows to $Ca < 1$ the non-wetting surface continues to exhibit $\theta = 180^\circ$, while the wetting surface exhibits a contact angle that decreases with $Ca$.

The change of behavior at $Ca \approx 1$ is shown via the standard deviation of the microscopic contact angle in Fig. 5(b). For the non-wetting surface $\theta = 180^\circ$ at all times, therefore the fluctuations are small and independent of the capillary number. For the wetting surface at large capillary numbers the fluctuations are also small, however shortly before $Ca = 1$ they begin to grow dramatically as $Ca \to 1$. This behavior is reminiscent of a phase transition, where fluctuations increase around the critical point. Taking this analogy further, this suggests that not only does the contact angle change for $Ca < 1$, but, more importantly, the flow enters a different flow regime. Note that this result is unrelated to the aforementioned touch-down events, which are instabilities of the apparent contact angle.

Fig. 5 shows the horizontal and vertical liquid sheet ejection velocities, measured at the rim of the liquid sheet. Fig. 5 shows that a cusp forms in the interface at the same time for both wetting and non-wetting surfaces. Consequently, one can expect the liquid sheet, which forms promptly after the cusp can be detected, to be ejected at the same velocity. This is confirmed in Fig. 5 which demonstrates that the liquid sheets are ejected at the same angle, independent of wetting properties. As time progresses the figures show that the trajectories of the liquid sheet are nearly identical for the wetting and non-wetting case.
IV. DISCUSSION

The key to understanding the unusual contact line behavior in splashing is in contrasting it with that of slow-moving contact lines. A stationary contact line will approach a homogeneous surface at an angle $\theta_0$ that is determined purely by wetting properties. If the contact line is forced to move, as in the classic case of a solid being plunged into a liquid bath, the shape of the interface will be determined by both the capillary number, which describes the balance between surface tension and viscous forces, and gravity. In addition, the assumption is made that the contact line is moving slowly enough that the microscopic contact angle is independent of the capillary number and is equal to Young’s angle $\theta_0$, which now serves as a boundary condition at the surface. For advancing contact lines this results in a critical capillary number at which this boundary condition cannot be satisfied. Consequently, theory predicts that above this critical capillary number wetting failure will be observed in the form of air bubble entrainment at the contact line. Additionally, it is predicted that the critical capillary number depends on the wetting properties of the surface.

The splash of the liquid drop occurs in multiple stages. Shortly before impact, the bottom surface of the drop is deformed by the rising gas pressure in the decreasing gap between the liquid and solid. When the liquid makes contact with the substrate, the air directly beneath the drop is trapped into a small bubble confined to the center of the deposited liquid, while the liquid begins spreading radially outward in the form of an axisymmetric...
ric lamella. Our simulations of lamella creation, described in more detail in Ref. [22], are consistent with the predictions made in Refs. [11] and [9]. The contact line moves fastest immediately after impact, and proceeds to rapidly decelerate as shown in Fig. 2. Fig. 2 further reveals that at the moment of thin-sheet formation the capillary number (on the right axis) of the contact line is in the unstable regime: at atmospheric pressure \( t_{\text{sheet}} = 0.21 \text{ms} \) with \( \text{Ca}(t_{\text{sheet}}) = 2.9 \). Therefore at all times between impact and sheet creation \( \text{Ca} > 2.9 \).

The time of thin-sheet creation, \( t_{\text{sheet}} \), varies with multiple parameters, most importantly with the ambient gas pressure. However, in all cases we find that thin-sheet creation occurs when \( \text{Ca}(t_{\text{sheet}}) \geq 1 \). Indeed, for the points shown in Fig. 3(a) a thin-sheet is created with \( 2 < \text{Ca}(t_{\text{sheet}}) < 7.5 \), for the 32 cP drops in Fig. 3(b) \( 1.6 < \text{Ca}(t_{\text{sheet}}) < 5.1 \), and for the 9.4 cP drops in Fig. 3(b) \( 0.7 < \text{Ca}(t_{\text{sheet}}) < 3.0 \). A wide range of parameters was investigated in Ref. [10]. Impact velocity, drop size, surface tension, density, surface tension, viscosity of both the liquid and the gas, and the gas molecular weight were varied. It was invariably found that splashing can occur only when the contact line is moving at a large \( \text{Ca} \).

The high resolution of our simulations allow us to determine that at such high \( \text{Ca} \) the contact line is advancing via a "rolling" motion. In both the wetting, \( \theta_0 = 0^\circ \), and non-wetting, \( \theta_0 = 180^\circ \), case, an ultra-thin air gap extends underneath the drop, as can be seen in Fig. 3(a-c). This is equivalent to a dynamic contact angle \( \theta = 180^\circ \) that is independent of the static wetting properties. In other words, splashing is independent of wetting, because splashing occurs at large \( \text{Ca} \), at which the static wetting properties do not influence the shape of the contact line. Although we can identify this air film in simulations, in experiments we are only able to detect the thicker gas film that is present after \( t_{\text{sheet}} \). However, the ultra-thin air film was already experimentally observed by Kolinski et al. via total internal reflection measurements and is consistent with the analysis in Ref. [4].

The presence of this air film can have a profound effect on drop impact. If the substrate is sufficiently smooth and the impact velocity sufficiently low, the ultra-thin air persists underneath the drop and isolates it from substrate, so that a drop can rebound from a wetting substrate, as if it were super-hydrophobic. However, drops splash at much higher impact velocities. In this case, Kolinski et al. observed that the air film behind the advancing contact line was closed within several microseconds, and attributed the effect to interactions between the liquid and solid. This is consistent with the observation that while splashing, which originates directly at the contact line, is independent of the wetting properties for both contact angles smaller than \( 90^\circ \) and larger than \( 90^\circ \), the slower dynamics following splashing are still determined by the static contact angle. For example, the maximum spreading diameter of an impacting drop does depend on the properties of the solid.

We emphasize that the wetting independence of splashing is a direct result of contact line dynamics. While our simulations agree with Ref. [5] with respect to the presence of a boundary layer and also find a characteristic "rolling" motion of the contact line, we find, in agreement with Ref. [17] that these phenomena result from the rapid motion of the contact line.

The behavior of the splashing contact line at high \( \text{Ca} \) provides a means of testing splashing theories, such as the model recently proposed by Riboux and Gordillo or by Liu et al. In the Riboux and Gordillo model the wetting properties influence the calculated lubrication lift force on the edge of the spreading liquid. As the liquid edge rises, its rim increases in size due to surface tension and, consequently, the bottom surface of the rim is forced downward. Depending on which of these effects dominates, the lamella either continues moving upward and eventually breaks apart to form a splash, or rewets the substrate, which prevents splashing. The lubrication force calculated is dependent strongly on the shape of the advancing contact line set by the microscopic contact angle, (c.f. footnote [35] in Ref. [5]). Consequently, the theory predicts the splashing threshold to strongly depend on the wetting properties of the substrate; this is inconsistent with the results presented here from experiments and simulations. Furthermore, this model predicts that splashing occurs at time \( t_{\text{e,crit}} \) after impact, calculated from Eqn. 1 in Ref. [5]. In contrast, we find that in all cases \( t_{\text{sheet}} > t_{\text{e,crit}} \). The disparity is most evident for drops of higher viscosity near threshold pressure, for which \( t_{\text{sheet}} \) is largest. For these drops, \( t_{\text{e,crit}} \), which does not depend on pressure, is smaller than \( t_{\text{sheet}} \) by several orders of magnitude. Together, these results suggest that the microscopic basis of this and similar theories should be revisited. In contrast, Liu et al. propose that splashing is caused by the Kelvin-Helmholtz instability in the air film that was observed in Ref. [17] and in our simulations. While our results are not a direct test of this model, they are consistent with its implicit contact angle independence.

V. CONCLUSION

Splashing arises from the interaction of three phases: the liquid drop, the solid substrate, and the ambient gas. It is therefore surprising that the most basic measure of this interaction, the contact angle \( \theta_0 \), does not influence the outcome of drop impact for the impact parameters we have investigated. Our experiments show that both the time of the splash, as well as the splashing thresholds are independent of \( \theta_0 \). Direct numerical simulations allow us to probe the advancing liquid-gas interface at nm length scales and show that the shape of this interface is the same for \( \theta_0 = 0^\circ \) and \( \theta_0 = 180^\circ \).

Splashing occurs when the liquid is spreading rapidly across the substrate, at capillary numbers \( \text{Ca} > 1 \). In this regime, both experiments and simulation suggest...
gest that the advancing contact line spreads over a short-lived thin film of air. Understanding the dynamics of this air film, both its rapid growth at $t_{\text{sheet}}$ (where we define $t_{\text{sheet}}$ as the time when the thin sheet first starts to appear at the front of the expanding lamella) that leads to splashing, and how $t_{\text{sheet}}$ is set by ambient pressure, is crucial to forming an accurate model of splashing.

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