Dense Suspension Splat: Monolayer Spreading and Hole Formation After Impact

Luuk A. Lubbers,1,2 Qin Xu,3 Sam Wilken,1 Wendy W. Zhang,1 and Heinrich M. Jaeger1

1 James Franck Institute and Department of Physics, The University of Chicago, Chicago, Illinois 60637, USA
2 Physics of Fluids Group, University of Twente, P. O. Box 217, 7500 AE Enschede, The Netherlands

We investigate experimentally and numerically the evolution of dense suspension drops that collide against a smooth solid surface and flatten into a rapidly expanding monolayer. This creates a lace-like pattern of particle clusters separated by particle-free regions. Both the expansion dynamics and the development of the spatial inhomogeneity are captured quantitatively by simple models derived from balancing forces acting on individual particles.

PACS numbers: 82.70.Kj, 45.70.Qj, 82.70.Dd, 47.57.Qk, 47.57.Gc

Since the pioneering work by Worthington [1] the spreading of liquids droplets upon impact has remained an active research area [2–4]. One reason for its enduring appeal is its intricacy: even a quantity as basic as the maximum extent of a water splotch on a glass surface is controlled by a nonlinear interplay of macroscopic flow effects such as inertia and viscous drag, microscopic properties such as substrate wettability and contaminants. A trace amount of small dust particles can alter the surface tension by collecting in a local region at the water surface and thence give rise to a Marangoni flow that alters the final splat size [5–7]. Larger suspended particles pin the contact line, again changing the spreading dynamics [8].

Here we examine post-impact spreading of suspensions of rigid, non-Brownian particles at high volume fraction (60% or above). This high concentration regime has so far received little study [9]. Our experiments show that their presence produces a qualitatively different outcome (Fig. 1): the plug flattens into a splat comprised of a single layer of close-packed particles immersed in a thin liquid layer. As the splat expands, void-like regions appear and grow, causing the final splat to display a lace-like pattern of particle clusters separated by particle-free regions. We develop particle dynamics models for the splat expansion as well as the instability and obtain good agreements between the model predictions and the measurements.

While the spreading dynamics is a sensitive function of wettability, particle concentration and surface tension in the previously examined, pure liquid or dilute particle concentration regimes, the splat expansion dynamics in the high concentration regime is robust. Changing the substrate wettability, surface tension and liquid viscosity introduces small perturbations. The single-particle dynamics governing monolayer splat expansion also contrasts with the complex rheological response exhibited by dense suspension in shear [10–14], especially while confined [15–17]. This is because the impact induced expansion rapidly switches off interactions among multiple particles.

Our results are relevant to determining the cohesive strengths of colloidal semiconductor quantum dots by measuring their maximal splat size after impact [18] as well as applications such as thermal spray coating of sintered powders [19] and additive manufacturing using inkjet printing [20, 21]. Splat formation in these processes, as in our experiment, is shaped by inertia being much larger than surface tension, thermal agitation and viscous drag.

FIG. 1. (Color online) Dense suspension impact, splat, and instability. (a) Side view: A cylindrical plug impacts a smooth dry glass surface, splashes by ejecting particles upwards and flattens into a monolayer. (b) Bottom view: The initial, nearly circular splat expands. Inhomogeneities appear as regions of particle clusters separated by particle-free regions (dark). (c) Close-up: clusters drag streaks of liquid along as they move outwards. (d) Substrate area coverage as function of time. Once the covered area approaches a constant value (shaded region), particles are spread out in a monolayer.
Experiments— Dense suspensions were made by adding spherical ZrO₂ particles (\(d = 250 \pm 22 \mu m\), \(\rho_p = 5.68 \times 10^3 \text{kg} \cdot \text{m}^{-3}\)) to water or silicon oils. Letting the particles sediment inside a straight cylindrical syringe produces packing fractions of \(\phi = 0.61 \pm 0.02\). As gravity pulls the suspension down, a pinch-off occurs below the cylinder opening [22, 23]. In the dense suspension limit studied here, the plugs preserve the cylindrical shape of the syringe, resulting in a plug radius \(R_p \approx 2.25 \pm 0.05 \text{mm}\), and have a height \(L \sim 2R_p\). The substrate was a smooth, horizontal glass plate \(1.6 \pm 0.03 \text{m}\) below the syringe.

Figure 1 shows typical image sequences of the impact, recorded by high-speed video. We denote \(t = 0\) as the moment when a monolayer first forms. Before this moment the cylinder-shaped plug flattens into a single-particle layer. This time point can be defined precisely by viewing plug impact onto a transparent glass slide from below and plotting the substrate coverage area as a function of time (Fig. 1(c)). The transition to constant area indicates the monolayer onset.

After \(t = 0\), the monolayer expands radially and develops holes. We measure the expansion by azimuthally averaging the particle density as a function of radial distance. The splash’s edge \(R(t)\) corresponds to the sharp transition zone from high density to zero density. Fig. 2(a) plots \(R(t) - R_0\), the difference between the splash radius and the initial monolayer radius \(R_0\). We have also measured the velocity field \(U_r(r)\) by azimuthally averaging the particle motion throughout the splash (Fig. 2(b)). These measurements reveal an internal straining flow, starting at 0 velocity at the small dead zone of immobile particles at the center of impact (shaded region). At later times, this internal straining flow weakens but retains its form. Finally we quantify the time evolution of the spatial inhomogeneity in terms of the area fraction in the splash occupied by the particle-free regions. Due to the linear straining flow, the instability grows fastest near the outer edge and slower in the interior. To capture this trend, we divide the splash into an inner and outer annulus that contain approximately the same particles over time, and plot the average area fraction of void regions within each annulus as a function of time. Fig. 4(b) shows that the measured void fraction initially grows rapidly, then slows and saturates.

In our experiments, the particle-based Weber number \(W_e = \rho_p U_0^2 d/\gamma\), where \(U_0 = \dot{R}(t = 0)\) is the initial expansion speed for the monolayer, and the Stokes number \(St = \rho_p d U_0/\mu\) are both large. Using speeds at the expanding edge, the water and silicone oil suspension impacts featured in Fig. 2(a) have \(W_e \approx 520\) (water) and 1900 (silicone oil), and \(St\) values of 7400 (water), 4000 (1.8 cSt oil) and 800 (10 cSt oil). Moreover, the rate of strain is so large that \(W_e\) and \(St\) are both much larger than 1 over almost all the splash interior. The dot-dash line corresponds to \(W_e = 1\), the line \(St = 1\) lies even nearer to the x-axis.

Splat Expansion— Given \(W_e \gg 1\) and \(St \gg 1\) we consider the following particle-inertia dominated model. We assume the splash expands as fast as particles at the splash’s leading edge can move and that these particle motions are unaffected by collisions. As a result, the only forces acting on the leading-edge particles are surface tension and drag due to motion relative to the liquid layer and/or the solid substrate. First, to estimate the force due to surface tension, we note that in the experiments the particle remains fully coated by the suspending liquid throughout the expansion. This gives rise to an asymmetric free surface shape: a thin liquid film coats the front surface of the particle at the outermost edge while the rear half of the particle remains immersed in a thicker liquid layer (Fig. 3). This yields a retarding force \(F_\gamma = \alpha \pi \gamma d / 2\). Because the value of the constant \(\alpha\) depends on the free-surface asymmetry, it varies from par-
force critical separation $s$ chain model for splat instability particle-free regions emerge from variations in the initial radial velocity field. Beyond a critical separation $s_c$ between adjacent particles, bridge-like menisci transform into trailing liquid streaks and the force switches from a bridging force $F_b$ to a trailing streak resistance $F_c$ acting solely on the faster moving particle in front. The model also produces good agreement with data from measurements. Second, the drag experienced by a leading-edge particle moving outwards with speed $R(t)$ has several distinct contributions. Measurements suggest the dominant contribution $F_\mu$ is viscous drag due to a thin trailing liquid streak [25]. If the average liquid layer thickness in the splat is $h$, then the average viscous stress experienced by the particle as it drags a liquid streak along is $\mu R/h$. If we assume in addition that this viscous stress acts over $\pi d^2/4$, the projection of the drop surface area in the direction of motion, then $F_\mu$, the total drag due to the trailing streak, is $(\mu R/h)(\pi d^2/4)$. Comparison with measured data presented later will show that this expression gives a quantitatively correct description for splat expansion at high liquid viscosities.

Requiring $ma = F = -F_\gamma - F_\mu$ where $m$ is the particle mass and $a$ its acceleration at the leading edge yields an evolution equation for the splat radius $R(t)$:

\[ \rho \left( \frac{\pi d^3}{6} \right) \frac{dR}{dt} = - \left( \frac{\mu R}{h} \right) \left( \frac{\pi d^2}{4} \right) - \alpha \gamma \frac{\pi d}{2}. \]  

(1)

Since the volume of liquid inside the suspension is conserved over time and the liquid layer is much thinner than the particle diameter, the unknown liquid layer thickness $h(t)$ is directly related to $R(t)$ via $(1 - \phi)V_p = \pi R^2(t)h(t)$, where $V_p = \pi R_p^2 L$ is the volume of the suspension plug.

FIG. 3. (Color online) In the leading-edge model for splat expansion, the splat edge, initially ejected with horizontal speed $R(t = 0)$, slows over time due to resistance by surface tension $F_\gamma$ and drag force $F_\mu$ from a trailing liquid film. In the chain model for splat instability particle-free regions emerge from variations in the initial radial velocity field. Beyond a critical separation $s_c$ between adjacent particles, bridge-like menisci transform into trailing liquid streaks and the force switches from a bridging force $F_b$ to a trailing streak resistance $F_c$ acting solely on the faster moving particle in front.

Slowed by viscous drag. This shows that the proposed expression for $F_\mu$ is quantitatively accurate. As far as we are aware, this simple drag law has neither been proposed nor tested against data in previous studies.

**Splat Instability**—Inspection of the magnitudes of forces in the leading-edge model shows that the monolayer splat regime is characterized by large particle inertia, small surface tension and small viscous drag effects. This in turn suggests a physical mechanism for the observed spatial inhomogeneity: the particle-free regions originate as small velocity and packing inhomogeneities within the initially close-packed monolayer splat. Subsequent rapid expansion of the splat amplifies the inhomogeneities, creating a lace pattern. This mechanism is qualitatively different from capillarity induced aggregation, which proceeds much more slowly [27].

A one-dimensional (1D) model based on this scenario gives reasonable agreement with measured growth rates for the spatial inhomogeneity. The model considers a chain of $N$ particles which lie along a ray emanating from the center of the splat (Fig. 3). Because surface tension and viscous drag merely perturb the dominant inertial motion, simple approximations will turn out to be sufficient for a quantitatively accurate description. Specifically, each particle in the chain experiences viscous drag $F_\mu = (\mu R_i/h)(\pi d^2/4)$ where $R_i$ is the speed of the $i$th particle in the chain. Initially the particles in the chain are closely packed together and each experiences cohesive capillary forces $F_\gamma$ with neighbors ahead of and behind itself. We approximate $F_\gamma$ as the cohesion exerted by an axisymmetric, static liquid bridge connecting two fully wetted spheres [25, 28] (This is a rough upper bound on the actual forces, because the liquid surface in the monolayer only bridges the top surfaces of the particles and because particles are in rapid relative motion so the liquid surface is less curved than a static calculation indicates). As the gap between the neighboring particles exceed a critical value $s_c$, the cohesive capillary interaction switches off ($F_\gamma = 0$). Instead, motivated by images from the experiment showing faster moving particles leaving streaks of liquid behind themselves, we require that a particle far ahead of its neighbor in the chain model experiences a retarding force due to surface tension $F_\gamma = \alpha \pi \gamma d/2$, while the left-behind neighbor no longer is pulled forward force by a liquid bridge.

The dashed line in Fig. 2(a) gives the position of the outermost particle in the N-particle chain and agrees well with the measured evolution. Comparisons with the other two systems also show good agreement and are given in [25]. Finally, we calculate the void fraction evolution from the chain model and plot the results in 4(b). The agreement is also good. More importantly, the calculated instability dynamics is robust when changing model parameters. Altering the value for $s_c$ by 40% from $d/4$ used in generating the chain model result presented in 4(b), or using an initial velocity fluctuation that
is half, or double the 10% value used, produces negligible changes.

**2D Simulation of Instability**— We next refine the chain model by going to 2D by prescribing the same $F_\mu$ but generalizing $F_b$ and $F_c$ (inset to Fig. 4(a) and [25]) to include capillary interactions with all nearest neighbors, not only those along a radial direction. The inset in Fig. 4(b) shows two snapshots from the simulation: initially the splat is so densely packed that it appears uniformly black. As the expansion proceeds, voids appear and grow, with the growth rate being faster in the outer regions. In Fig. 4(b) we also plot the void area fraction calculated from the simulation. Including the interaction with azimuthal neighbors allows the 2D simulation to track the initial void growth rate more accurately than the chain model. This results in a noticeably better fit to the measured evolution.

The simulation also allows us to test our starting assumption that the radial expansion causes the particle dynamics in the monolayer splat to be decoupled, thereby rendering the splat evolution simple. In Fig. 4(a) we plot $\bar N_b$, the average number of nearest neighbors experiencing cohesive capillary interaction, as a function of normalized radial distance. The different curves correspond to different radial strain $\epsilon = [R(t) - (R_0 - R_{DZ})]/(R_0 - R_{DZ})$, with $R_{DZ}$ the radius of the dead zone. This quantity $\bar N_b$ is difficult to extract from the experiment but gives direct insight into the degree of collective interactions present. Initially particles everywhere in the splat are closely packed and have on average 5.5 neighbors. As the radial expansion proceeds and the radial strain grows large, many particles lose cohesive capillary interactions with nearest neighbors, particularly those along the azimuthal direction. This effect is most pronounced near the outer edge, where the expansion speed is the largest. Finally, as the monolayer splat expansion slows, a large outer area in the splat are occupied by particles experiencing one cohesive capillary bond on average.

**Conclusions**— The monolayer splat analyzed here forms from macroscopic particles moving at several meters-per-second, and thus falls in the same $We_p \gg 1$ and $St \gg 1$ regime seen in many technologically relevant applications [18–21]. The high impact speeds used in these processes ensure that particle inertia remains important despite the smaller particles used. Increasing the viscosity of the suspending liquid so that the particle dynamics becomes overdamped should halt the expansion and therefore the lace instability. Reducing the impact speed so that surface tension dominates should cause the expansion to be dominated by the motion of cohesive particle clusters rather than the outermost particle. Other parameter changes raise more complex questions, such as how the monolayer dynamics change when the splat becomes several particles deep, or whether a qualitatively different dynamic appears at lower volume fractions due to long-range viscous flow coupling [29–32]. The findings presented here provide a solid starting point for tackling these issues.

**Acknowledgements**— We thank I.R. Peters and M.Z. Miskin for discussions. We also thank D. Lohse and J.H. Snoeijer for bringing Chicago and Enschede together. This work was supported by NSF through its MRSEC program (DMR-0820054) and fluid dynamics program (CBET-1336489).

FIG. 4. (Color online) Instability dynamics. (a) Average number of capillary-bridge bonds per particle, $\bar N_b$, as function of $\tau/R(t)$, the radial distance normalized by current splat radius, for different values of radial strain $\epsilon = [R(t) - (R_0 - R_{DZ})]/(R_0 - R_{DZ})$. Inset: 2-dimensional generalization of $F_b$ and $F_c$. A cohesive capillary bridge bond between neighboring particles becomes a trailing streak if the neighboring particle lies outside a wedge of opening angle $2\delta$ and radius $(d/2) + s_b$ (shaded region). (b) Area fraction of particle-free regions in circular annuli within the splat as a function of time. The boundary between the inner and outer annuli is chosen to lie at $\rho U_b d/\gamma = 150$, where $U_b$ is the initial speed of the particle at the boundary. Experiments (● ▲), one-dimensional chain model (solid and dashed lines), and two-dimensional numerical model (○ △) agree quantitatively. Inset: snapshots from the simulation.


The value of $\alpha$ is insensitive to the contact line dynamics. Impacts using plugs suspended in silicone oil show the same expansion dynamics as those suspended in water even though oil wets the dry glass substrate but water does not. This insensitivity arises because the region of the liquid surface affected by the substrate wetting properties corresponds to a very small portion of the full area when the liquid motion Weber number are large and $We_p \gg 1$.

See the details in the Supplemental Material.

Varying $\alpha$ by 10% still allows a fit within the error bar range of experiments.