Coexistence of solid and liquid phases in impacting colloidal drops

P. Shah\textsuperscript{1,\dagger}, S. Arora\textsuperscript{1,\dagger}, M.M. Driscoll \textsuperscript{1,*}

\textsuperscript{1}Department of Physics and Astronomy, Northwestern University

\dagger These authors contributed equally to this work.

*corresponding author

Abstract

Complex fluids exhibit a variety of exotic flow behaviours under high stresses, such as shear thickening and shear jamming. Rheology is a powerful tool to characterise these flow behaviours over the bulk of the fluid. However, this technique is limited in its ability to probe fluid behaviour in a spatially resolved way. Here, we utilise ultrahigh-speed imaging and the free-surface geometry in drop impact to study the flow of colloidal suspensions, and for the first time observe coexisting solid and liquid phases. In addition to observing Newtonian-like spreading and bulk shear jamming, we observe the transition between these regimes in the form of localised patches of jammed suspension in the spreading drop. We capture shear jamming as it occurs via a solidification front travelling from the impact point, and show that the speed of this front is set by how far the impact conditions are beyond the shear thickening transition.
1 Introduction

Complex fluids, such as particulate suspensions and polymer solutions, exhibit a variety of exotic flow behaviours, for instance shear thickening and solidification via jamming. These behaviours are particularly relevant to development of smart materials, such as body armours and soft robots. Rheometry is traditionally used to characterise complex fluids. However, this technique traditionally provides measurements averaged over the bulk of the fluid and obscures the information on local variations in flow. The free-surface geometry in drop impact systems offer a unique lens to probe these flow properties, as it provides spatially resolved data on the manifestations of non-Newtonian flow. Here, we use ultrahigh-speed imaging to study the drop impact of colloidal suspensions over a large range of volume fractions and impact velocities, thus sampling impact behaviour from liquid-like spreading to solid-like jamming. Combined with input from rheological data, our measurements offer a more holistic picture of complex fluid flow.

An extensive understanding has been developed for the dynamics of a Newtonian fluid drop impacting a dry solid substrate. However, the vastly different flow properties of complex fluids substantially modify impact dynamics. Past studies have explored the spreading and splashing of a variety of complex fluids, but each has largely focused on a relatively narrow slice of the vast parameter space — for example, a particular range of suspension volume fractions. Colloidal suspensions offer a convenient control parameter — volume fraction — to scan suspension behaviours ranging from Newtonian-like to shear thickening. Here, we report a systematic study of colloidal suspension impact spanning a wide range of volume fractions (0.09 $\leq \phi \leq 0.50$) and
For our experiments, we synthesize charge-stabilized silica spheres (diameter 830 ± 20 nm, Fig. 1a) using the Stöber process\textsuperscript{14, 15} and suspend them in water. Spherical drops of diameter $d_0 = 3.0 \pm 0.1 \text{ mm}$ are formed by drawing a known volume of fluid (15 µL) into a micropipette. We set the impact velocity by changing the height from which the drops are released, and record the drops impacting on a dry glass substrate using a high-speed camera. To minimize the effects of particle sedimentation, all samples are re-suspended immediately before the experiments using a vortex mixer. All experiments are performed in a humidity chamber, which additionally mitigates air currents (see Methods for details).

To connect impact behaviours with rheological properties, a mapping between impact velocity and rheological parameters such as shear rate or shear stress is necessary. Precisely quantifying shear rates in drop impact systems is challenging due to the nonuniformity of shear in both space and time. However, a simple dimensional argument can be used to estimate the shear rate at impact. At the instant of impact, the bottom point on the drop comes to rest, while the apex continues to fall at the impact velocity $u_0$, as the information of impact has not had time to propagate. Dividing this difference in speeds, $u_0$, by the drop size $d_0$ thus provides an estimate of the maximum shear rate experienced by the drop at the moment of impact: \( \dot{\gamma} = u_0/d_0 \). With the drop size of 3 mm, we could access shear rates in the range $233 \text{ s}^{-1} \leq \dot{\gamma} \leq 1333 \text{ s}^{-1}$. Thus, we are able to span a large spectrum of flow behaviours in these suspensions, and observe how non-Newtonian flow give rise to a rich variety of post-impact phenomena. The results we present here take us closer to an
understanding of the shear jamming transition and the properties of shear jammed solids.

2 Results

Bulk rheometry measurements [Fig. 1b] demonstrate the variety of flow behaviours exhibited by our suspensions. At low $\phi$ (black and pink lines), the fluid viscosity is constant, akin to a Newtonian fluid. Shear thinning (decreasing viscosity) becomes pronounced as $\phi$ is increased (green and purple curves), and shear thickening (indicated by increasing viscosity) appears for $\phi \geq 0.47$ at high shear stresses (orange, blue, and red curves). We observe fascinating consequences of this non-Newtonian rheology in our impact experiments. At $\phi = 0.47$ where weak shear thickening appears at high stresses in bulk rheology, we observe patches of localised solidification during spreading — panel 3 of Fig. 2a shows small solid-like bumps that protrude from the spreading drop, but vanish in panels 4 and 5 [SI video 1]. At higher $\phi$, we observe partial solidification of the drop — Panel 2 in Fig. 2b shows that the bottom part of the drop acts as a solid, while the top part remains fluid and flows over the solidified region throughout panels 3-5 [SI video 2]. Finally, at $\phi = 0.49$ and high impact velocities, most of drop solidifies as shown in Fig. 2c [SI video 3]. Here, we show that this variety of solidification behaviours is a direct consequence of shear jamming\textsuperscript{10}, evidenced by their occurrence much below the static jamming threshold\textsuperscript{2019controlling}.

We encapsulate this broad range of impact outcomes in a $\phi - u_0$ state diagram [Fig. 3]. Green circles, indicating simple spreading [SI video 4], dominate the low $\phi$ and low $u_0$ region. With increasing $\phi$ or $u_0$, the localised solidification regime appears (orange diamonds), followed
by the bulk solidification regime (blue triangles) where a larger and larger portion of the drop solidifies upon impact. The transition between these regimes is a function of both $\phi$ and $u_0$, as all regimes can be accessed by varying either of the parameters while keeping the other constant. Additionally, we find that the drop behaviour is very sensitive to small changes in $\phi$, consistent with the transition to shear thickening in rheological measurements [Fig. 1b].

To quantify this range of impact outcomes, we compute the normalized maximum diameter of the impacted drops, $\beta = d_{max}/d_0$, and plot this metric against $u_0$ [Fig. 4a]. For $\phi \leq 0.47$, $\beta$ increases with increasing impact velocity. However, $\beta$ drops to 1 at $\phi \geq 0.49$ and high impact velocities. This is because the drop no longer spreads after impact (lower inset). This result is consistent with recent studies that observed similar solidification in suspension impact at high $\phi$ [8, 11].

Our drops remain solid for a few milliseconds; however they spread like a liquid over the timescale of a second [SI video 5]. Thus, the solid-like state we observe is transient in nature, further evidence that this solidification is a direct result of shear jamming. A recent result suggests that the substrate wettability affects this timescale of unjamming [11], but this problem remains largely unexplored.

At $\phi \leq 0.47$, the drops spread in a manner qualitatively similar to Newtonian fluids [SI video 4]. Previous experiments [17] with Newtonian fluids have shown that $\beta$ scales as the dimensionless parameter $ReWe^{1/2}$, where $Re$ is the impact Reynolds number ($\rho u_0 d_0/\eta$, where $\rho$ is the fluid density and $\eta$ is the fluid viscosity) and $We$ is the Weber number ($\rho u_0^2 d_0/\sigma$, where $\sigma$ is the fluid surface tension). For impacting colloidal drops, the calculation for the Weber number remains
identical to Newtonian fluids, where we use the density of the colloidal suspension and the surface tension of the suspending fluid. Estimating the Reynolds number is less straightforward, however, due to non-constant fluid viscosity of complex fluids. Immediately after impact, the shear rate experienced by the drop is at its maximum ($\dot{\gamma} = u_0/d_0$). As the fluid spreads and slows down, the shear rate continuously drops to zero. We used the average of these two extremes, $\dot{\gamma}_{avg} = u_0/2d_0$, as the simplest estimate of shear rate throughout the spreading process. We then used the viscosity value corresponding to $\dot{\gamma}_{avg}$ from our bulk rheological data to calculate the effective Reynolds number, $Re_{eff}$ at each $\phi$ and $u_0$. In Fig. 4b, we plot $\beta$ in the spreading regime against $Re_{eff}We^{1/2}$; the dashed black line indicates the power-law fit:

$$
\beta = 0.81(Re_{eff}We^{1/2})^{0.164}.
$$

(1)

The exponent of this fit shows excellent agreement with that reported by Scheller et al.\textsuperscript{17} for viscous Newtonian fluids, $\beta = 0.61(ReWe^{1/2})^{0.166}$. Thus, our simple estimate of shear rate and in turn effective viscosity, provides a useful framework to quantify the maximum spread of impacting colloidal drops, even well into the shear thinning regime.

In the localised solidification regime [orange diamonds in Fig. 3, SI video 1] the bulk of the drop still spreads like a Newtonian fluid [Fig. 4b], but shear thickening is apparent via solidified patches that appear and then disappear after a few milliseconds. Our observation of this regime coincides with the onset of weak shear thickening in the bulk rheology data [orange curve in Fig. 1b]. Moreover, the transient nature of these patches is strong evidence that regions of high viscosity are embedded in a lower-viscosity fluid phase. However, for higher $\phi$ where shear thickening is pronounced, the drop does not spread due to bulk shear jamming.
For $\phi \geq 0.49$, a large fraction of the drop solidifies upon impact. However, further analysis reveals that the top portion of the drop remains a liquid. To quantify the dynamics of this partially solidified state, we measure the height of the tallest point on the drop as a function of time [Fig. 5a]. Consistent with another study of impacting shear-thickening drops, we observe two regimes in the $h$ vs. $t$ curve. Immediately after impact, $h$ decreases linearly with time, and then plateaus at a constant value $h_{\text{min}}$. The slope of the linearly decreasing regime, $u^*$, is identical to the impact velocity, $u_0$ [Fig. 5b]. This is strong evidence that any shear from the impact event has not yet propagated to the top portion of the drop, and hence the top portion must still remain a liquid. We quantify the spatial extent of solidification by plotting the normalized minimum height, $h_{\text{min}}/d_0$ against $u_0$ [Fig. 5c]. The increase in $h_{\text{min}}/d_0$ with $u_0$ indicates that a larger and larger volume of the drop is solidified as the impact velocity is increased. Interestingly, at high impact velocities, $h_{\text{min}}/d_0$ plateaus to a value smaller than 1, indicating that the solidified drop also undergoes compression along the impact direction. Furthermore, the high temporal resolution (100,000 fps) of our imaging enables us to capture the details of this solidification as it occurs.

Immediately after impact, we observe a disturbance travelling upward along the drop surface over hundreds of microseconds [orange and green arrows in Fig. 6a]. To better visualise this disturbance, we subtract successive frames of the image sequence, so that only the parts that change between frames are highlighted [right panel of Fig. 6a, SI video 6]. The location of the front is given by the lower end of the bright edge [Fig. 6b]. As this front travels upward, the portion of the drop above the front still maintains its pre-impact curvature [red circles in Fig. 6a], indicating that it is unaffected by the impact event until the front reaches it (consistent with $u^* = u_0$, Fig. 5b). The
angular location of this front plotted against time reveals that the front travels at a constant speed, \( u_{\text{front}} \) [slope of the line in Fig. 6c]. \( u_{\text{front}} \) increases with increasing \( u_0 \), and its value is several times larger than \( u_0 \) [Fig. 6d]. As evident from the rheology, the suspension thickens when the applied shear surpasses a critical value. Indicated by the dotted lines in Fig. 6c, the critical shear rate where thickening is observed, \( \dot{\gamma}_c \), is much lower for \( \phi = 0.50 \) than for \( \phi = 0.49 \). We plot \( u_{\text{front}} \) against the excess shear rate over this critical value, \( \dot{\gamma} - \dot{\gamma}_c \), and the data indeed collapses on a single curve for both \( \phi \) [Fig. 6f]. Thus, the speed of this disturbance is set by this excess shear rate.

As the impact velocity is increased, a larger and larger volume of the drop solidifies upon impact. Strikingly at \( \phi = 0.50 \) and \( u_0 = 4 \, \text{m/s} \), we observe the drop bounce off the substrate, with the coefficient of restitution \( \epsilon = 0.1 \) [SI video 7]. By coupling this coefficient of restitution with the drop’s compression along the impact axis, we can semi-empirically estimate the elastic modulus of the solidified drop. The drop impacts the substrate with an initial velocity \( u_0 \), remains in contact with the substrate for time \( \Delta t = 200 \, \mu\text{s} \) (measured from our high-speed videos), and then rebounds with the final velocity \( \epsilon u_0 \). While in contact with the substrate, we measure that the drop is compressed by the amount \( \Delta x = 0.24 \, \text{mm} \). We calculate the force experienced by the drop upon impact using momentum conservation:

\[
F = \frac{m \Delta u}{\Delta t} = \frac{m(1 + \epsilon)u_0}{\Delta t},
\]

To convert the force to a stress, we divide by the contact area for a Hertzian contact:\[18\], \( \pi a^2 = \pi d_0 \Delta x / 2 \):

\[
\sigma = \frac{F}{\pi d_0 \Delta x / 2} = \frac{2m(1 + \epsilon)u_0}{\pi d_0 \Delta x \Delta t},
\]
The strain experienced by the drop is \( \gamma = \Delta x/d_0 \). Thus, the elastic modulus of the rebounding drop can be computed as

\[
E = \frac{\sigma}{\gamma} = \frac{2m(1 + \epsilon)u_0}{\pi(\Delta x)^2 \Delta t}.
\]  

(4)

using \( m = 2.25 \times 10^{-5} \text{ kg} \), we find \( E = 5 \text{ MPa} \). A more thorough estimate using Hertz’s equations\(^{[13]}\) for two colliding elastic bodies lead to a similar estimate of \( E \). Calculating the elastic modulus in this way for other impact conditions is challenging, as measuring the contact time in the absence of rebound is nontrivial.

3 Discussion

In sum, our analysis presents the following picture of the drop dynamics. Upon impact, the drop experiences a very large shear stress at the impact point. At high enough volume fractions and impact velocities, this stress manifests itself as pockets of localised solidification embedded in the spreading liquid phase. At even higher volume fractions or stresses, a larger and larger fraction of the drop solidifies after impact, but some volume at the top remains liquid. Therefore, the shear front must be dissipating as it moves upward, and the stress falls below the critical stress for shear thickening before the entirety of the drop is solidified. Moreover, at the highest impact velocity, the drop rebounds, and the coefficient of restitution allows us to estimate the elastic modulus of the shear jammed solid, \( E = 5 \text{ MPa} \). Thus, our drop impact experiments provide a unique window to observe shear jamming as it occurs, and give rise to a number of questions about the nature of both the shear jamming transition and the resulting jammed solid.
The occurrence of localised solidification coincides with the appearance of weak shear thickening in our bulk rheology data. The fact that these solidified patches vanish over a few milliseconds is strong evidence that they are regions of high viscosity embedded in a lower-viscosity fluid phase. Recent rheological studies using boundary stress measurements (BSM) have reported finite regions of enhanced stress in silica suspensions\cite{19, 20}. In these works, Rathee et al.\cite{19, 20} argued that the transition from shear thickening to shear jamming is governed by the growing size of such localised shear jammed regions. Our observations of transient localized solidification are thus striking visual evidence of such a mechanism. Further spatially resolved stress measurements performed on impacting drops\cite{21} could provide more information on the nature of localised solidification in the absence of confinement.

In the bulk solidification regime, the nature of the upward-travelling front raises a number of interesting questions. Before the front reaches the top, the speed of the top part of the drop \( u^* \) is identical to \( u_0 \), and the curvature of the top portion is the same as it was before impact. This confirms that the information of the impact event reaches the top portion only with the front, thus establishing that it is a solidification front. Why the speed of this front is constant along the drop surface is a very intriguing question. One would expect a shear front to travel through the bulk of the drop, upward from the region in contact with the substrate. Given the visual nature of our measurements on an opaque drop, we can naturally observe this front only on the surface. The most likely explanation, therefore, is that the front we measure is this bulk shear front after it interacts with the drop boundary.
Our experiments are especially well-positioned to capture such a front due to the free-surface conditions here that are absent in other studies of shear fronts\textsuperscript{22-26}. Past work has established that shear fronts in dense suspensions are not a result of densification, and their velocity is set by the external driving speed\textsuperscript{25}. The dependence of the front speed on $\dot{\gamma} - \dot{\gamma}_c$ in our experiments suggests that suspension properties near the shear jamming transition are governed by the distance from the onset of shear thickening. This is consistent with measurements in static jamming, where material properties depend on the distance from the critical point\textsuperscript{27}. The functional form of this dependence potentially contains insights into the nature of the shear jamming transition. Numerical work exploring the impact of suspension drops, although incredibly challenging due to the strong role of hydrodynamics in colloidal systems, might provide crucial information in this respect. Unfolding the physics of these fronts will not only extend constitutive models for complex fluid rheology to much higher stress regimes, but will also help us understand more about the nature of the shear jamming transition.

Due to the transient nature of the shear jammed state, characterising the jammed solid that is created at impact is challenging. Using the coefficient of restitution of the rebounding drop, we were able to estimate the elastic modulus of the solid phase, $E = 5 \text{ MPa}$. As rebound only occurred at one impact velocity, how the elastic properties of shear jammed drops are controlled by the impact conditions remains obscure. The use of superhydrophobic substrates promotes rebound, even in Newtonian liquid drops\textsuperscript{28}. Further colloidal drop impact experiments on superhydrophobic surfaces could extend the parameter space where drops rebound, and thus provide the information essential to understand what controls the properties of this elastic state. Numerous other properties
of the shear jammed solid are of interest: When and how would such a solid fracture? How broad is its linear elastic regime? How do these properties compare to those of static jammed solids?

In conclusion, we conduct highly time-resolved drop impact experiments and systematically probe suspension flow ranging from Newtonian-like to shear jamming. We show that the impact behaviour in the spreading regime can be quantitatively understood via an effective viscosity framework, and that the solidification behaviours at high $\phi$ and $u_0$ are direct consequences of shear jamming. The free-surface geometry in our system provides direct visual information on how the shear jamming transition occurs, both in parameter space and in time. Shear jamming occurs via a solidification front, the speed of which is set by how far into the shear thickening regime the applied shear rate is. Furthermore, we see this transition occur via a localised solidification regime that cannot be observed via bulk measurements. We believe that drop impact is a powerful experimental tool to investigate macroscopic properties of complex fluids, and provides information that compliments the data from bulk rheometry.

4 Methods

Colloidal sample preparation

We fabricated silica spheres in our lab using the Stöber synthesis method. The particle size was determined by the number of feeds: we performed 14 feeds after the initiation of the reaction, resulting in particles with a diameter of $830 \pm 20 \text{ nm}$. The reaction mixture was centrifuged and re-suspended in ethanol 3 times; the suspension was then gravity separated to improve monodis-
persity. The particles were then imaged on the Hitachi S4800 Scanning Electron Microscope [Fig. 1a]. The particle size was characterized by measuring the diameter of a representative sample of 100 particles in ImageJ, and the polydispersity reported is the standard deviation in particle size.

A concentrated stock suspension of the silica spheres was prepared in water (with no surfactant), and the weight fraction was measured by drying 100 \( \mu L \) of the stock suspension. The density of silica (2 \( g/cm^3 \)) was used to convert weight fractions into volume fractions. Dilutions were then performed to prepare samples of desired volume fractions. The uncertainty in volume fractions reported is 0.5\% (0.005) or less, determined by repeated measurements. When not in use, all the sample tubes were sealed using Parafilm and stored in a refrigerator to minimize evaporation and contamination.

**Experimental setup** We used Fisherbrand plain glass slides as the hydrophilic impact substrate. The slides were cleaned using a 2.5M solution of NaOH in ethanol and water to remove organic impurities. A micropipette was used to form colloidal drops. The micropipette was mounted on a vertically moving pipette holder to vary impact velocities. We used 15 \( \mu L \) of fluid to obtain drops of 3.0 ± 0.1 mm diameter. The setup was enclosed in a humidity chamber with the relative humidity maintained between 70–80\% using a saturated solution of NaCl in water, and the humidity was monitored in real time during experiments. Before every trial of the impact experiments, a vortex mixer was used to re-disperse the sample, ensuring that it was consistently well-mixed.

The impacting drops were backlit using a white LED light, and filmed using two high-speed
cameras. The first camera, a Phantom V2512, captured the side-view of the impacting drop at 100,000 frames per second. The second camera, a Phantom V640L, filmed at 20,000 fps. It was tilted at an angle of 15° to gather information on how the impact affected the top surface of the drop. The experiment was repeated at least 5 times for each impact condition to ensure reproducibility.

**Rheological studies**

Stress-controlled rheological measurements were performed on the colloidal samples over 0.09 ≤ φ ≤ 0.50. The measurements were done on a TA Instruments Discovery HR-2 rheometer at room temperature (∼ 21°C) using the cone-plate geometry with 40 mm diameter and a 1° cone angle. The truncation gap was 25 µm. We covered the edges of the samples with a microscope immersion oil to minimize evaporation. The samples were pre-sheared to remove effects of shear history.

**Data analysis**

All high-speed videos were background-divided and analysed using ImageJ. The plots were made using python, and all errors reported are standard deviations over multiple trials. The maximum drop spread $d_{max}$ was determined by locating the frame in the impact timeseries where the extent of the spreading drop was the greatest. The height of the tallest point on the drop relative to the substrate, $h$, was measured for each frame in the image sequence. The minimum height $h_{min}$ was defined as the drop height at the crossover point between the decreasing and the plateau regimes in the $h$ vs. $t$ plot. The time of first observation of $h_{min}$, measured since the impact event, was defined as $t^*$. The slope of the linearly decreasing regime in the $h$ vs. $t$ plot was defined as $u^*$. To calculate the coefficient of restitution, the speed of the drop before impact $u_0$, and the speed
after rebound, $u_f$ were computed using several frames of the image sequence. The coefficient of restitution was then computed as $\epsilon = u_f / u_0$.

To calculate the speed of the upward-moving front, the side-view impact videos recorded at 100,000 fps were used. For every frame of the image sequence, the pixel-wise difference between consecutive frames was taken in ImageJ, so that only the elements that changed between consecutive frames (corresponding to the location of the moving front) were highlighted [SI video 7]. This enabled us to locate the jamming front with a time uncertainty of 10 $\mu$s. The images were then adjusted for brightness and contrast to enhance the moving front. The vertical height $h_{\text{front}}$ of the disturbance from the impact substrate was measured for each frame of the image sequence, until the front was no longer visible. For every high-speed video, the left and right half of the drop were separately analyzed to obtain two datasets for $h_{\text{front}}(t)$. In order to convert $h_{\text{front}}$ to the position along the drop surface, $r\theta_{\text{front}}(t)$, we approximated the drop profile as a circle of radius $r = 1.5$ mm (disregarding the slight deviation from spherical shape during front propagation), and used the relation $h_{\text{front}}(t) = r(1 - \cos \theta_{\text{front}}(t))$, such that $\theta_{\text{front}}(0) = 0$ at the impact point, to obtain the angle $\theta_{\text{front}}(t)$. A line was then fit to the $r\theta_{\text{front}}$ vs. time plots, and the slope, averaged over the two halves of the drop and several movies for each impact condition [Fig. 6c], was reported as $u_{\text{front}}$ with error bars indicating the standard deviation.

Acknowledgements We thank Jeff Richards, Sid Nagel, and Xiang Cheng for useful discussions. This work was supported by the National Science Foundation under award number DMR-2004176. This work made use of the EPIC facility of Northwestern University’s NUANCE Center, which has received support
from the SHyNE Resource (NSF ECCS-2025633), the IIN, and Northwestern’s MRSEC program (NSF DMR-1720139). We thank the Richards Lab at Northwestern University for the use of their rheometry facilities.

**Competing Interests** The authors declare that they have no competing financial interests.

**Correspondence** Correspondence and requests for materials should be addressed to MMD.

(michelle.driscoll@northwestern.edu).

**Author contributions** SA contributed to the conception of the work, experimental design, and data acquisition and analysis. PS contributed to data interpretation and analysis and drafted the manuscript. MMD contributed to the conception of the work, data analysis and interpretation, and drafted the manuscript.
1. Stickel, J. J. & Powell, R. L. Fluid mechanics and rheology of dense suspensions. *Annual Review of Fluid Mechanics* **37**, 129–149 (2005).

2. Osswald, T. & Rudolph, N. Polymer rheology. *Carl Hanser, München* (2015).

3. David, N. V., Gao, X.-L. & Zheng, J. Q. Ballistic Resistant Body Armor: Contemporary and Prospective Materials and Related Protection Mechanisms. *Applied Mechanics Reviews* **62** (2009). 050802.

4. Rus, D. & Tolley, M. T. Design, fabrication and control of soft robots. *Nature* **521**, 467–475 (2015).

5. Josserand, C. & Thoroddsen, S. T. Drop impact on a solid surface. *Annual review of fluid mechanics* **48**, 365–391 (2016).

6. Bergeron, V., Bonn, D., Martin, J. Y. & Vovelle, L. Controlling droplet deposition with polymer additives. *Nature* **405**, 772–775 (2000).

7. Blackwell, B. C., Deetjen, M. E., Gaudio, J. E. & Ewoldt, R. H. Sticking and splashing in yield-stress fluid drop impacts on coated surfaces. *Physics of Fluids* **27**, 043101 (2015).

8. Boyer, F., Sandoval-Nava, E., Snoeijer, J. H., Dijksman, J. F. & Lohse, D. Drop impact of shear thickening liquids. *Physical review fluids* **1**, 013901 (2016).

9. Peters, I. R., Xu, Q. & Jaeger, H. M. Splashing onset in dense suspension droplets. *Physical review letters* **111**, 028301 (2013).
10. Jørgensen, L., Forterre, Y. & Lhuissier, H. Deformation upon impact of a concentrated suspension drop. *Journal of Fluid Mechanics* **896** (2020).

11. Bertola, V. & Haw, M. D. Impact of concentrated colloidal suspension drops on solid surfaces. *Powder Technology* **270**, 412–417 (2015).

12. Thoraval, M.-J. *et al.* Nanoscopic interactions of colloidal particles can suppress millimetre drop splashing. *Soft matter* **17**, 5116–5121 (2021).

13. Kim, G., Kim, W., Lee, S. & Jeon, S. Impact dynamics of a polystyrene suspension droplet on nonwetting surfaces measured using a quartz crystal microresonator and a high-speed camera. *Sensors and Actuators B: Chemical* **288**, 716–720 (2019).

14. Stöber, W., Fink, A. & Bohn, E. Controlled growth of monodisperse silica spheres in the micron size range. *Journal of colloid and interface science* **26**, 62–69 (1968).

15. Zhang, L. *et al.* Hollow silica spheres: synthesis and mechanical properties. *Langmuir* **25**, 2711–2717 (2009).

16. Bi, D., Zhang, J., Chakraborty, B. & Behringer, R. P. Jamming by shear. *Nature* **480**, 355–358 (2011).

17. Scheller, B. L. & Bousfield, D. W. Newtonian drop impact with a solid surface. *AIChE Journal* **41**, 1357–1367 (1995).

18. Landau, L. D., Lifshitz, E. M., Kosevich, A. M. & Pitaevskii, L. P. *Theory of elasticity: volume 7*, vol. 7 (Elsevier, 1986).
19. Rathee, V., Blair, D. L. & Urbach, J. S. Localized stress fluctuations drive shear thickening in dense suspensions. *Proceedings of the National Academy of Sciences of the United States of America* **114**, 8740–8745 (2017).

20. Rathee, V., Blair, D. L. & Urbach, J. S. Localized transient jamming in discontinuous shear thickening. *Journal of Rheology* **64**, 299–308 (2020).

21. Cheng, X., Sun, T.-P. & Gordillo, L. Drop impact dynamics: Impact force and stress distributions. *Annual Review of Fluid Mechanics* **54**, null (2022).

22. Waitukaitis, S. R. & Jaeger, H. M. Impact-activated solidification of dense suspensions via dynamic jamming fronts. *Nature* **487**, 205–209 (2012).

23. Han, E., Peters, I. R. & Jaeger, H. M. High-speed ultrasound imaging in dense suspensions reveals impact-activated solidification due to dynamic shear jamming. *Nature communications* **7**, 1–8 (2016).

24. Peters, I. R., Majumdar, S. & Jaeger, H. M. Direct observation of dynamic shear jamming in dense suspensions. *Nature* **532**, 214–217 (2016).

25. Han, E., Wyart, M., Peters, I. R. & Jaeger, H. M. Shear fronts in shear-thickening suspensions. *Phys. Rev. Fluids* **3**, 073301 (2018).

26. Rømcke, O., Peters, I. R. & Hearst, R. J. Getting jammed in all directions: Dynamic shear jamming around a cylinder towed through a dense suspension. *Phys. Rev. Fluids* **6**, 063301 (2021).
27. Liu, A. J. & Nagel, S. R. The jamming transition and the marginally jammed solid. *Annual Review of Condensed Matter Physics* **1**, 347–369 (2010).

28. Richard, D., Clanet, C. & Quéré, D. Contact time of a bouncing drop. *Nature* **417**, 811–811 (2002).
Figure 1: **Rheology of the colloidal suspensions.** **a** SEM image of the colloidal silica spheres used in our drop impact experiments; the sphere diameter is $830 \pm 20$ nm. **b** Bulk rheological flow curves: the colloidal suspension exhibits viscous flow, shear thinning, and shear thickening as $\phi$ is increased. The grey triangle in the bottom right indicates the rate limit of the rheometer.
Figure 2: **Exotic impact behaviours of colloidal suspension drops.** a Timeseries of a $\phi = 0.47$ colloidal drop expanding after impacting at $u_0 = 3.0 \text{ m/s}$ [see also SI video 1]. The spreading drop shows transient pockets of localized solidification, indicating the onset of shear thickening. 

b Timeseries of a $\phi = 0.49$ colloidal drop impacting at $u_0 = 2.0 \text{ m/s}$ [see also SI video 2]. The bottom half of the drop solidifies, while the still-fluid top portion flows over it. 

c Timeseries of a $\phi = 0.49$ drop impacting at $u_0 = 3.0 \text{ m/s}$ [see also SI video 3]. While most of the drop is solidified, the top portion of the drop is in the liquid phase. All scale bars are 1 mm.
Figure 3: **State diagram of colloidal drop impact.** $\phi - u_0$ state diagram summarizing impact regimes; representative snapshots corresponding to these regimes are shown on the right. Green circles denote simple spreading behaviour, which dominates the low $\phi$, low $u_0$ region. Orange diamonds indicate that transient pockets of localised solidification were observed during spreading. Blue triangles correspond to the partial/full solidification regime, where the bottom portion of the drop jams after impact, but a shrinking region at the top remains fluid.
Figure 4: **Quantifying post-impact spreading.** a Normalised maximum diameter, $\beta = d_{\text{max}}/d_0$, as a function of $u_0$ for various volume fractions $\phi$. For $\phi \geq 0.49$ and high impact velocities, $\beta$ drops to 1, indicating the drop does not spread. Insets show representative snapshots of simple spreading (upper), localised solidification (middle), and bulk solidification (lower). Dotted lines are guides to the eye, and the dashed black line indicates $\beta = 1$. b Normalised maximum diameter, $\beta$, for $\phi \leq 0.47$ plotted against the dimensionless parameter $Re_{\text{eff}}We^{1/2}$. The exponent of the power law fit (dashed line, $\beta = 0.81(ReWe^{1/2})^{0.164}$) is in excellent agreement with the scaling reported for Newtonian fluids.\footnote{17}
Figure 5: **Characterisation of the partial solidification regime.**  

a Height of the tallest point on the drop from the impact substrate, plotted against time, for $\phi = 0.50$, $u_0 = 3$ m/s. $h$ decreases at the speed $u^*$ until time $t^*$, then plateaus at the value $h_{\text{min}}$. Inset: post-impact snapshot of a drop at the minimum height $h_{\text{min}}$.  

b $h$ decreases at a speed identical to the impact velocity, indicating that over the timescale $t^*$, the top portion of the drop is unaffected by the impact event. Dashed line corresponds to $u^* = u_0$.  

c $h_{\text{min}}/d_0$ vs. impact velocity $u_0$. $h_{\text{min}}/d_0$ increases with increasing impact velocity, and then plateaus at a value less than 1, indicating finite compression of the drop along impact axis. Dashed line indicates $h_{\text{min}}/d_0 = 1$. 


Figure 6: **Dynamics of the solidification front.** a Timeseries of a $\phi = 0.50$ drop impacting at $u_0 = 2.0$ m/s [see also SI video 6]. Right panels are images obtained by subtracting consecutive frames, so that the edge of the solidification front is highlighted (shown by arrows). The red circle indicates the drop profile before impact. Even at 0.19 ms, the portion of the drop above the front maintains its pre-impact curvature. Scale bar is 1 mm. b Schematic of a subtracted image of the moving solidification front, outlining relevant parameters. The height, $h_{\text{front}}$, of the edge of the white outline gives the location of the front, which is then converted to $r\theta_{\text{front}}$ using the spherical geometry. c Example datasets of $r\theta_{\text{front}}$ vs. $t$ for $\phi = 0.49$ and $u_0 = 3$ m/s. $r\theta$ vs. $t$ is a straight line, the slope being the front speed along the surface, $u_{\text{front}}$. d $u_{\text{front}}$ plotted against $\dot{\gamma}$. e High-$\phi$ bulk rheological data from Fig.1b re-plotted as shear stress vs. shear rate. Dotted lines indicate the onset shear rates $\dot{\gamma}_c$ for shear thickening. f The $\frac{2u_{\text{front}}}{d_{\text{front}}}$ data for $\phi = 0.49$ and $\phi = 0.50$, when plotted against $\dot{\gamma} - \dot{\gamma}_c$, collapses on a single curve.