Shear jamming in colloidal drop impact

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Understanding the rich and nuanced physics of drop impact is crucial in many processes: for example, geological erosion, industrial coating, forensics, and agrochemical delivery\(^1\), and the drop impact of Newtonian fluids has been a studied extensively for over a century\(^1-3\). However, literature on the impact dynamics of complex fluids is relatively sparse, despite the ubiquity of complex fluids in many natural and industrial impact processes. Here, we present a systematic study of colloidal drop impact over a large range of volume fractions and impact velocities. We observe behaviors ranging from liquid-like spreading to solid-like jamming, delineate them in a state diagram, and connect them to the bulk rheological behavior\(^4,5\) of our colloidal system. These experiments allow for an exploration of the rich flow behaviors that are only apparent in the absence of confinement and in the presence of spatiotemporally varying stresses. Furthermore, our observations of nonuniform jamming behaviors are direct visual evidence for recent reports of localised stress fluctuations\(^6,7\) and dynamically propagating jamming fronts\(^8-11\).

When a liquid drop impacts on a solid substrate [Fig. 1b], several parameters govern the impact dynamics\(^12\): impact velocity and angle, the properties of the impacting liquid, the solid substrate, and the ambient gas all play crucial roles in the post-impact behavior. As a result of many
studies for over a century, a fairly comprehensive understanding has been built of the dynamics of a single Newtonian fluid drop impacting a dry solid substrate[1,2]. The highly non-Newtonian rheology of complex fluids can lead to a diversity of exotic flow behaviours in these materials. However, there is lack of systematic studies exploring the impact of complex fluid drops. Previous work has largely focused on the impact dynamics of non-Brownian suspensions[12–14] and polymeric fluids[15–17]; only a few have conducted experiments with colloidal suspensions[18–20].

Here, we report a systematic experimental study of the impact dynamics of colloidal silica suspensions [Fig. 1a] with volume fractions $0.09 \leq \phi \leq 0.50$, and impact velocities $0.7 \text{ m/s} \leq u_0 \leq 4.0 \text{ m/s}$. In this range of $\phi$ and $u_0$, the suspension exhibits a wide range of flow behaviors[21] from Newtonian-like to discontinuous shear thickening (DST)[22]. Not only does this system allow us to access shear rates near the limits of conventional rheometers, but it also provides direct visual evidence of the effect of spatiotemporally varying stresses on flow behavior [Fig. 1b,c]. As rheometry only provides the bulk-averaged properties of fluids under confinement, information gained from impact experiments in the near-total absence of confinement can complement rheological measurements to build a more holistic understanding of the flow behavior of colloidal fluids under high stresses.

We synthesized silica spheres of diameter $830 \pm 20 \text{ nm}$ [Fig. 1a] using the well-known Stöber process[23,24]. Drops 3 mm in size were formed by drawing a known volume of fluid into a micropipette and the impact velocity was set by changing the height from which drops were released. The drops were impacted on a dry hydrophilic glass substrate. We recorded the impacting
drops at 100,000 frames per second (fps). To minimize the effects of particle sedimentation, all samples were sonicated before impact experiments. All experiments were performed in a humidity chamber, which also mitigated air currents.

When a fluid drop impacts and spreads onto a substrate, it experiences a shear that is nonuniform both over time and over position in the drop. During this process, the kinetic energy of the drop gets converted into surface energy accompanied by viscous dissipation. The maximum shear rate experienced by the impacting drop can be estimated as the ratio of impact velocity and drop diameter, \( \frac{u_o}{d_o} \). Thus, for our system with \( d_o = 3.0 \pm 0.1 \) mm, we effectively varied the maximum shear rate \( \dot{\gamma} \) between 233-1333 s\(^{-1}\). From our bulk rheological data [Fig. 2b], we expect to observe the whole range of shear thinning, continuous shear thickening, and discontinuous shear thickening behaviors. We also note that the highest shear rate of 1333 s\(^{-1}\) allows us to probe a regime approaching the working limit of typical rheometers.

Scanning the impact dynamics over both \( \phi \) and \( \dot{\gamma} \), we observed a range of behaviors not exhibited by Newtonian drop impact. At varying \( \phi \) and constant \( \dot{\gamma} \), we observed the drop behavior transitioning from simple spreading to partial jamming to a fully jammed state [Fig. 2a]. Conversely, at constant \( \phi \), we observed a similar transition with increasing \( \dot{\gamma} \). This transition can be approximately mapped to the range of volume fractions and shear rates where shear thickening behavior is observed in a rheometer [Fig. 2b].

We quantified the dynamics of this transition by measuring the normalized maximum spread of the impacted drops \( \lambda = \frac{d_{max}}{d_0} \) as a function of shear rate \( \dot{\gamma} = \frac{u_o}{d_0} \) over the whole range
of volume fractions. As shown in Fig. 2c, for $\phi \leq 0.47$ the normalized spread increases with shear rate due to the increasing kinetic energy available. The normalized spread decreases with increasing $\phi$, a reflection of the rising effective viscosity of the suspension. The most striking feature of this plot, however, is that for the highest volume fractions of 0.49 and 0.50, the maximum spread $\lambda$ drops to 1 at high shear rates. This is because the drop no longer spreads after impact, as seen from panel 3 in Figure 2a. Markedly different from its Newtonian counterparts, this behavior is quantitative evidence that the drop partially or fully solidifies upon impact. Furthermore, the shear rates at which $\lambda$ drops to 1 are consistent with the onset of shear thickening that we observed in bulk rheological data [Fig. 2b]. All of these behaviors were captured over a timescale of milliseconds using high-speed imaging. However, the drops were observed to relax and spread in a liquid-like manner over much longer timescales of seconds, when external shear was no longer present. This is further evidence that the short-timescale response is a direct result of shear jamming. The long-timescale spreading is likely governed by surface properties. A different study of dense colloidal drop impact has reported similar spreading on glass substrates, while they observed the drops to remain jammed for days on hydrophobic PTFE substrates\textsuperscript{18}.

To summarize the range of outcomes observed in the impacting colloidal drops, we constructed a $\phi - \dot{\gamma}$ state diagram [Fig. 3]. Gray squares, indicating simple spreading, dominate the low volume fraction and low-shear region. At $\phi = 0.47$ and $\dot{\gamma} > 1000s^{-1}$, we observe the first signs of non-Newtonian behavior in the form of pockets of localized jamming that emerge and quickly disappear over milliseconds [Fig. 1b]. This regime coincides with the first appearance of shear thickening in bulk rheology [$\phi = 0.47$ curve in Figure 2b]. At even higher $\phi$, where rheo-
logical measurements show strong continuous and discontinuous shear thickening, we observe the transition to a fully jammed drop via a partial jamming regime. As evident from the state diagram, the drop behavior is very sensitive to small changes in $\phi$, consistent with the rheological transition to shear thickening.

A noteworthy feature observed in our experiments is the occurrence of localized jamming [Fig. 1b, yellow region in Figure 3]. Past studies have reported regions of high localized stress using spatially resolved stress measurements on silica suspensions in both the CST\textsuperscript{6} and DST\textsuperscript{7} regimes, leading to high local viscosity regions akin to a jammed solid-like phase. Our observations of localized jamming are striking visual evidence of such a mechanism, that we are only able to access due to the absence of confinement in our system. We note that past stress measurements were conducted under a constant external applied stress, as opposed to the spatiotemporally varying stress present in drop impact. Further spatially resolved stress measurements\textsuperscript{25} performed on impacting drops could help shed more light on this phenomenon.

In the partial jamming regime [Fig. 1c, blue region in Fig. 3], we observe a fluid-like depression at the top of the drop [Fig. 4b], and we hypothesize that this is correlated with larger and larger portion of the drop exhibiting solid-like behavior with increasing shear rate. We measure the maximum size $\delta$ of this depression. Figure 4d shows the normalized depression diameter $\delta/d_0$ plotted against impact velocity. At $\dot{\gamma} \gtrsim 667 s^{-1}$, the depression size falls and plateaus to zero, indicating that the transition to a fully jammed state is complete.

The depression at the apex of the drop, shrinking with increasing shear rate, is consistent
with an upward propagating jamming front. As further evidence of this propagation, we observed a disturbance travelling from the bottom to the top of the drop [Fig. 4a] in both the partially and fully jammed regimes. Similar shear-driven jamming fronts have been observed in both externally impacted\textsuperscript{8–10} and sheared\textsuperscript{11} cornstarch suspensions. Figure 4c shows the normalised speed of the solidification front, $u_{front}$, plotted against the impact velocity $u_0$; we note that $u_{front}$ is several times larger than $u_0$. Whether the solidification fronts observed in our experiments are connected to numerical observations of shock fronts\textsuperscript{26,27} in soft systems, needs further investigation using measurements of local density fluctuations during impact.

Based on the observed behaviors in the localized, partial, and shear jamming regimes in our experiments, the following overall picture emerges. As a dense suspension drop impacts the substrate, it is subjected to a spatially and temporally varying shear over the drop volume. The pockets of localized jamming at relatively lower $\phi$ and $\dot{\gamma}$ are likely indicative of transient regions of high localized stress. In the partial jamming regime, an increasing volume of the drop jams as the shear rate is increased, quantified by the shrinking size of a liquid-like depression at the apex of the drop. At even higher $\phi$ and $\dot{\gamma}$, the size of this depression drops to zero, indicating a fully jammed regime.

In summary, our highly time-resolved colloidal drop impact experiments allow us to systematically probe flow regimes of colloidal suspensions ranging from Newtonian-like to shear jamming. The absence of confinement in the drop impact system enables us to gain visual information on the effect of localised stresses in a non-Newtonian fluid which is not accessible via bulk
rheometry. Behaviours such as partial and full jamming are a direct consequence of the nonuniform shear that propagates up the drop in form of a solidification front. We constructed a $\phi - \dot{\gamma}$ state diagram capturing simple spreading, localized jamming, partial jamming, and full jamming regimes, and connected them to the rheological properties for our suspensions. It is also worth noting that although jamming occurred over milliseconds, the suspension drops relaxed into their fluid-like state over longer timescales in the absence of shear.

In the future, spatiotemporally resolved stress measurements in drop impact systems could provide further insights into the effect of spatially nonuniform shear on impacting drops. Understanding the nature of the solidification front we observed traveling up the drop could provide valuable insights into the broader problem of jamming front propagation. We note that we observe signatures of shear jamming only in a narrow range of $\phi$. Elongated particle suspensions are shown to exhibit shear jamming for a broader range of $\phi$ in both experiments and simulations. Drop impact experiments studying anisotropic suspensions would both enhance the range of state space where we would expect to observe jamming, and enrich our understanding of the flow behavior of dense suspensions with an additional orientational degree of freedom.

**Methods**

**Colloidal sample preparation**

We prepared silica spheres in our lab using the Stöber silica synthesis method. The size of the particles was controlled by the number of ‘feeds’ in the reaction. We performed 14 feeds after the initiation of the reaction, resulting particles with a diameter of $830 \pm 20$ nm. In order to clean
the particles, the reaction mixture was centrifuged and re-suspended in ethanol 3 times. We then gravity separated the suspension to improve monodispersity, and then re-suspended the particles in water. The particles were characterized using Scanning Electron Microscopy on the Hitachi S4800 instrument. We characterized the particle size and polydispersity by measuring a representative sample of 100 particles.

We prepared a concentrated stock solution of the silica colloids in water, and measured the weight fraction by drying 100 µL of the concentrated suspension. We used a density of 2 g/cm³ for silica colloidal spheres to convert weight fractions into volume fractions. We then performed dilutions to prepare samples of desired volume fractions. All the sample tubes were sealed using Parafilm and stored in a refrigerator when not in use to minimize evaporation and contamination.

**Experimental set-up**

We used Fisherbrand plain glass slides as the hydrophilic substrate. The slides were cleaned using a 2.5M NaOH solution in ethanol and water to remove any organic impurities. A micropipette was used to form colloidal drops that were impacted on the substrate from varying heights. The micropipette was mounted on a vertically moving pipette holder to vary impact velocities. We used 15 µL of fluid to obtain drops of 3.0 ± 0.1 mm diameter. This whole setup was enclosed in a humidity chamber, and the relative humidity was maintained between 70-80% using a saturated solution of NaCl in water. The humidity was monitored in real-time during experiments. Before every impact experiment, we used a vortex mixer to re-disperse the sample to ensure that the sample was well-mixed.
The impacting drops were backlit using a white LED light, and filmed using two time-synchronised high-speed cameras. The first camera, a Phantom V2512, captured the side-view of the impacting drop at 100,000 frames per second (fps). The second camera, Phantom V640L, filmed at an angle of $15^\circ$ below horizontal at 20,000 fps, so as to gather more information on the impact behavior and how it affected the top surface of the drop. Each experiment was repeated at least 5 times to ensure reproducibility.

**Rheological studies**

We performed stress-controlled rheological measurements on the colloidal samples with $0.09 \leq \phi \leq 0.50$. The measurements were done on a TA Instruments Discovery HR-2 rheometer at room temperature ($\sim 21^\circ$ C) using the cone-plate geometry with a $2^\circ$ cone angle. We covered the edges of the samples with a microscope immersion oil to minimize evaporation. The samples were pre-sheared to remove any effects of shear history.

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Figure 1: Shear jamming in drop impact. a Electron microscopy image of the colloidal silica we synthesized for our experiments. b Timeseries of a $\phi = 0.47$ colloidal drop expanding after impacting at $u_0 = 3$ m/s ($\dot{\gamma} = 1000$ s$^{-1}$), showing transient pockets of localized jamming. Scale bars are 1 mm. c Timeseries of a $\phi = 0.49$ colloidal drop impacted at $u_0 = 2$ m/s ($\dot{\gamma} = 667$ s$^{-1}$) showing partial solidification from the bottom. Scale bars are 1 mm.
Figure 2: **Quantifying impact outcomes.**

**a** Images of colloidal drops at maximum expansion $d_{max}$ for $\phi = 0.09, 0.49, \text{ and } 0.50$ at constant shear rate $\dot{\gamma} = 667 \text{ s}^{-1}$. The scale bars are 1 mm.

**b** Rheological flow curves for a range of $\phi$ plotted against shear rate $\dot{\gamma}$, showing Newtonian-like, shear thinning, and the transition to shear thickening behavior as $\phi$ is increased.

**c** Normalised maximum diameter $\lambda$ as a function of shear rate $\dot{\gamma}$ for various volume fractions $\phi$. $\lambda$ increases with increasing $\dot{\gamma}$ for $\phi < 0.49$. However for $\phi \geq 0.49$ and high shear rates, $\lambda$ drops to 1, indicating the drop stops spreading at high $\phi$ and $\dot{\gamma}$. Additionally, $\lambda$ decreases with increasing $\phi$ for constant $\dot{\gamma}$, as expected for a more viscous suspension.
Figure 3: **State diagram for colloidal drop impact.** $\phi - \dot{\gamma}$ state diagram summarizing the diverse outcomes after impacting a colloidal drop onto a solid substrate. Representative snapshots corresponding to these behaviors are shown on the right. Solid square grey symbols indicate simple spreading behavior, which dominates the low $\phi$, low $\dot{\gamma}$ region. Yellow open circles indicate the transient pockets of localised jamming observed. Half-solid blue symbols indicate the partial jamming regime, where the bottom portion of the drop acts like a solid, but some of the top portion behaves as a fluid. Finally, the solid red symbols correspond to fully jammed drops which exhibit no fluidlike depression at the apex.
Figure 4: **Upward-moving solidification fronts.** a Timeseries of a $\phi = 0.50$ drop, arrows highlight the disturbance (jamming front) moving upward along the drop surface. b Timeseries of a $\phi = 0.49$ drop illustrating partial jamming: much of the drop volume is solidified at impact, but a liquid-like depression of width $\delta$ is observed at the apex of the drop. c The normalised speed, $u_{\text{front}}/u_0$, of the upward-travelling front observed in partial and full jamming regimes, plotted against impact velocity. $u_{\text{front}}$ is several times larger than $u_0$. d The normalised maximum diameter, $\delta/d_0$ of the top depression vs. impact velocity. The size of the depression decreases with increasing impact velocity, indicating that an increasing portion of the drop is in the solid-like phase with increasing shear rate.