nature portfolio

Peer Review File

Coexistence of solid and liquid phases in shear jammed colloidal drops

Reviewers' comments:

Reviewer #1 (Remarks to the Author):

In this manuscript, Shah, Arora & Driscoll report on the experimental results of the impact of drops of colloidal suspensions on a large range of volume fractions and impact velocities. Using high-speed imaging of the drop interface, the authors show that, during the impact, the colloidal suspension within the drop displays behaviors that range from Newtonian-like to shear thickening and shear jamming. The behaviors can even coexist as is demonstrated by the emergence of patches of jammed suspension in drop impacts in transition regimes.

This manuscript aims to characterize an interesting and relevant problem of complex-fluid dynamics that is challenging due to the limitations of current techniques in terms of spatial and temporal resolutions. Authors have managed to obtain key pieces of information although these limitations and interpret them. Furthermore, they have done a good job explaining carefully the experimental techniques they use, as well as the derived phenomenological laws. The outcome of the experiments is very well described, classified, and analyzed. The bibliography may be missing some key articles for the Newtonian drop impact, but otherwise, it seems to me to be complete. Videos are well-edited, plots and diagrams are clear, and the writing style is simple, direct, and clear. In general, the article is scientifically sound and very well structured. For the reasons above and considering the requirements for publication of Communications Physics, I recommend the manuscript for publication in its current form. Below I include some questions and comments to be considered by the authors for improving the content of the article.

To summarize, this manuscript that reports on the impact of colloidal-suspension drops is well written and scientifically sound. For this reason, I recommend its publication in Communications Physics.

Comments to the Authors

This section includes scientific comments for the authors.

• Major questions

– – In the case of Newtonian fluids, there has been great progress in the last years concerning the flow inside the drop during the early stages of impact. Theoretical predictions have been shown to match the results of both numerical simulations and experiments. Results include predictions for the velocity field, pressure, and shear as a function of space and time. I was wondering if these results could be used for determining a map for the shear rate for the Newtonian flow case. I think they may be helpful to determine which regions are more prone to suffer shear thickening or jamming of the colloidal suspensions. I believe this should be at least commented in the manuscript. The two references below are important.

1. Drop impact on a solid surface: short time self-similarity. J Philippi, PY Lagrée, A Antkowiak. Journal of Fluid Mechanics 795, 96-135

2. Dynamics of drop impact on solid surfaces: evolution of impact force and self-similar spreading. L Gordillo, TP Sun, X Cheng. Journal of Fluid Mechanics 840, 190-214.

I have the feeling that the cushion shape that you have on the top of the drop may be related to the shear distribution. It may also help to do some better predictions on the solidification-front motion.

– My second concern is about compression. Are we really sure that the solidified region in the partial solidification is compressed? Couldn't it be a flow-past object configuration instead responsible for the decreased h_{\min}/d_{0}? I think that these concerns could be easily figured out if the volume of the drop (in the apparent maximum compression) is obtained from image analysis.

• Minor corrections

– L23. I suggest adding “under dynamic conditions” or “under impulsive forces” to be more specific.

– L46. Please specify what you mean by information. Sound waves? Deformation?

– L50. give -> gives

– L51. Post-impact -> impact.

– L58. Quantify high shear stresses in the text.

– L67. Problem with reference.

– L100. Please provide an interval of confidence for the coefficient and exponent of the power-law fit.

– L106. Please consider including some more precise description for the time at which the patches exist (maybe by comparing with some temporal feature of drop impact, like spreading time, etc.)

– L140. “drop bounces off”

– L200. I think that the next question is: Why is drop impact better than other (dynamic) setups that could be more controlled?

– Figure 3. I believe a Phase diagram will be more appropriate than a Phase Diagram

– Figure 4. People will refer to “drop maximum spreading” instead of “post-impact spreading”

– Figure 5. People use “apex” instead of “tallest point”.

Reviewer #2 (Remarks to the Author):

This manuscript studies the impact of colloidal drops, varying the volume fraction of the particles and the impact velocity, while keeping constant the size of the particles. The authors observe shear jamming at large volume fractions and impact velocities. They correlate these observations with the rheology of the suspensions. They also observe the propagation of the solidification front on the side of the drop. Overall, the observations of jamming are impressive and the experiments provide a careful and thorough investigation of the problem. I therefore recommend publication in Communications Physics. The authors may consider the minor suggestions listed below:

1. I think that the term "Coexistence of solid and liquid phases" in the title is misleading. It does not convey the key information that the solid phase appears due to the impact. Using the term "shear jamming" in the title would probably be more clear. Same remark for the abstract.
2. Line 19: "it provides spatially resolved data": It probably provides more information on the spatial and temporal resolution, but I would not say "resolved".
3. Line 19-20: "on the manifestations of non-Newtonian flow": captures only some of the non-Newtonian effects.
4. Line 20: "ultrahigh-speed imaging" -> high-speed imaging
5. A simple estimate from the scale in Fig. 1a gives a typical diameter for the spheres around 600 nm,

which is significantly lower from the 830 +/- 20 nm reported in the manuscript. Can the authors explain this difference?
6. What is the contact angle of the suspensions? Does it vary with the concentration of the suspension?
7. Line 27: The introduction on references 6-13 could probably be expanded to better put this manuscript in the context of previous studies. Especially, I think that it would be useful to explain how previous studies compare to the study in this manuscript, including in terms of particle sizes, range of concentrations and results obtained.
8. Line 30: It would probably be useful to give an introduction on shear thickening with some key review articles on the topic. It would be useful also to give a simple overview of when shear thickening is expected, according to the results published previously, in terms of shear rates and concentrations of the suspensions, and explain which parameters are important to influence it.
9. Line 37: What is the typical sedimentation time of the particles?
10. Line 48: Boyer et al. [1] made a different choice to estimate the maximum shear rate in the drop during impact. It would be useful to understand why the authors are making a different choice here.
11. Line 67: Typo " jamming thresholdjames2019controlling"
12. Line 87: I do not think that Ref. 17 is the best choice to model the maximum spreading of the impacting drop, especially considering that it completely ignores the effect of contact angle, that is probably relevant here. The authors should probably use a more recent theory such as the one presented in Lee et al. [2]. At least, the comparison with this more recent theory should be provided in a Supplementary Material file.
13. Line 91: It would be useful to provide the equation used to calculate the density of the colloidal suspension.
14. Line 91-92: What value of the surface tension is used in this study? Does it vary with the concentration of the suspension?
15. Line 111-112: What is the "further analysis" mentioned here to support the statement that the top of the drop remains liquid?
16. Line 116-117: Previous studies such as [3–5] had already observed that the top of the drop initially continues to fall at the impact velocity. It would be worth mentioning here.
17. Fig. 6: The shear rate estimation experienced during impact should have a different name from the shear rate measured in the rheometer, as they are of completely different nature. It is critical to understand clearly which one is used in the discussions and figures.
18. Line 137: I am not convinced that a shear rate above the critical shear rate for jamming is really meaningful. See for example Fig. 6e.
19. Line 138: "Thus, the speed of this disturbance is set by this excess shear rate.": I would recommend to replace this affirmation by something like "this suggests that…", considering that this interpretation is highly hypothetical.
20. Line 140-141: I think it would be useful to repeat here the hydrophilic nature of the target surface, as it is not common to see rebound on a hydrophilic surface.
21. Line 177: "in the absence of confinement": This statement implies that the confinement by surface tension in the drop is not important. I would either rephrase the statement, or formulate explicitly this hypothesis with supporting evidence.
22. Line 183-187: I guess that the distribution of shear stress within the impacting drop plays a key role in the dynamics of the shear front. The authors could benefit from looking at the following studies which provide information on the velocity profile [3,6].
23. The naming of the SI videos that I have received did not include their numbering mentioned in the text. The authors should ensure that the names of the videos are consistent with the naming of the files in the final publication.
24. Ref. 21 should be updated.
25. Fig. 4: I think it would be useful to plot the maximum spreading diameter as a function of the effective Reynolds number as well as the Weber number, to understand the range of these parameters covered in the experiments. This could be put in a Supplementary Material file is necessary.
26. Fig. 4b: I am not convinced that the choice of the effective viscosity or the choice of the model for the maximum spreading diameter are the best to collapse the data. I still see a systematic deviation

when increasing the concentration instead of an "excellent agreement".
27. Fig. 5b: I think that plotting the ratio u*/u0 as vertical axis would make it easier for the reader to understand the deviation from the free fall approximation u*=u0.
28. Fig. 5b: I am a bit confused how one data point can be above the dashed line. This would mean that the top of the drop could fall faster than the impact velocity? Some careful checking or comments on this would be useful.

References

[1] F. Boyer, E. Sandoval-Nava, J. H. Snoeijer, J. F. Dijksman, and D. Lohse, Phys. Rev. Fluids 1, 013901 (2016).
[2] J. B. Lee, N. Laan, K. G. de Bruin, G. Skantzaris, N. Shahidzadeh, D. Derome, J. Carmeliet, and D. Bonn, J. Fluid Mech. 786, R4 (2016).
[3] G. Lagubeau, M. A. Fontelos, C. Josserand, A. Maurel, V. Pagneux, and P. Petitjeans, J. Fluid Mech. 713, 50 (2012).
[4] L. Gordillo, T.-P. Sun, and X. Cheng, J. Fluid Mech. 840, 190 (2018).
[5] B. R. Mitchell, J. C. Klewicki, Y. P. Korkolis, and B. L. Kinsey, J. Fluid Mech. 867, 300 (2019).
[6] J. Philippi, P.-Y. Lagrée, and A. Antkowiak, J. Fluid Mech. 795, 96 (2016).

We are thankful to both the referees for their time and efforts in reviewing our work. We are glad to know that both referees find our work highly relevant and technically sound, and recommend it for publication. We are grateful for the constructive comments provided by both the referees. In the revised manuscript, we have added important details and made changes in phrasing to remove ambiguities, and we believe incorporating this feedback from both referees has improved our manuscript. We have marked all the changes made to the manuscript in red for easy identification. Where appropriate, we have consolidated the remarks by both referees and used the following colour-code:

Referee 1: red

Referee 2: purple

– – In the case of Newtonian fluids, there has been great progress in the last years concerning the flow inside the drop during the early stages of impact. Theoretical predictions have been shown to match the results of both numerical simulations and experiments. Results include predictions for the velocity field, pressure, and shear as a function of space and time. I was wondering if these results could be used for determining a map for the shear rate for the Newtonian flow case. I think they may be helpful to determine which regions are more prone to suffer shear thickening or jamming of the colloidal suspensions. I believe this should be at least commented in the manuscript. The two references below are important.

1. Drop impact on a solid surface: short time self-similarity. J Philippi, PY Lagrée, A Antkowiak. Journal of Fluid Mechanics 795, 96-135

2. Dynamics of drop impact on solid surfaces: evolution of impact force and self-similar spreading. L Gordillo, TP Sun, X Cheng. Journal of Fluid Mechanics 840, 190-214.

I have the feeling that the cushion shape that you have on the top of the drop may be related to the shear distribution. It may also help to do some better predictions on the solidification-front motion.

22. Line 183-187: I guess that the distribution of shear stress within the impacting drop plays a key role in the dynamics of the shear front. The authors could benefit from looking at the following studies which provide information on the velocity profile [3,6].

[3] G. Lagubeau, M. A. Fontelos, C. Josserand, A. Maurel, V. Pagneux, and P. Petitjeans, J. Fluid Mech. 713, 50 (2012).

[6] J. Philippi, P.-Y. Lagrée, and A. Antkowiak, J. Fluid Mech. 795, 96 (2016).

We thank the referees for these detailed remarks. Although the above studies of low-viscosity Newtonian fluids have established the velocity and pressure fields within the drop, they are not directly applicable in the case of high-viscosity systems. The highly non-Newtonian nature of colloidal suspensions adds further complexity, as the fluid viscosity is a strong function of the applied shear. We have elaborated on these challenges in comparison to Newtonian studies in a new discussion paragraph (line 193).

– My second concern is about compression. Are we really sure that the solidified region in the partial solidification is compressed? Couldn't it be a flow-past object configuration instead responsible for the decreased h_{\min}/d_{0}? I think that these concerns could be easily figured out if the volume of the drop (in the apparent maximum compression) is obtained from image analysis.

We thank the referee for this observation. At high impact velocity and ϕ, in addition to observing $h_{min}/d_0 < 1$, we observe a slight bulging in the direction perpendicular to the impact direction. Our initial phrasing may have inadvertently suggested that the drop volume is not constant after impact. We have now added this clarifying information to the manuscript (Line 136).

– L46. Please specify what you mean by information. Sound waves? Deformation?

We are grateful to the referee for pointing out the ambiguity in phrasing. We have now replaced the word 'information' with 'shear caused by impact' as marked in red (Line 63).

– L58. Quantify high shear stresses in the text.

Per the referee's suggestion, we have added this information to the text (Line 76).

– L200. I think that the next question is: Why is drop impact better than other (dynamic) setups that could be more controlled?

We believe that drop impact is an especially powerful tool because of the ability to probe suspension behaviour under large instantaneous shear. We have modified our statement to reflect this (Line 244).

– L106. Please consider including some more precise description for the time at which the patches exist (maybe by comparing with some temporal feature of drop impact, like spreading time, etc.)

We are grateful to the referee for this comment. The patches appear during the spreading phase (~1 ms timescale) and last well into the receding phase, anywhere between 5 and 50 ms. Our observations suggest that these patches occur in a more or less random way, and there is a large variance in their lifetime. We have now included as much clarifying information as possible (Paragraph on line 131). We believe the stochastic nature of these patches point to the mechanism of shear thickening suggested by Rathee et al.

– Figure 3. I believe a Phase diagram will be more appropriate than a Phase Diagram

We agree with the referee that 'phase diagram' is often used in such context. However, we chose the word 'state diagram' for our classification of post-impact states of the drop in order to avoid semantic confusion with phase transitions.

– L23. I suggest adding "under dynamic conditions" or "under impulsive forces" to be more specific.

– L50. give -> gives
– L51. Post-impact -> impact.

– L67. Problem with reference.
– L100. Please provide an interval of confidence for the coefficient and exponent of the power-law fit.
– L140. "drop bounces off"
– Figure 4. People will refer to "drop maximum spreading" instead of "post-impact spreading"
– Figure 5. People use "apex" instead of "tallest point".

We thank the referee for the detailed editing comments above. We have made the necessary changes to the manuscript and marked them in red for easy identification.

1. I think that the term "Coexistence of solid and liquid phases" in the title is misleading. It does not convey the key information that the solid phase appears due to the impact. Using the term "shear jamming" in the title would probably be more clear. Same remark for the abstract.

We thank the referee for this remark. We have edited the title and abstract which we believe better describes the work.

2. Line 19: "it provides spatially resolved data": It probably provides more information on the spatial and temporal resolution, but I would not say "resolved".

Per the referee's suggestion, we have changed the phrase to 'with spatial and temporal resolution' to avoid sematic ambiguity.

3. Line 19-20: "on the manifestations of non-Newtonian flow": captures only some of the non-Newtonian effects.

Although we agree with the referee that we only study some of the non-Newtonian effects, it is clear that the drop impact system could be used to study a wide range of non-Newtonian flow behaviours.

4. Line 20: "ultrahigh-speed imaging" -> high-speed imaging

11. Line 67: Typo " jamming thresholdjames2019controlling"
13. Line 91: It would be useful to provide the equation used to calculate the density of the colloidal suspension.
14. Line 91-92: What value of the surface tension is used in this study? Does it vary with the concentration of the suspension?
15. Line 111-112: What is the "further analysis" mentioned here to support the statement that the top of the drop remains liquid?

19. Line 138: "Thus, the speed of this disturbance is set by this excess shear rate.": I would recommend to replace this affirmation by something like "this suggests that…", considering that this interpretation is highly hypothetical.
20. Line 140-141: I think it would be useful to repeat here the hydrophilic nature of the target surface, as it is not common to see rebound on a hydrophilic surface.

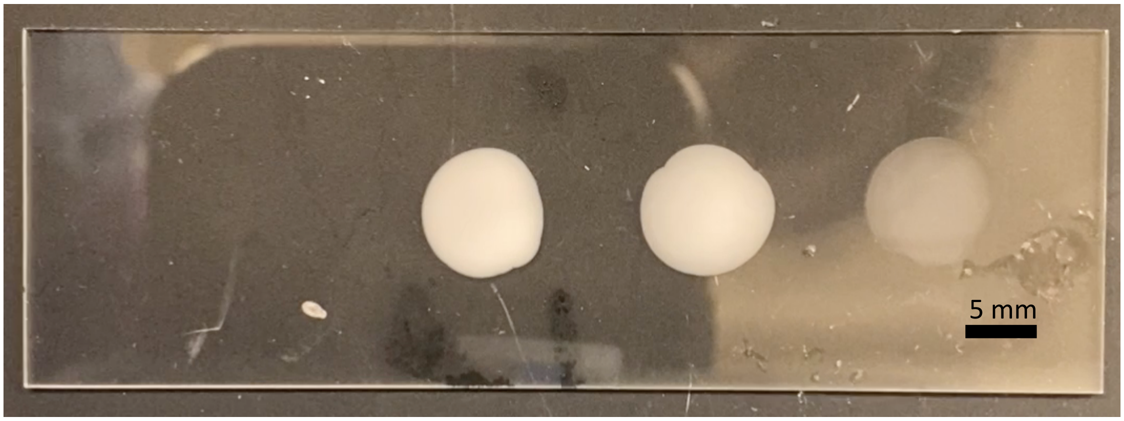
We thank the referee for these comments that help us add important details. We have added the relevant equations and explanations to clarify the points above.

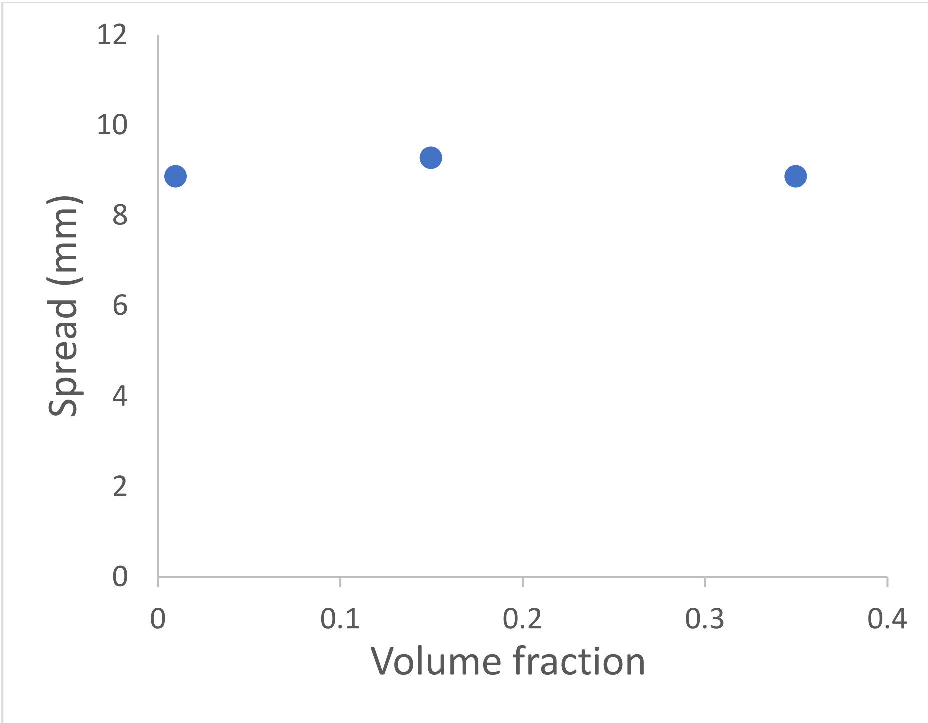
5. A simple estimate from the scale in Fig. 1a gives a typical diameter for the spheres around 600 nm, which is significantly lower from the 830 +/- 20 nm reported in the manuscript. Can the authors explain this difference?

We are very grateful to the referee for noticing this discrepancy. We accidentally used an SEM image from another synthesis run (not used for drop impact experiments). We have now replaced the figure with the right image, and our re-analysis has verified that the particle size indeed is 830 +/- 20 nm.

6. What is the contact angle of the suspensions? Does it vary with the concentration of the suspension?

Here, we show drops at $\phi = 0.35, \phi = 0.15$, and $\phi = 0.01$ drops (left to right) placed on a hydrophilic glass plate similar to the one used for experiments. We see that all drops spread to the same extent (average 9.0 mm), shown quantitatively in the plot below. Using the drop volume of 5 μL and this spreading size, we use the spherical cap approximation and estimate the contact angle of 4^{o}. Thus, the contact angle does not vary significantly with ϕ. Furthermore, given the short timescale of the phenomena we report in this work, we believe any small variation in contact angle would not significantly affect impact outcomes.





7. Line 27: The introduction on references 6-13 could probably be expanded to better put this manuscript in the context of previous studies. Especially, I think that it would be useful to explain how previous studies compare to the study in this manuscript, including in terms of particle sizes, range of concentrations and results obtained.

We thank the referee for this comment, and we have now expanded this paragraph to include more details about past studies (paragraph at Line 26).

8. Line 30: It would probably be useful to give an introduction on shear thickening with some key review articles on the topic. It would be useful also to give a simple overview of when shear thickening is expected, according to the results published previously, in terms of shear rates and concentrations of the suspensions, and explain which parameters are important to influence it.

Per the referee's suggestion, we have now included a paragraph outlining the current understanding of shear thickening and added relevant references (Line 40).

9. Line 37: What is the typical sedimentation time of the particles?

Calculated as $\frac{9\eta}{2\Delta\rho g a}$, where η is the viscosity of water, $\Delta\rho$ is the difference in densities of silica and water, g is acceleration due to gravity, and a is particle radius, the sedimentation time of the particle is approximately 1.1 seconds.

10. Line 48: Boyer et al. [1] made a different choice to estimate the maximum shear rate in the drop during impact. It would be useful to understand why the authors are making a different choice here.

[1] F. Boyer, E. Sandoval-Nava, J. H. Snoeijer, J. F. Dijksman, and D. Lohse, Phys. Rev. Fluids 1, 013901 (2016).

At the end of paragraph 1, page 5 of Boyer et al., they use the formula $\dot{\gamma} = \frac{u_0}{d_0}$, same as we use on Line 48. Is the referee referring to another work by Boyer et al. that we are not aware of?

12. Line 87: I do not think that Ref. 17 is the best choice to model the maximum spreading of the impacting drop, especially considering that it completely ignores the effect of contact angle, that is probably relevant here. The authors should probably use a more recent theory such as the one presented in Lee et al. [2]. At least, the comparison with this more recent theory should be provided in a Supplementary Material file.

[2] J. B. Lee, N. Laan, K. G. de Bruin, G. Skantzaris, N. Shahidzadeh, D. Derome, J. Carmeliet, and D. Bonn, J. Fluid Mech. 786, R4 (2016).

26. Fig. 4b: I am not convinced that the choice of the effective viscosity or the choice of the model for the maximum spreading diameter are the best to collapse the data. I still see a systematic deviation when increasing the concentration instead of an "excellent agreement".

We have now modified the text to reflect these discrepancies, and also added the above references [Line 128]. Accounting for surface wettability is challenging for colloidal suspensions. However, given that these suspensions are highly shear thinning, we are pleasantly surprised by the agreement we see in the scaling.

16. Line 116-117: Previous studies such as [3–5] had already observed that the top of the drop initially continues to fall at the impact velocity. It would be worth mentioning here.

[3] G. Lagubeau, M. A. Fontelos, C. Josserand, A. Maurel, V. Pagneux, and P. Petitjeans, J. Fluid Mech. 713, 50 (2012).
[4] L. Gordillo, T.-P. Sun, and X. Cheng, J. Fluid Mech. 840, 190 (2018).
[5] B. R. Mitchell, J. C. Klewicki, Y. P. Korkolis, and B. L. Kinsey, J. Fluid Mech. 867, 300 (2019).

We are thankful to the referee for pointing out these relevant studies of Newtonian impact. We have added text to the manuscript comparing our results with the observations mentioned above [Line 152].

17. Fig. 6: The shear rate estimation experienced during impact should have a different name from the shear rate measured in the rheometer, as they are of completely different nature. It is critical to understand clearly which one is used in the discussions and figures.

We are thankful to the referee for raising this important concern. We have now renamed the shear rate estimation at impact as $\dot{\gamma}_{impact}$ to differentiate it from $\dot{\gamma}$ measured in the rheometer.

18. Line 137: I am not convinced that a shear rate above the critical shear rate for jamming is really meaningful. See for example Fig. 6e.

We agree with the referee that Fig. 6e suggests that excess shear rate beyond the critical value is unphysical, and appreciate that further explanation is needed to clarify this important nuance. This is true in a rheometer, as resistance to shear in a thickened fluid causes the shear rate to not increase even though applied shear stress increases. However, we believe that the shear rate estimated by $\dot{\gamma} = \frac{u_0}{d_0}$ is a physically relevant quantity *at the instant of impact*. The rationale is as follows: at the moment of impact (before the front has travelled across the drop over ~100 microseconds, as our results show), the impact point comes to an abrupt halt while the drop apex still moves at the impact velocity (Fig. 5b). Therefore, the velocity difference u_0 exists across the drop size d_0 , leading to the above instantaneous shear rate estimate. The front propagation gives us a timescale over which effects of shear jamming are apparent over the whole drop, and we believe that the inability to access high shear rates in a rheometer is a direct consequence of the longer timescale over which shear is applied in rheometry measurements. We emphasize this ability to apply instantaneous high shear is the greatest strength of studying suspension behaviour via drop impact. As per in the previous comment, separating the name for the two shear rates --- one measured in the rheometer and one estimated for impact experiments --- is a useful suggestion in this regard, and we have made this change throughout the manuscript.

21. Line 177: "in the absence of confinement": This statement implies that the confinement by surface tension in the drop is not important. I would either rephrase the statement, or formulate explicitly this hypothesis with supporting evidence.

We have amended our statement to read 'in free-surface systems' as confinement with surface tension is expected to play a role for the particle size used in our experiments [Line 221].

24. Ref. 21 should be updated.

We have now added the new arXiv preprint to the bibliography.

25. Fig. 4: I think it would be useful to plot the maximum spreading diameter as a function of the effective Reynolds number as well as the Weber number, to understand the range of these parameters covered in the experiments. This could be put in a Supplementary Material file is necessary.

We agree that the range of Reynolds and Weber numbers is key information, as the physics of spreading can vary drastically with this information. We have added to the manuscript the range of both numbers scanned in our experiments [Line 119].

27. Fig. 5b: I think that plotting the ratio u/u0 as vertical axis would make it easier for the reader to understand the deviation from the free fall approximation u*=u0.*

We believe that these two ways of visualising the data are equivalent, however we slightly prefer the plot in its current form.

28. Fig. 5b: I am a bit confused how one data point can be above the dashed line. This would mean that the top of the drop could fall faster than the impact velocity? Some careful checking or comments on this would be useful.

We note that statistically we would expect one-third of the points drawn from a distribution should lie more than a standard deviation away from the mean. Additionally, we believe our argument that the drop apex falls essentially at the impact velocity is strengthened as there are points distributed on both sides of the mean.

Reviewers' comments:

Reviewer #2 (Remarks to the Author):

The authors have mostly addressed my comments, and I still recommend publication in Communications Physics.
However, it is important to remind the authors that all replies discussed in the reply to referees should be added to the manuscript. The review process is not designed to answer the questions of the referees, but to better explain and argue the material presented in the manuscript.

Following my previous comments, I still have the following recommendations:
1) The authors must include the contact angle information in the manuscript to properly describe their experiments. This is critical due to the influence of the contact angle on the maximum spreading of the drop.
2) The authors should also add their estimate of the sedimentation time. They should also comment in the text on the fact that the sedimentation time is probably much shorter than the typical time to complete the experiments, which could lead to significant variations of the volume fraction estimates.
3) The authors misunderstood the work of Boyer et al. Page 5 of Boyer et al. indeed mentions the shear rate U/D. However, they clearly state that it is an "estimate of the minimum shear rate experienced by an impacting drop", and not of an estimate of the maximum shear rate as used in the manuscript. The correct estimate of the shear rate is given on page 6, using the minimal height of the drop rather than the full drop diameter. In the more realistic estimate of Boyer, the typical maximum shear rates experienced during drop impact are not accessible with a typical rheometer, significantly affecting the conclusions of the authors. It is therefore important to either correct the estimate of the maximum shear rate, or at least discuss the different estimate provided by Boyer et al. and its implications. This shear rate estimated by Boyer et al. could be typically an order of magnitude larger, which would also significantly affect the estimated effective Reynolds number.
4) The statement "We note that more recent works [8, 9] have modified the scaling by Scheller et al. [28] to additionally account for surface wettability." is simply wrong. The models proposed in [8, 9] are completely different from the work of Scheller et al. [28]. Moreover, the work of Laan et al. did not include any wettability effect.
5) The authors should better explain in the text what they argue is the physical meaning of the "excess shear rate", as detailed in the reply to referee.
6) Ref. 21 (now 11) is still not updated according to my recommendations: Cheng, X., Sun, T.-P., & Gordillo, L. (2022). Drop Impact Dynamics: Impact Force and Stress Distributions. Annual Review of Fluid Mechanics, 54, 57–81.
7) The reply to my comment 28 does not seem related to my comment. In Fig. 5b, the authors are claiming that for the point above the dashed line, within error bars, the falling velocity of the drop apex is larger than the impact velocity! I only see 2 possibilities from this observation:
- If the authors are correct about the accuracy of their measurements, it means that some mechanism is accelerating the drop apex during impact! This is quite a shocking discovery to me and should be discussed by the authors! Of course, simple estimates can easily prove that gravity is not responsible for this discrepancy.
- Otherwise, it means that the error bars presented in that figure are wrong. In that case, I strongly recommend the authors to double check their measurements and explain how they obtain these error bars.

We thank both the referees for their time and effort, and have addressed all their comments below and in the manuscript. We have also added a Supplementary Information Document discussing some of the finer points in more detail. We are glad to see that both referees recommend the manuscript for publication with these changes.

Below is a point-by-point response, color coded as:

Red: Referee 1
Purple: Referee 2

The authors replied this to my first question in my report:

"We thank the referees for these detailed remarks. Although the above studies of low-viscosity Newtonian fluids have established the velocity and pressure fields within the drop, they are not directly applicable in the case of high-viscosity systems. The highly non-Newtonian nature of colloidal suspensions adds further complexity, as the fluid viscosity is a strong function of the applied shear. We have elaborated on these challenges in comparison to Newtonian studies in a new discussion paragraph (line 193). "

The authors also added this phrase in the manuscript: " Although recent studies of low-viscosity Newtonian fluids have established the velocity and pressure fields within an impacting drop, they are not applicable in case of high-viscosity systems. "

I am still confused. One of the references I suggested (see 1) not only reports a large amount of data for high-viscosity-drop impacts but also provides a theory that matches them well. I made a fast check, and the viscosities reported in the manuscript are in the same range as those studied in the reference. Could this be please checked carefully? Concerning the phrase added, it is misleading and should be rephrased. I propose something like this:

"Although recent studies of Newtonian fluids in a large range of viscosities[7,10] have established the velocity and pressure fields within an impacting drop, they are not applicable in the case of colloidal systems. "

We are thankful to the referee for pointing out the high-viscosity work. We have now updated the statement in the manuscript to reflect this comment (line 234).

1) The authors must include the contact angle information in the manuscript to properly describe their experiments. This is critical due to the influence of the contact angle on the maximum spreading of the drop.

We have now included the contact angle information in the Supplementary Information document and added the contact angle estimate to the manuscript (line 66).

2) The authors should also add their estimate of the sedimentation time. They should also comment in the text on the fact that the sedimentation time is probably much shorter than the *typical time to complete the experiments, which could lead to significant variations of the volume fraction estimates.*

We have added to the manuscript calculations for sedimentation time, both over particle diameter (1.1 s) and over the drop size (~ 4000 s). We have emphasized that samples are vortex mixed before each experimental trial, hence the sedimentation time over drop size is much longer than the typical time to complete the experiments (line 50).

3) The authors misunderstood the work of Boyer et al. Page 5 of Boyer et al. indeed mentions the shear rate U/D. However, they clearly state that it is an "estimate of the minimum shear rate experienced by an impacting drop", and not of an estimate of the maximum shear rate as used in the manuscript. The correct estimate of the shear rate is given on page 6, using the minimal height of the drop rather than the full drop diameter. In the more realistic estimate of Boyer, the typical maximum shear rates experienced during drop impact are not accessible with a typical rheometer, significantly affecting the conclusions of the authors. It is therefore important to either correct the estimate of the maximum shear rate, or at least discuss the different estimate provided by Boyer et al. and its implications. This shear rate estimated by Boyer et al. could be typically an order of magnitude larger, which would also significantly affect the estimated effective Reynolds number.

We are thankful to the referee for pointing this out, we have now edited our shear rate estimate as well as the scaling to reflect this more realistic estimate (line 124 in the manuscript). We note that this change only changed the scaling exponent within error (line 133).

4) The statement "We note that more recent works [8, 9] have modified the scaling by Scheller et al. [28] to additionally account for surface wettability." is simply wrong. The models proposed in [8, 9] are completely different from the work of Scheller et al. [28]. Moreover, the work of Laan et al. did not include any wettability effect.

We have updated the text to clarify this point (line138) and added a more extensive discussion with other scaling plots in the SI document.

5) The authors should better explain in the text what they argue is the physical meaning of the "excess shear rate", as detailed in the reply to referee.

We have now added the text from the response to referees to the manuscript in further detail (line 253).

6) Ref. 21 (now 11) is still not updated according to my recommendations: Cheng, X., Sun, T.-P., & Gordillo, L. (2022). Drop Impact Dynamics: Impact Force and Stress Distributions. Annual Review of Fluid Mechanics, 54, 57–81.

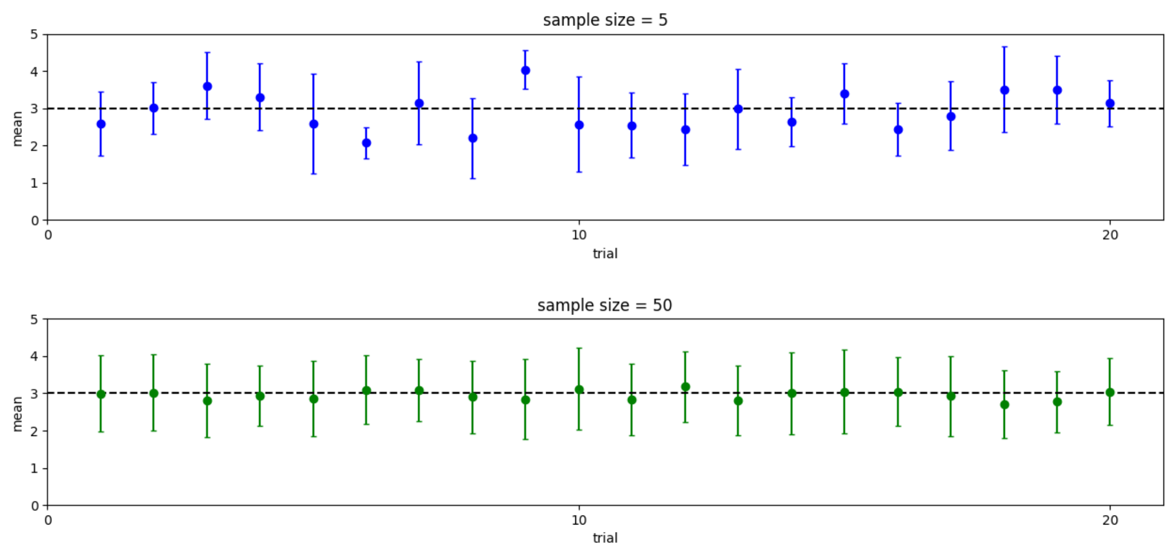
Ref. 11 is updated to the one mentioned above by the referee (line 396).

7) The reply to my comment 28 does not seem related to my comment. In Fig. 5b, the authors are claiming that for the point above the dashed line, within error bars, the falling velocity of the

drop apex is larger than the impact velocity! I only see 2 possibilities from this observation:
- If the authors are correct about the accuracy of their measurements, it means that some mechanism is accelerating the drop apex during impact! This is quite a shocking discovery to me and should be discussed by the authors! Of course, simple estimates can easily prove that gravity is not responsible for this discrepancy.
- Otherwise, it means that the error bars presented in that figure are wrong. In that case, I strongly recommend the authors to double check their measurements and explain how they obtain these error bars.

In Fig. 5b, the fact that one datapoint lies more than an error bar away from the mean does not represent a velocity measurement greater than the impact velocity, but this is the result of a statistical fluctuation due to the small sample sizes in our data (5 datapoints per sample). We elaborate further on this point below, and we have added a clarifying statement in the manuscript.

For all our data including that reported in Fig. 5b, the error bars represent one standard deviation in the measurements, calculated over at least 5 trials. To elaborate further on how a measurement could lie more than one standard deviation away from the true mean, especially for small sample sizes (5 trials in our case), we are attaching synthetic data generated from a gaussian distribution. We plot random samples of size 5 drawn from a gaussian, with a mean of 3 and standard deviation of 1 (top panel):



Every data point in the top panel is the average of 5 samples drawn from the distribution, and error bars are the standard deviation over those 5 samples. For a small number of data points, the error bars around the sample mean do not intersect the true mean (black dashed line), even though they are sampled from the same distribution.

If we increase the size of each sample to 50 (bottom panel), the variation in the sample means and standard deviations is much smaller, and the sample mean always lies within an error bar of the true mean. Therefore, we believe that the point lying above the u*=u_0 line is due to the small sample size in our experiments. To clarify this, we have emphasized the small sample size in the manuscript, and added that error bars of one standard deviation imply a 68% confidence interval (line 339). We have copied the python code to generate the above artificial data at the end of this document.

```
import numpy as np

import matplotlib.pyplot as plt

n = 20

fig, ax = plt.subplots(2,1)

mean = []
error = []
#make n synthetic data points
for i in range(n):


      mu = 3; sigma = 1;
      #draw five values from a gaussian distribution
      values = np.random.normal(mu, sigma, 5)
      #compute the mean and standard deviation of the values
      mean.append(np.mean(values))
      error.append(np.std(values))


ax[0].errorbar(np.arange(n)+1, mean, yerr = error, ls='None', marker =
'o',capsize = 2, c= 'b')
ax[0].plot([0,n+1],[mu,mu],ls='--',c='k')
ax[0].set_ylim(0,5)
ax[0].set_xlim(0,n+1)
ax[0].set_ylabel('mean')
ax[0].set_xlabel('trial')
ax[0].set_title('sample size = 5')
ax[0].set_xticks([0,10,20])

mean = []
error = []
#make n synthetic data points
for i in range(n):


      mu = 3; sigma = 1;
      #draw fifty values from a gaussian distribution
      values = np.random.normal(mu, sigma, 50)
      #compute the mean and standard deviation of the values
      mean.append(np.mean(values))
      error.append(np.std(values))


ax[1].errorbar(np.arange(n)+1, mean, yerr = error, ls='None', marker =
'o',capsize = 2, c= 'g')
ax[1].plot([0,n+1],[mu,mu],ls='--',c='k')
ax[1].set_ylim(0,5)
ax[1].set_xlim(0,n+1)
ax[1].set_ylabel('mean')
ax[1].set_xlabel('trial')
ax[1].set_title('sample size = 50')
ax[1].set_xticks([0,10,20])

plt.tight_layout()
plt.show()
```

REVIEWERS' COMMENTS:

Reviewer #2 (Remarks to the Author):

The authors have answered all my comments. The work presented in this manuscript is excellent and the publication will probably have a significant impact in the research field. I am therefore glad to recommend publication in Communications Physics.

I just have a very minor comment that the authors can consider before publication if they want. I do not need to revise the reply of the authors before publication.
1) I think we agree that all physical values of u* must be smaller than u0. Any value larger than u0 is most likely wrong physically, and therefore reflect some kind of measurement error.
2) The authors have now clarified that the error bars do not quantify a physical estimation of the measurement error, but are just “one standard deviation over at least 5 trials”.
3) Having a data point significantly above u0 is a strong indication that the authors have some significant error in their measurement method, at least larger than the distance to u0.
4) The measurement error shown here, with the physically wrong values of u*, is of course reasonable and fine for publication. But I feel that it is a larger measurement error than most publications in the field.
5) Providing more information on the definitions and measurement methods would probably clarify the origin of this measurement error:
- How is u* measured? Is it obtained from some kind of arbitrary fit as displayed in Fig. 5a? Or is based on a rigorous measurement protocol? How are the starting time and end time chosen to obtain the fitted value? Is u* obtained by least square fitting of the data points?
- What is the resolution of the images used for the measurements of h, the physical size of the pixels? This will give some idea of the measurement accuracy in Fig. 5a.
- How is the impact velocity obtained? Is it a theoretical value (free fall?), or is it measured experimentally for each impact?
I guess that these clarification steps should help the authors identify the origin of their measurement error, and eventually correct it, or give the reader the information to judge the accuracy of the data provided.

We thank the referee for their time to review our revised manuscript one more time, and we are glad to know that the referee finds our answers satisfactory and recommends our manuscript for publication. Below, we address the referee comments.

Reviewer #2 (Remarks to the Author):
The authors have answered all my comments. The work presented in this manuscript is excellent and the publication will probably have a significant impact in the research field. I am therefore glad to recommend publication in Communications Physics.

I just have a very minor comment that the authors can consider before publication if they want. I do not need to revise the reply of the authors before publication.

1) I think we agree that all physical values of u must be smaller than u0. Any value larger than u0 is most likely wrong physically, and therefore reflect some kind of measurement error.*
2) The authors have now clarified that the error bars do not quantify a physical estimation of the measurement error, but are just "one standard deviation over at least 5 trials".
3) Having a data point significantly above u0 is a strong indication that the authors have some significant error in their measurement method, at least larger than the distance to u0.
4) The measurement error shown here, with the physically wrong values of u, is of course reasonable and fine for publication. But I feel that it is a larger measurement error than most publications in the field.*

We are pleased to know that our clarification of the error bar calculation is satisfactory.

5) Providing more information on the definitions and measurement methods would probably clarify the origin of this measurement error:
- How is u measured? Is it obtained from some kind of arbitrary fit as displayed in Fig. 5a? Or is based on a rigorous measurement protocol? How are the starting time and end time chosen to obtain the fitted value? Is u* obtained by least square fitting of the data points?*
- What is the resolution of the images used for the measurements of h, the physical size of the pixels? This will give some idea of the measurement accuracy in Fig. 5a.
- How is the impact velocity obtained? Is it a theoretical value (free fall?), or is it measured experimentally for each impact?
I guess that these clarification steps should help the authors identify the origin of their measurement error, and eventually correct it, or give the reader the information to judge the accuracy of the data provided.

We thank the referee for these suggestions. We have now added this information to the methods section of the manuscript, and reiterated how the error bars are calculated in the figure captions.