Peer Review File

Manuscript Title: Shell Buckling for Programmable Metafluids

Reviewer Comments & Author Rebuttals

Reviewer Reports on the Initial Version:

Referees' comments:

Referee #1 (Remarks to the Author):

A. Summary of the key results

A new class of "metafluid" composed of highly deformable air-filled capsules suspended in an incompressible surrounding fluid is presented. By harnessing the shell buckling of these quite easy-to-make building blocks, the authors can reversibly tune the elastic, optical and rheological properties of these suspensions at different scales ranging from the micrometer to the centimeter. A large number of configurations are presented in this work paving the way for numerous applications in various areas such as soft robotics.

B. Originality and significance:

Buckling of viscoelastic spherical shells has been already intensively studied but harnessing this mechanism for the applications presented in this work is undoubtedly original and significant.

C. Data & methodology:

The approach developed by the authors is very convincing. Data quality is good but the quality of presentation might be improved by clarifying the figures. As an example, the schemata shown in Figure 1D that illustrate the micrometer-scale capsules fabrication is misleading for the reader since one can think the capsules are filled with water. Obviously, these objects are filled with air but this information is hidden in the supplementary materials and not easy to find. As another example, Figure 3 is really too busy and difficult to read in my opinion. The authors may split it into several figures for the sake of clarity.

D. Appropriate use of statistics and treatment of uncertainties

Statistics were not used here and treatment of uncertainties is correct.

E. Conclusions:

On my opinion, the conclusions are robust, valid and reliable. The take-home messages are quite clear even if the potential applications might be expanded.

F. Suggested improvements:

1) In several places in the text, the authors claim that shell buckling is reversible. Nevertheless, that does not seem to be the case when looking at the end of the cycle not shown in the first movie. Could the authors comment on that point?
2) All experiments were only performed under one loading/unloading cycle. What happens for several cycles? Do performances deteriorate? For all mentioned potential applications, that must be a crucial point to be investigated in this paper.
3) How the elastic properties of the shell were characterized? What is their bulk modulus? Does this parameter have an influence on shell buckling?
4) What about the shell viscoelasticity (corresponding to the imaginary part of the shear modulus)? Is this parameter important?
5) As far as I understood, the metafluids comprising micrometer-scale capsules are suspended in silicone oil unlike centimeter-scale capsules are suspended in water? Why choose this viscous fluid for smallest capsules? What is its viscosity? Does it have any influence on shell buckling dynamics?
6) Typo remark: in page 4 "(realized by filling the WITH water and 12 shells…)
7) The authors claim that the improved transmission (and optical contrast) beyond a critical pressure is due to the collapse of capsules that behave like small optical lenses focusing incoming light. Are all collapsed capsules oriented along the same direction? If not, all these small lenses will focus in random directions providing a blurry image, isn't it? Could the authors comment more on that point?

G. References:

The references are appropriate and do not need to be completed by other works.

H. Clarity and context:

To conclude, this is a very interesting work with appropriate abstract and clear summary. The introduction is well written and the conclusions sum up the main points of this study quite well. However, there are so many (experimental and numerical) results on so many configurations that this paper is not always easy to follow. On my opinion, some clarifications on the above-mentioned points should be provided for possible publication in Nature.

Referee #2 (Remarks to the Author):

Metamaterials with enhanced functionalities have been revolutionizing a variety of scientific fields in recent years. For instance, these materials use their very structure in lieu of their composition to obtain unique acoustic, thermal, and mechanical properties. Routes for the design and fabrication of these materials have been largely studied in the soft matter community. Yet, as rightfully pointed out by the authors, those systems are primarily solid, leaving their liquid counterparts relatively unexplored. Starting from this observation the authors have devised a meta-fluid system. The idea is beautiful: they use thin elastomeric and deformable spherical capsules enclosing air that they embed in an incompressible liquid. When compressed, the pressure in the system increases, as one would expect from a compressible fluid, before experiencing a sudden drop as one or more capsule buckle. The unloading pathway differs from this loading scenario, yielding a nonlinear and hysteretic pressure-volume curve. This relation between pressure and volume is the core of the author's work; and everything in the paper unfolds from that nonlinearity. They first rationalize this curve, demonstrate its universal character, and then exemplify its applicability to a variety of systems.

Overall, I am enthusiastic about the work, and I find the system reported by the authors to be novel and potentially exciting to a broad readership. I would certainly be supportive of publication in Nature if the authors can address the following points.

1. The idea reported in the manuscript is intriguing as the author produce a new type of fluid with unique and tunable properties. Yet, too rarely the authors envision that these fluids might flow. This is somehow disappointing… To be clear: I appreciate that the system should first be studied in a quasi-static framework and in quiescent conditions. But the fluid that surrounds the capsules should be more than a way to load these capsules. Therefore, the discussion of Fig4 D and E should be largely expended. Likewise, too little is said about the rheology of this peculiar type of suspension. A detailed study should be provided for the microscopic system: what is the rheology of this fluid, i.e., its storage and loss modulus. Surely these properties depend on the shear rate… I would advise to move some of the other applications from the main text to extended figures to make up some room for a new and extended discussion on rheology and fluid dynamics. In particular the collapsed spheres appear to jam. See for example: Bourrianne, Philippe, et al. "Tuning the shear thickening of suspensions through surface roughness and physico-chemical interactions." Physical Review Research 4.3 (2022): 033062.

2. The authors demonstrate the generic nature of their idea, going from macroscale to microscale. This is certainly an asset of the work; yet little is provided in terms of a global understanding of the physics of the problem. I appreciate that the authors have FEM simulations reproducing most features of the system, but I would love to see more scaling laws and theory, which will ultimately help providing design guidelines. Specifically, it would be helpful to know when the plateau in pressure occurs and how long it is. Likewise, the results somehow resemble well known pressure-volume nonlinear relation, such as the one found when inflating a hyper-elastic balloon. The similarities and differences with this system should be discussed.

3. While the figures are superb, the manuscript needs significant re-writing to be adapted to a broad readership. Too many details of the method (experimental and numerical) crowd the results that the authors are reporting. More emphasis should be brought to the said results. Likewise, the manuscript appears to be missing a full-fledged introduction. I appreciate that the abstract serves this purpose to some extent, yet, it would certainly serve the manuscript to detail further relevant aspects of the

context of this study.

Some more minor comments:

1. The fabrication technique of the large capsules is like that reported in E. JambonPuillet et al. Nature Phys 2020, which should thus be cited.
2. What explains the qualitative difference between the curves in Fig1C and E?
3. What if the pressure is not uniform across the sample? As one would expect in an flow. How would the metafluid then behave? Should this point be a concern for the programmability of the system?
4. In Fig3A, why not putting the dotted line plateau in between Phold and Pcrush?

In conclusion, I would like to reiterate my support toward publication. The main reasons for that positive assessment are the novelty of the work, the elegance of the system and its potential impact to the scientific community.

Referee #3 (Remarks to the Author):

The paper " Metafluids: harnessing shell buckling for programmable response" by Djellouli and al. introduces suspensions made from hollow shells that can collapse when the hydrostatic pressure of the surrounding fluid exceeds a certain threshold. They characterize and model the behavior of single particles as well as collections of such shells and show that the suspension as a whole exhibits enhanced compressibility as a result of the collapse of the shells and a large hysteresis in the pressure-volume curve. In fact in the case where the shell are cm-sized, the compressibility become effectively infinite: the pressure plateaus as the volume is increased. They also fabricate sub-mm sized shells and in this case, the compressibility is not infinite, yet it becomes much larger. In a second time, they demonstrate how to use these suspensions in gripping and mechanical computing applications. The enhanced compressibility provides an advantageous saturation of the gripping force, which in turn makes a forgiving gripper than can grab fragile objects. The nonlinearity of the pressure-volume curve is also used to create mechanical logic gates. Finally, they exploit the shape-change of the suspension's shells from spherical to flat to achieve tunable light absorption and rheology.

Overall, although the concept of suspensions made from collapsing particles that lead to a highly nonlinear pressure-volume curve with enhance compressibility is not new (e.g. ref 27), many ideas put forward in this manuscript are novel and stimulating: the idea of using simple spherical shells, their manufacturing at the sub-millimeter scale and their use in applications such as gripping, logic, optics and rheology are all innovative and could lead to better integrated adaptive mechanical response. Embedding functionality in the fluid is complementary to the approach traditionally employed in mechanical metamaterials, where the functionality is integrated in elastic structures instead and could help applications in robotics and medicine applications.

However, some results appear incomplete and the understanding of the phenomena appear a bit too preliminary for a journal of the caliber of Nature. In particular, where the modeling of single shells and of shells in hydrostatic condition is well treated, I would except a more thorough treatment of the metafluid-structure interaction that leads to a delayed bulging of the tube (Fig. 3b) as well as a more in depth treatment of the rheology from a theoretical and/or computational perspective (Fig. 4DE). How do the shells flow? How is their buckling distributed in space? Would it be possible to propose a continuum model for the flow of such metafluid?

In conclusion, although I find the idea put forward in the paper stimulating and of potential broad relevance, I feel that the paper at its current stage of development is not yet suitable for a top-tier journal like Nature. I list a few more points below, which might help the authors to further improve their manuscript.

*1 Could the author explain why the pressure-volume curve in the case of the sub-mm shells does not exhibit the clean plateau of the cm-shell? Is it due to polydispersity?

*2. For the logic gate (fig 3C). What is the benefit of using such fluid instead of using valves? In this case it is not clear to me that the metafluid offers a better level of integration, since many mechanical apparatus are used in the setup.

*3 on the same figure 3c. I find that the setup and experimental protocol are not clearly illustrated and explained.

*4 on fig 3B. How would you model such metafluid at the continuum level? is the shape of the bulge different between the metafluid and ordinary incompressible fluids?

*5. on fig 4DE: The rheology should be better described using theory or computation (e.g. Lattice Boltzmann could be a suitable technique here). Can the shear flow also deform the shell and hence affect their buckling? Do the particles migrate in the Poiseuille flow of fig 4d?

Typo:
" Further, we point out that if THE pressure"

Author Rebuttals to Initial Comments:

Response to the Referees

Shell Buckling for Programmable Metafluids
Manuscript number: 2023-05-09013B

Adel Djellouli[a,1], Bert Van Raemdonck[b,1], Yang Wang[a,1], Yi Yang[a], Anthony Caillaud[a], David Weitz[a], Shmuel Rubinstein[c], Benjamin Gorissen[b,2], and Katia Bertoldi[a,2]

[a]J.A. Paulson School of Engineering and Applied Sciences, Harvard University, USA; [b]Department of Mechanical Engineering, KU Leuven and Flanders Make, Belgium; [c]The Racah Institute of Physics, The Hebrew University, Israel; [1]These authors contributed equally.; [2]These authors contributed equally.

Response to Referee #1

A new class of "metafluid" composed of highly deformable air-filled capsules suspended in an incompressible surrounding fluid is presented. By harnessing the shell buckling of these quite easy-to-make building blocks, the authors can reversibly tune the elastic, optical and rheological properties of these suspensions at different scales ranging from the micrometer to the centimeter. A large number of configurations are presented in this work paving the way for numerous applications in various areas such as soft robotics..

We thank the Referee for the thorough review and the in-depth comments that helped us improve the quality of the manuscript. We are glad to see that they acknowledge the novelty of harnessing shell buckling to program the response of a fluid.

Comment 1A

In several places in the text, the authors claim that shell buckling is reversible. Nevertheless, that does not seem to be the case when looking at the end of the cycle not shown in the first movie. Could the authors comment on that point?

As described in our response to Comment 1B, we performed additional experiment where we subjected our metafuid to 100 loading/unloading cycles. Our experimental results indicate the behavior of the metafluid between cycles is almost identical. Additionally, in Fig. R1 we focus on a single shell and show both its pressure-volume response and experimental snapshots at the beginning, immediately after buckling and at the end of a loading/unloading cycle. They clearly show that the shell buckling is fully reversible as the shell goes back to its initial spherical shape at the end of the cycle.

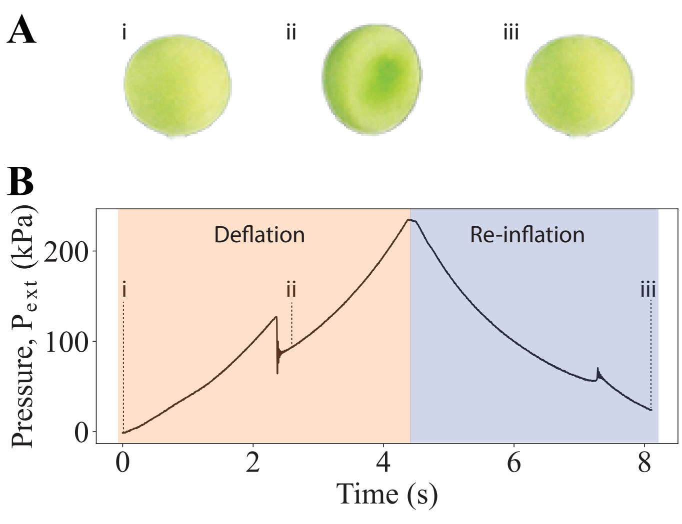

Fig. R1. Reversibility in shape. (A) Experimental snapshots of one shell at the beginning, immediately after buckling and at the end of a loading/unloading cycle. (B) Evolution of pressure as function of time during a loading/unloading cycle.

Comment 1B

All experiments were only performed under one loading/unloading cycle. What happens for several cycles? Do performances deteriorate? For all mentioned potential applications, that must be a crucial point to be investigated in this paper.

To address this point, we have conducted additional experiments on a metafluid with initial bulk modulus $K_0 = 6$ MPa and critical buckling pressure $P_{cr}^{up} = 125kPa$ (realized by filling a container with $V_{tot} = 60$ ml with water and one capsule with $G = 350kPa$, $t = 2$ mm and $R_o = 10$ mm). In these experiments, we performed 110 loading cycles at a frequency of about 0.1 Hz, using a syringe pump (Harvard apparatus 33 DDS) and measured the pressure inside the container with a differential pressure sensor (60 PSI Ashcroft® GV Pressure Transducer). The results shown in Fig. R2 indicate that the behaviour of the metafluid is mostly invariant. These results are consistent with recent research that has demonstrated the stability of nonlinearities in the pressure-volume domain over for 50.000 cycles (1). Nevertheless, it is important to acknowledge that the long-term behavior of the capsules is influenced by the diffusion of air through their walls, posing a potential source of alteration.

To clarify this important point we have added the following text to Supplementary Information (Section S2.1)

> "In addition to the results presented in the main text, we also characterize the effect of both cyclic loading and the loading rate on the pressure-volume characteristics of the metafluid. In order to assess the effect of cyclic loading, we place a capsule with $R_o = 10$ mm and $t = 2$ mm in a container with volume $V_{tot} = 60$ ml and perform 110 loading/unloading cycles. As shown in Fig. R2, we find that pressure volume curves are mostly invariant, except for a small reduction in the critical snapping pressures which decrease by 5 kPa over more than 100 cycles. As such, these results indicate that cyclic loading has a minimal effect on the behavior of metafluids comprising centimeter scale capsules. Nevertheless, it is important to acknowledge that the long-term behavior of the capsules may be influenced by the diffusion of air through their walls, posing a potential source of alteration."

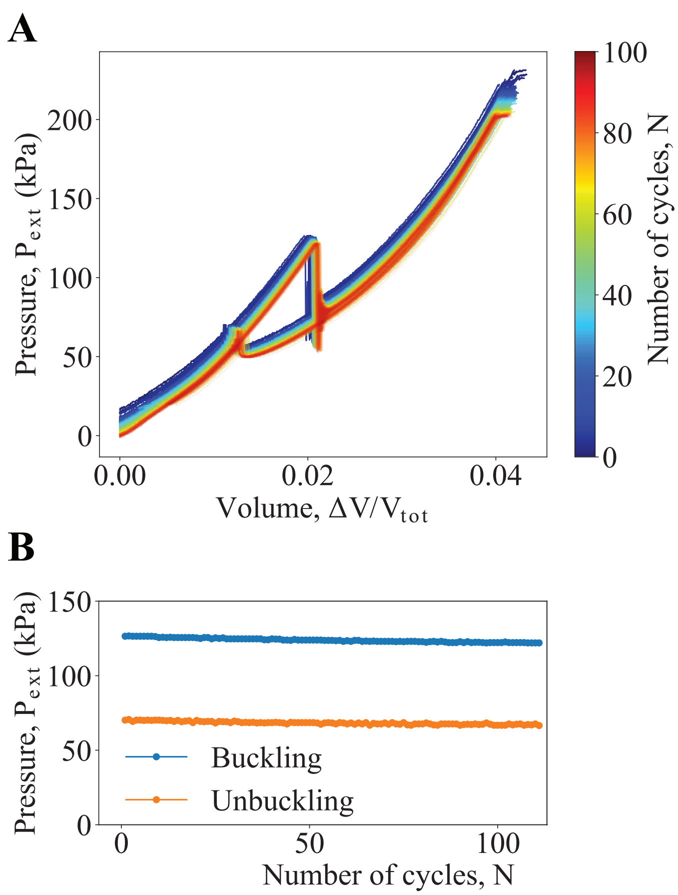

Fig. R2. **Cycling loading of a centimeter scale capsule** (A) Pressure-volume curve recorded over 110 loading/unloading cycles. (B) Evolution of the critical pressures over the cycles.

Comment 1C

How the elastic properties of the shell were characterized? What is their bulk modulus? Does this parameter have an influence on shell buckling?

To characterize the response of the elastomers used to fabricate the shells both at the macro- and micro-scale, we conducted uniaxial tensile test on dogbone samples made of Zhermack Elite Double 32 (green rubber) and PDMS. To describe this, we have expanded Section S3.1 of Supporting Information to read as

> "**Material model.** To determine hyperelastic material models capable of adequately capturing the response of the silicone rubber used to make the capsules, we perform uniaxial tensile tests on dogbone samples made out of PVS (Green silicone rubber) and PDMS. For the PVS material we find an almost linear relation between true stress and true strain which is accurately captured by fitting a Neo-Hookean model to the data (Fig. R3A). Differently, for PDMS the Neo-Hookean model fails to capture its stress-strain response, but models with two parameters such as the first order Mooney-Rivlin and Ogden models closely follow the data (Fig. R3B). The coefficients that produce the optimal fits for these models are shown in table R1, where $\lambda = 1 + \epsilon$ represents the stretch. Note that all models assume that the material is incompressible. Even though for PDMS the different models produce different estimates for the initial shear modulus of the material G, the pressure-volume characteristics of a PDMS capsule align well when the pressure is normalized by the shear modulus (Fig. R3C). The reason is that for small strains each of the stress-strain relations in table R1 reduces to the linear form $\sigma = 3G\epsilon$ and that the deformed shape of a shell is independent of $\Delta P/G$ (2)."

Material	Model	Stress-strain relation	Parameters	Shear modulus
PVS	Neo-Hookean	$\sigma = 2C_{10}\left(\lambda^2 - \frac{1}{\lambda}\right)$	C_{10} = 215	G = 430
PDMS	Neo-Hookean	$\sigma = 2C_{10}\left(\lambda^2 - \frac{1}{\lambda}\right)$	C_{10} = 306	G = 612
PDMS	Mooney-Rivlin	$\sigma = 2\left(C_{10} + \frac{C_{01}}{\lambda}\right)\left(\lambda^2 - \frac{1}{\lambda}\right)$	C_{10} = 685, C_{01} = -499	G = 371
PDMS	Ogden	$\sigma = \frac{2\mu}{\alpha}\left(\lambda^{\alpha} - \frac{1}{\lambda^{\alpha/2}}\right)$	μ = 453, α = 4.55	G = 453

Table R1. Material parameters for both PVS (green rubber) and PDMS.

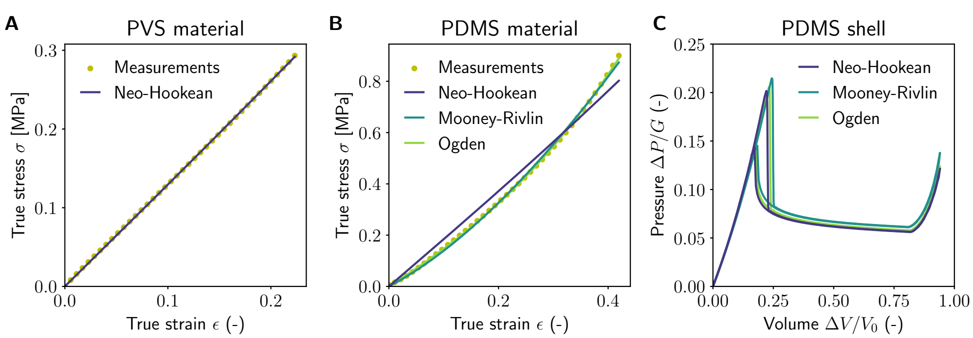

Fig. R3. Material model. (A)-(B) Experimentally measured (markers) and numerically predicted (solid lines) true stress-strain curve for a dogbone sample of (A) PVS and (B) PDMS under uniaxial tension. (C) Simulated pressure-volume characteristics for a shell with $\eta = 0.22$ with the fitted material models for PDMS.

Regarding the bulk modulus, it is well known that both elastomers are nearly incompressible. Consequently, they are characterized by ν approaching 0.5 and K tending towards infinity (where ν and K represent the initial Poisson's ratio and bulk modulus, respectively). Considering the classical buckling pressure of elastic thin spherical shells, which is given by

$$p_{cr} = \frac{2E}{\sqrt{3(1-\nu^2)}}\left(\frac{t}{R}\right)^2, \qquad \text{[R1]}$$

we anticipate that as ν approaches 0.5, the value of p_{cr} will increase. It's worth mentioning that recent research has demonstrated that Eq. (R1) effectively describes also the response of thick spherical shells (3).

In response to this comment, we have added the following text to Supporting Information

> **Effect of bulk modulus.** It is well known that elastomers are nearly incompressible. Consequently, they are characterized by ν approaching 0.5 and K tending towards infinity (where ν and K represent the initial Poisson's ratio and bulk modulus, respectively). Considering the classical buckling pressure of elastic thin spherical shells, which is given by (4)

$$\Delta P_{cr}^{up} = \frac{2E}{\sqrt{3(1-\nu^2)}}\eta^2 \qquad \text{[R2]}$$

By introducing

$$E = \frac{9KG}{3K+G} \text{ and } \nu = \frac{3K-2G}{2(3K+G)} \qquad \text{[R3]}$$

Eq. (R2) can be rewritten in terms of K/G as

$$\frac{\Delta P_{cr}^{up}}{G} = 4\frac{\frac{K}{G}}{\sqrt{\left(\frac{K}{G}\right)^2 + \frac{4}{3}\frac{K}{G}}}\eta^2. \qquad \text{[R4]}$$

As such, we anticipate that as K becomes larger, the value of ΔP_{cr}^{up} will increase asymptotically. To verify this point, we perform a number of FE simulations on a shell with $\eta = 0.22$ where we systematically vary the normalized bulk modulus K/G from 10^1 to 10^9 (note that for PDMS $K/G \approx 5 \times 10^3$ (5)). The results of Fig. R4 show that for $K/G >$ 1e3 the change in ΔP_{cr}^{up} is less than 0.1 as the K is varied from 10^3 to 10^9. Further, in Fig. R4B we compare the evolution of ΔP_{cr}^{up} as a function of K as predicted by Eq. (R4) with the data extracted from our FE simulations. Again, we find that the dependence of ΔP_{cr}^{up} on K is weak. Finally, in Fig. R4C we show the evolution of the critical volumes on volume controlled loading and unloading as a function of K and again find a weak dependence. Since approximating the material as incompressible introduces a negligible error, we use a fully incompressible material model in the simulations.

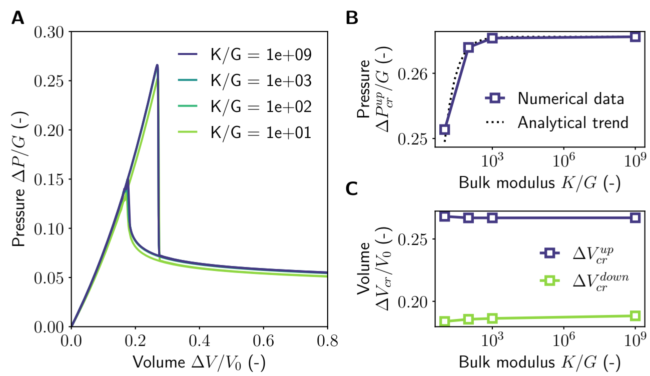

Fig. R4. Influence of the material bulk modulus. **A**, Simulated pressure-volume characteristics for a shell with $\eta = 0.22$ made out of a Neo-Hookean material for different values of the normalized bulk modulus K/G. **B**, Relation between the normalized bulk modulus K/G and the normalized shell critical pressure $\Delta P_{cr}^{up}/G$. The dashed line represents the analytical scaling law given by Eq. (R4). **C**, Relation between the normalized bulk modulus K/G and the change in volume at which the instabilities on volume-controlled loading (ΔV_{cr}^{up}) and unloading (ΔV_{cr}^{down}) occur for the curves plotted in A.

Comment 1D

What about the shell viscoelasticity (corresponding to the imaginary part of the shear modulus)? Is this parameter important?

As correctly pointed out by the Reviewer, our modeling approach does not account for viscoelasticity. This omission stems from the observation that, for both rubber materials utilized in our experiments (specifically, Zhermack Elite Double 3 and PDMS), the loss modulus is at least one order of magnitude lower than the storage modulus (6). Consequently, although there may be a delayed response (7), our spherical shells have the capacity to undergo rapid snapping at short time scales. This observation is corroborated by the experimental findings presented in Figure S6 of the Supplementary Information. In this figure, we demonstrate that the pressure-volume curve of the metamaterial remains nearly constant across various loading rates, spanning from 0.01 mm/s to 10 mm/s.

Comment 1E

As far as I understood, the metafluids comprising micrometer-scale capsules are suspended in silicone oil unlike centimeter-scale capsules are suspended in water? Why choose this viscous fluid for smallest capsules? What is its viscosity? Does it have any influence on shell buckling dynamics?

Our micrometer-scale capsules were suspended in two different media, namely glycerol (Figure 1 of the main text) and silicone oil (Figure 4 of the main text). The choice of glycerol was primarily influenced by the requirement for extended duration static experiments, where a viscous fluid like glycerol effectively prevented air from diffusing out of the PDMS membrane. In the case of Figure 4, we opted to suspend the micrometer-scale capsules in silicone oil due to the necessity for index-matching in the optical experiments showcased in Figure 4A of the main text. For consistency, the same suspension medium was utilized for Figures 4D and 4E. It's worth noting that the viscosity of the PDMS solution used in Figure 4 is 100 cSt (as detailed in Section S2.2 of Supplementary Information).

Finally, it is important to highlight that research has demonstrated that, across a wide range of viscosities, the speed of buckling remains unaffected by the viscosity of the external fluid (8).

Comment 1F

The authors claim that the improved transmission (and optical contrast) beyond a critical pressure is due to the collapse of capsules that behave like small optical lenses focusing incoming light. Are all collapsed capsules oriented along the same direction? If not, all these small lenses will focus in random directions providing a blurry image, isn't it? Could the authors comment more on that point?

This is a good point. As shown in Fig. 3C-(i) of the revised main text, spherical and collapsed capsules exhibit distinct scattering behaviors when interacting with incident rays. To further investigate the influence of these diverse scattering behaviors on the optical characteristics of the metafluid, we conducted additional simulations where we investigated the response or larger arrays of capsules. More specifically, we considered a dense cluster of capsules (with outer radius $R_o = 250\mu m$) arranged on a 8×8×8 FCC lattice with a volume fraction of 0.6 (resulting to a center-to-center distance of $2.06R_o$ between neighboring spheres). We explored three scenarios: (1) an array of spherical capsules, (2) an array of randomly oriented collapsed capsules all with the convex side facing the rays, and (3) an array of randomly oriented collapsed capsules. Note that we chose all collapsed capsules to have the same projected area as the spherical ones. In the simulations rays arranged on a hexapolar pattern with a power of 1 W and a radius $R_{beam} = 500\mu m$ are initiated 5 mm away from the array of capsules. To quantify the light scattering behavior of the three considered arrays of capsules, we monitored the light reflected onto a square screen with an edge length of 15 mm positioned at a distance of 15 mm from the arrays (Fig. R5A).

In Fig. R5B we show the power density collected on the screen for the three considered arrays of capsules. We find that the transmitted power is much larger for the two arrays of collapsed capsules. Furthermore, we observe that the randomly oriented array of collapsed particles proves more effective in light transmission. To quantify this effect, we calculate the transmission, T, defined as the power collected on the panel divided by the input power. The results reported in Figure R5C show that the transmission for the array of ordered collapsed capsules is $\approx$ 100 larger than that of the array of spherical microcapsules - an increase that can be attributed to the lensing capability of the collapsed particles. Finally, when we compare the arrays of collapsed ordered and randomly oriented capsules, we find that the transmission is $\approx$ 60% higher for the latter. Such increase can be attributed to the decreased projected area of the randomly oriented capsules in the plane perpendicular to the incoming rays. Indeed when we measure the coverage area for the arrays of ordered and randomly oriented collapsed capsules, we find that the latter is reduced by 58. As such, these results suggest that the experimentally observed tunable optical behavior of our metafluid can be ascribed to a combination of the lensing effect and the reduced capsule coverage area within the randomly oriented array of collapsed capsules.

The following text has been added to Section S3.3 of Supporting Information to describe these new results

> **Array of capsules.** We also considered a dense cluster of capsules (with outer radius $R_o = 250\mu m$) arranged on a 8×8×8 FCC lattice with a volume fraction of 0.6 (resulting to a center-to-center distance of $2.06R_o$ between neighboring spheres). We explored three scenarios: (1) an array of spherical capsules, (2) an array of randomly oriented collapsed capsules all with the convex side facing the rays, and (3) an array of randomly oriented collapsed capsules. Note that we chose all collapsed capsules to have the same projected area as the spherical ones. In the simulations rays arranged on a hexapolar pattern with a power of 1 W and a radius $R_{beam} = 500\mu m$ are initiated 5 mm away from the array of capsules. To quantify this light scattering behavior of the three considered arrays of capsules, we monitored the light reflected onto a square screen with an edge length of 15 mm positioned at a distance of 15 mm from the arrays (Fig. R5A). In Fig. R5B we show the power density collected on the screen for the three considered arrays of capsules. We find that the transmitted power is much larger for the two arrays of collapsed capsules. Furthermore, we observe that the randomly oriented array of collapsed particles proves more effective in light transmission. To quantify this effect, we calculate the transmission, T, defined as the power collected on the panel divided by the input power. The results reported in Figure R5C show that the transmission for the array of ordered collapsed capsules is $\approx$100 larger that that of the array of spherical microcapsules - an increase that can be attributed to the lensing capability of the collapsed particles. Finally, when we compare the arrays of collapsed ordered and randomly oriented capsules, we find that the transmission is $\approx$60 higher for the latter. Such increase can be attributed to the decreased projected area of the randomly oriented capsules in the plane perpendicular to the incoming rays. Indeed when we measure the coverage area for the arrays of ordered and randomly oriented collapsed capsules, we find that the latter is reduced by 58. As such, these results suggest that the experimentally observed tunable optical behavior of our metafluid can be ascribed to a combination of the lensing effect and the reduced capsule coverage area within the randomly oriented array of collapsed capsules.

Further, we have modified the main text to read as

> "As shown in Fig. 3C-(i), the simulations show that spherical and collapsed capsules exhibit distinct scattering behaviors. When we then measure the power of the transmitted light through a microsuspension with $\varphi \approx 0.4$ and $P_{cr}^{up} \approx 380$, we find that the transmittance T suddenly increases from $T \approx 8$ to $T \approx 30$ at $P_{ext} \approx P_{cr}^{up}$ (Fig. 3c-(ii)). This increase can be attributed to a combination of the lensing effect, and the reduction in the coverage area of the capsules in the collapsed state (see Supplementary Materials for details)."

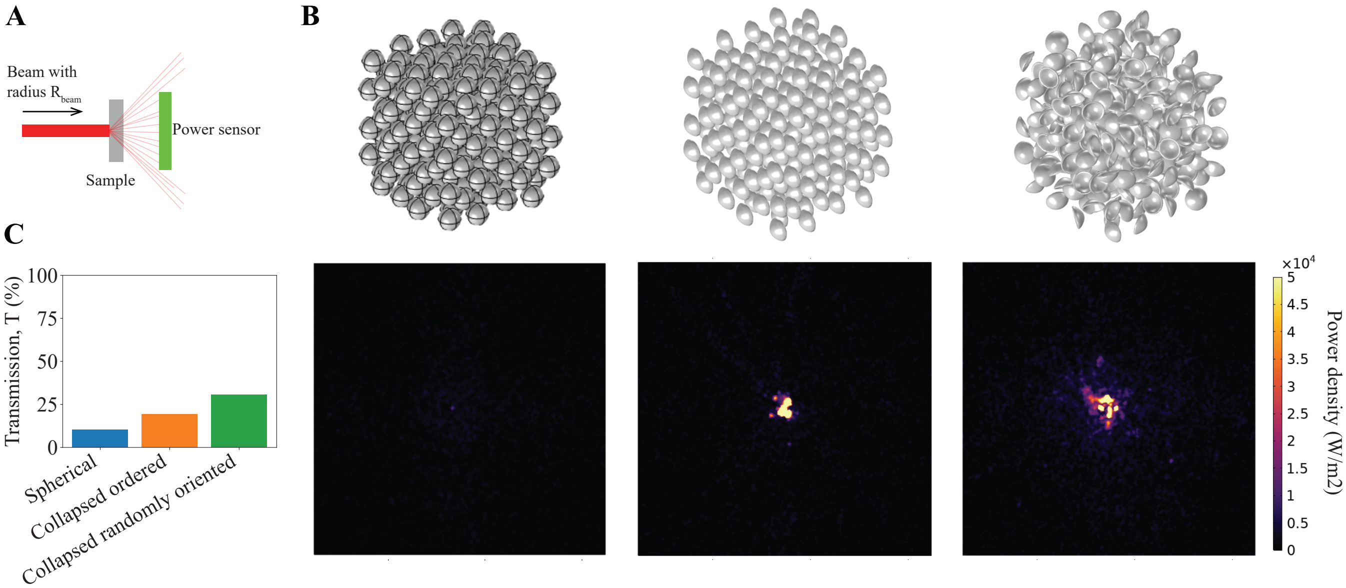

Fig. R5. Ray tracing simulations for a cluster of capsules. A Schematics of the simulation setup. **B** Heat map plots of the transmitted power density in the three different scenarios considered: cluster of spheres (left), cluster of collapsed capsules with their convex side facing the incident beam (middle) and cluster of randomly oriented collapsed capsules (right). **C** Transmitted power measured for the three different scenarios divided by the input power.

Response to Referee #2

Metamaterials with enhanced functionalities have been revolutionizing a variety of scientific fields in recent years. For instance, these materials use their very structure in lieu of their composition to obtain unique acoustic, thermal, and mechanical properties. Routes for the design and fabrication of these materials have been largely studied in the soft matter community. Yet, as rightfully pointed out by the authors, those systems are primarily solid, leaving their liquid counterparts relatively unexplored. Starting from this observation the authors have devised a meta-fluid system. The idea is beautiful: they use thin elastomeric and deformable spherical capsules enclosing air that they embed in an incompressible liquid. When compressed, the pressure in the system increases, as one would expect from a compressible fluid, before experiencing a sudden drop as one or more capsule buckle. The unloading pathway differs from this loading scenario, yielding a nonlinear and hysteretic pressure-volume curve. This relation between pressure and volume is the core of the author's work; and everything in the paper unfolds from that nonlinearity. They first rationalize this curve, demonstrate its universal character, and then exemplify its applicability to a variety of systems. Overall, I am enthusiastic about the work, and I find the system reported by the authors to be novel and potentially exciting to a broad readership. I would certainly be supportive of publication in Nature if the authors can address the following points.

We are pleased that the Referee finds our work potentially ex citing to a broad readership and is supportive of publication in Nature.

Comment 2A

The idea reported in the manuscript is intriguing as the author produce a new type of fluid with unique and tunable properties. Yet, too rarely the authors envision that these fluids might flow. This is somehow disappointing... To be clear: I appreciate that the system should first be studied in a quasi-static framework and in quiescent conditions. But the fluid that surrounds the capsules should be more than a way to load these capsules. Therefore, the discussion of Fig4 D and E should be largely expended. Likewise, too little is said about the rheology of this peculiar type of suspension. A detailed study should be provided for the microscopic system: what is the rheology of this fluid, i.e., its storage and loss modulus. Surely these properties depend on the shear rate... I would advise to move some of the other applications from the main text to extended figures to make up some room for a new and extended discussion on rheology and fluid dynamics. In particular the collapsed spheres appear to jam. See for example: Bourrianne, Philippe, et al. "Tuning the shear thickening of suspensions through surface roughness and physico-chemical interactions." Physical Review Research 4.3 (2022): 033062.

This is a very good point. Motivated by this comment, we characterized the rheology of the proposed metafluid using a parallel plate rheometer. The new results are reported in Fig. 4C of the revised main text (also included below as Fig. R6C for completeness). Further, the following paragraph has been added to the revised main text to describe them

> "The results from Fig. R6B suggest that the effective viscosity of the metafluids is higher when the capsules are collapsed. Nonetheless, it is important to note that in these experiments the metafluid is sheared at a rate $\dot{\gamma} \approx 1\ \mathrm{s}^{-1}$. To investigate how the effective viscosity of the metafluid is affected by the shear rate, we characterize its rheology using a parallel plate rheometer (see Supplementary Materials for details). The results reported in Fig. R6C indicate that the collapse of the particles have a profound effect on its rhelogy. In the presence of spherical capsules, the metafluid behaves as a Newtonian fluid with effective viscosity $\eta \approx 2.2\eta_0$ (η_0 denoting the viscosity of the solvent). However, the capsules collapse transforms it into a non-Newtonian shear-thinning fluid. In line with the results of Figs. R6A and B, the metafluid containing collapsed capsules exhibits high effective viscosity at low shear rates. Such behavior is ascribed to the formation of clusters by the collapsed particles, as previously reported for blood containing sickle-shaped red blood cells (9). As the shear rate increases, we observe an initial rapid decrease in effective viscosity, which we attribute to the gradual disruption of the clusters. Eventually, at $\dot{\gamma} \approx 10\ \mathrm{s}^{-1}$ the effective viscosity approaches a plateau at 1.3 η_0. This plateau can be attributed to the breakdown of particle clusters at such high strain rates. Finally, we note that at high shear rates the effective viscosity of the metafluid containing collapsed particles is lower than that of the metafluid containing spherical ones, likely because of the decrease in particle volume fraction caused by buckling. All together, these experiments highlight that the metafluid exhibit very rich rheology that can be tuned by controlling the level of the applied pressure. "

Furthermore, a detailed description of this new set of experiments has been added to Section S2.2 of Supporting Information

> **Rheology of the microsuspension.** To better understand the impact of capsule shape on the flow of metafluid, we characterize its rheology using a parallel plate rheometer (TA instruments Discovery HR 30) with two disks of 50 mm diameter separated by a gap of 500 μm. In all our tests, at room temperature (20 °C) we applied a ramp of increasing shear rate $\dot{\gamma}$ and swept it logarithmically from 1 s^{-1} to 100 s^{-1} with five points per decade and an averaging time of 30 s. We first performed the test with increasing shear rate and then repeated in decreasing order to check the reproducibility of the measurements. Note that the reported results are the average of the two tests, the variability measured is typically of the order of 3% of the average value at a given shear rate. Since using this setup we were unable to control external pressure applied to the metafluid, we prepares two suspensions:

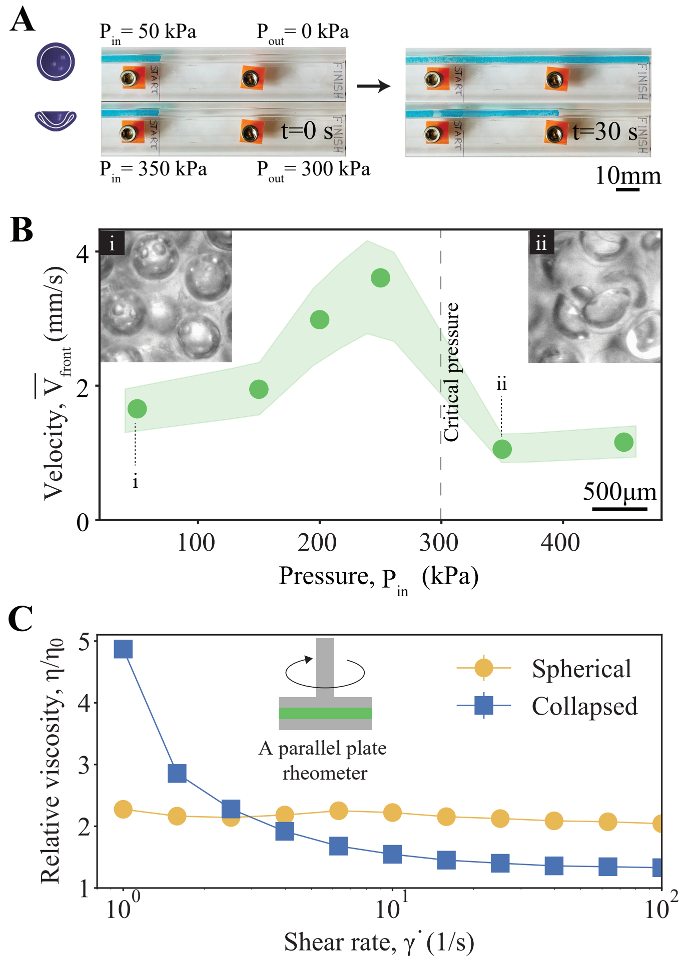

Fig. R6. Harnessing nonlinearity for tunable rheology. A Snapshots of the metafluid with $P_{cr}^{up} \approx 300$ flowing in a channel under constant difference of pressure $\Delta P = P_{in} - P_{out} = 50$ at $t = 0$ (left) and $t = 30$. Top: $P_{in} = 50$ so that the capsules are spherical. Bottom: $P_{in} = 350$ so that the capsules are collapsed. **B** Average front velocity as function of P_{in} for $\Delta P = 50$. The experimental data are depicted as average (markers) and standard deviation (shaded area). Snapshots show the flowing microsuspension for P_{in}= 50 and 350. **C** Flow curves for a metafluid containing spherical (yelow markers) and collapsed (blue markers) capsules.

- (i) a suspension with 30% volume fraction of spherical microcapsules (with $R_o = 40\ \mu m$ and $t = 8\mu m$) in silicone oil. Note that the miscrocapsules are fabricated following all steps shown in Fig. S3;
- (ii) a suspension with collapsed particles in silicone oil. To obtain the collapsed particles, we prepared the same volume of spherical capsules (with $R_o = 40\ \mu m$ and $t = 8\mu m$) as in (i) but this time we followed the process described in Fig. S3 up to Step 3. After that, we dried the inner core by evaporation (instead of sublimation), causing them to collapse permanently. The collapsed shells where then re-suspended in silicone oil to match the volume fraction of the first solution after collapse (upon pressurization).

In addition to these two suspensions, we also tested pure silicone oil (the solvent used for the two tested suspensions). The results are shown in Fig. R7A and indicate that the silicone oil behaves as a Newtonian fluid with viscosity $\eta_0 \approx 0.1$ Pa.s.

Finally, we performed oscillation sweeps with frequency $\omega \in [0.1, 10]$ Hz at low strain amplitudes (3% to remain in the linear-viscoelastic region) to probe the viscoelastic behavior of the metafluid with spherical and collapsed particles. As shown in Fig. R7B, we find that the suspension with spherical microcapsules is dominated by viscous effects which are linear with ω. Differently, the suspension with collapsed microcapsules behaves like a viscoelastic solid at low frequencies ($\omega < 1Hz$) where G' dominates. For $\omega > 1Hz$, the dominance of G'' indicates that viscous effects prevail in the metafluid.

Comment 2B

The authors demonstrate the generic nature of their idea, going from macroscale to microscale. This is certainly an asset of the work; yet little is provided in terms of a global understanding of the physics of the problem. I appreciate that the authors have FEM simulations reproducing most features of the system, but I would love to see more scaling laws and

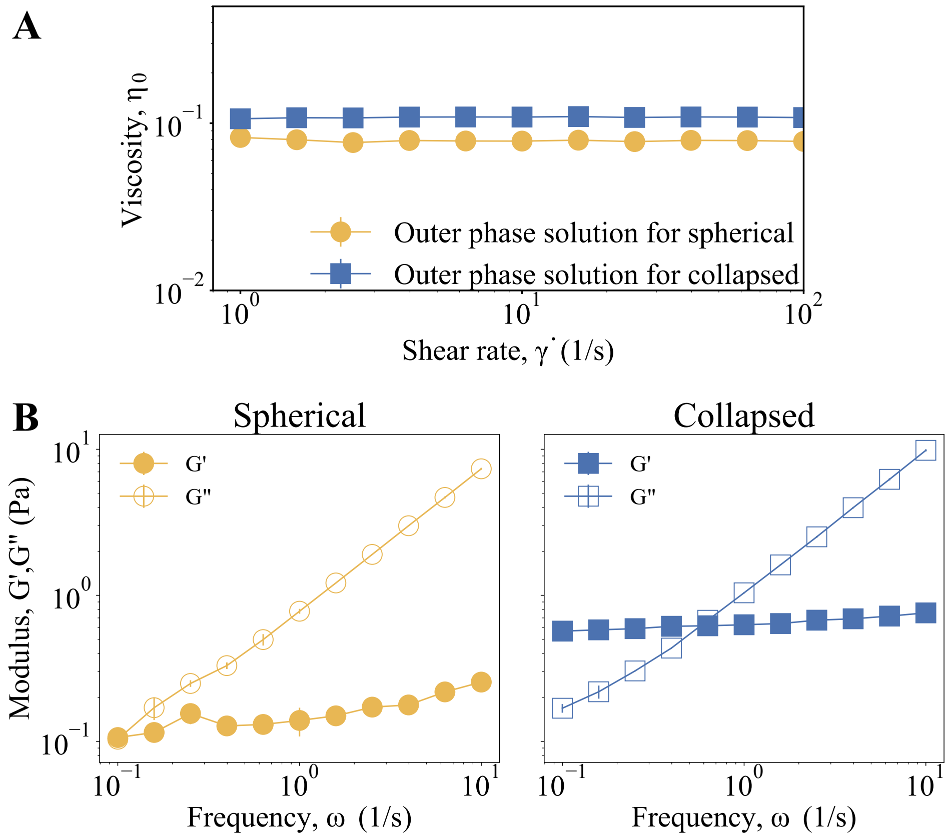

Fig. R7. Rheological measurements of the micro-suspensions. **(A)** Shear rate measurement of the outer phases used for the solutions comprising spherical and collapsed micro-capsules. **(B)** Oscillation sweep in frequency for micro-suspensions comprising spherical and collapsed micro-capsules.

theory, which will ultimately help providing design guidelines. Specifically, it would be helpful to know when the plateau in pressure occurs and how long it is. Likewise, the results somehow resemble well known pressure-volume nonlinear relation, such as the one found when inflating a hyper-elastic balloon. The similarities and differences with this system should be discussed.

This raises a valid point. In light of this comment, we've incorporated the following text and figure into Section S3.1 of the updated Supporting Information.

Scaling laws. We start by noting that analytical formulas are well established to describe the initial behavior of spherical shells subject to uniform external pressure (4, 10, 11). These formulas are derived using thin shell theory and therefore their accuracy is limited for capsules with $\eta \approx 0.3$ such as the microcapsules produced in this work. However, they can be still used to explain the general trends observed in the data.
Critical points: It is well known that for a thin spherical shell subjected to a pressure loading the difference between the external and internal pressure of the shells and volume at which buckling is triggered is given by (12)

$$\Delta P_{cr}^{up} = \frac{2E}{\sqrt{3(1-\nu^2)}}\left(\frac{t}{R}\right)^2. \qquad [R5]$$

For an incompressible material with $\nu = 0.5$, $E = 2G(1+\nu) = 3G$ and Eq. (R5) reduces to

$$\Delta P_{cr}^{up} = \frac{6G}{1.5}\eta^2 = 4G\eta^2, \qquad [R6]$$

where $\eta = t/R$. Further, the change in internal volume at which buckling is triggered can be expressed as (12)

$$\Delta V_{cr}^{up} = \eta V_0^{mid}, \qquad [R7]$$

where $V_0^{mid} = 4\pi R^3/3$ is the initial volume enclosed by the midsurface of the shell. Using Eq. (S3), V_0^{mid} can be rewritten as

$$V_0^{mid} = \frac{V_0}{(1-\eta/2)^3}, \qquad [R8]$$

where $V_0 = 4\pi R_i^3/3$ is initial volume of the gas-filled cavity (R_i denoting the inner radius of the shell). Taking a Taylor series, Eq. (R8) can be approximated with second order accuracy as

$$V_0^{mid} \approx V_0 \left(1 + \frac{3\eta}{2}\right). \qquad \text{[R9]}$$

Finally, substitution of Eq. (R9) into Eq. (R7) yields

$$\Delta V_{cr}^{up} \approx \eta \left(1 + \frac{3\eta}{2}\right) V_0, \qquad \text{[R10]}$$

As shown in Figs. R8A and B, Eqs. (R6) and (R10) closely match the numerically obtained values for ΔP_{cr}^{up} and ΔV_{cr}^{up} for small η. For $\eta > 0.15$, the accuracy of the equation becomes limited since the shells can no longer be assumed to be thin, but qualitatively the trends line up well. Next, we use these formulas to predict the total

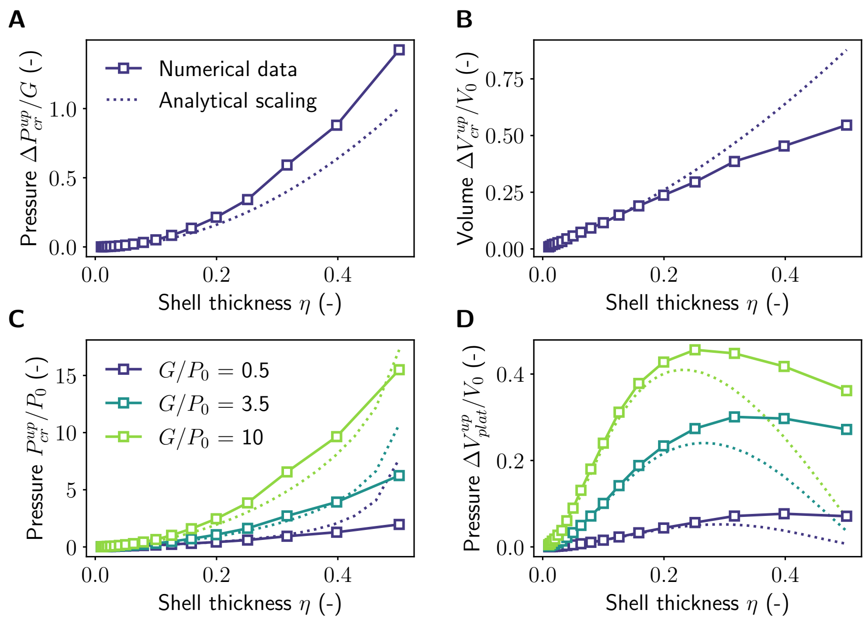

Fig. R8. Scaling of critical pressures and volumes with shell thickness and modulus. A, Evolution of the differential shell pressure at the critical point on loading ΔP_{cr}^{up} in function of the dimensionless shell thickness η. Square markers indicate data points obtained with the FE model while the dotted line corresponds to Eq. (R5). **B**, Evolution of the volume at the critical point upon loading, ΔV_{cr}^{up}, as function of η. The dotted line corresponds to Eq. (R9). **C**, Evolution of the total pressure at the critical point upon loading, P_{cr}^{up}, as a function of η for different values of the dimensionless shear modulus G/P_0. The square markers correspond to numerical FE results and the dotted lines to Eq. (R13). **D**, Evolution of the change in volume during the instability on pressure-controlled loading, ΔV_{plat}^{up}, as function of η for different values of G/P_0. The square markers correspond to FE resutls and the dotted lines correspond to results from Eq. (R16). The line colors correspond to the same values of G/P_0 as indicated in the legend of panel C.

external pressure at the critical point, P_{cr}^{up}

$$P_{cr}^{up} = \Delta P_{cr}^{up} + P_{int,cr}, \qquad \text{[R11]}$$

where $P_{int,cr}$ denoted the critical internal pressure caused by the compression of the gas inside the capsule. Using Eq. (S6), $P_{int,cr}$ can be expressed as

$$P_{int,cr} = P_0 \left(\frac{1}{1 - \frac{\Delta V_{cr}^{up}}{V0}} - 1 \right) = P_0 \left(\frac{1}{1 - \eta(1 + 3\eta/2)} - 1 \right), \qquad \text{[R12]}$$

where P_0 is the atmospheric pressure. Substitution of Eqs. (R6) and (R12) into Eq. (R11) yields

$$\frac{P_{cr}^{up}}{P_0} = 4\frac{G}{P_0}\eta^2 + \frac{1}{1 - \eta(1 + 3\eta/2)} - 1. \qquad \text{[R13]}$$

In Fig. R8C we compare the predictions of this equation to numerically obtained data and find that it accurately predicts the trend, especially for low values of η.
Change in volume upon collapse: Next, we derive a scaling law for the change in volume of the capsule as it suddenly collapses at P_{cr}^{up} when under pressure controlled conditions, ΔV_{plat}^{up}. Note that ΔV_{plat}^{up} is proportional

to the width of the plateau in the pressure-volume characteristic of a metafluid consisting out of many identical capsules under both pressure and volume control conditions (see Section S3.2). We start by noting that

$$\Delta V^{up}_{plat} = \Delta V^{up}_{cr,col} - \Delta V^{up}_{cr}, \tag{R14}$$

where $\Delta V^{up}_{cr,col}$ is the volume of the collapsed capsule after the instability and ΔV^{up}_{cr} is given by Eq. (R10). After collapse the capsule is highly deformed and its behavior is no longer captured accurately by analytical models. However, our FE simulations show that in the collapsed state $\Delta P_{shell}(\Delta V^{up}_{cr,col}) \ll P_{int}(\Delta V^{up}_{cr,col})$, so that the influence of the shell stiffness can be neglected. As such, $\Delta V^{up}_{cr,col}$ can be estimated from Eq. (S6) as:

$$\frac{\Delta V^{up}_{cr,col}}{V_0} = 1 - \frac{1}{\frac{P^{up}_{cr}}{P_0} + 1} \tag{R15}$$

Substitution of Eq. (R13) into Eq. (R15) yields

$$\frac{\Delta V^{up}_{plat}}{V_0} = 1 - \frac{1}{4\frac{G}{P_0}\eta^2 + \frac{1}{1-\eta(1+3\eta/2)}} - \eta\left(1 + \frac{3}{2}\eta\right). \tag{R16}$$

Eq. (R16) predicts that $\Delta V^{up}_{plat}/V_0$ reaches a maximum for intermediate values of η. This trend is confirmed by the numerical data shown in Fig. R8D. However, Eq. (R16) starts to largely underestimate the numerical data for η past this maximum largely due to the overestimation of ΔV^{up}_{cr}.

Comment 2C

While the figures are superb, the manuscript needs significant re-writing to be adapted to a broad readership. Too many details of the method (experimental and numerical) crowd the results that the authors are reporting. More emphasis should be brought to the said results. Likewise, the manuscript appears to be missing a full-fledged introduction. I appreciate that the abstract serves this purpose to some extent, yet, it would certainly serve the manuscript to detail further relevant aspects of the context of this study.

In response to this comment we have moved the description of the logic gate from the main text to Supporting Information, since it required a lot of details. Further, we have added an introductory paragraph that reads as

> "Unlike solid metamaterials, metafluids have the unique ability to flow and adapt to the shape of their container without the need for a precise arrangement of their constituent elements. Our goal is to realize a metafluid that not only possesses these remarkable attributes but also provide a platform for programmable elastic behavior, optical properties, and rheology. To achieve this, we focus on a suspension comprising elastomeric, highly deformable spherical capsules filled with air within an incompressible fluid. "

Comment 2D

The fabrication technique of the large capsules is like that reported in E. JambonPuillet et al. Nature Phys 2020, which should thus be cited

The paper has been cited.

Comment 2E

What explains the qualitative difference between the curves in Fig1C and E?

We interpret the Reviewer's comment as referring to the differences between Figs. 1C and 1F. The absence of a clearly defined plateau in the pressure-volume curve of the metafluid with microscale shells can primarily be attributed to geometric imperfections introduced during the fabrication process. These imperfections give rise to polydispersity and ultimately lead in a broad spectrum of buckling pressures. To emphasize this important aspect, we have incorporated the following text to page 2 of the revised main text:

> "Nevertheless, it is important to highlight that the metafluid incorporating microscale capsules does not exhibit a clear plateau in the pressure-volume curve. This deviation can be attributed to geometric imperfections introduced during the fabrication process, resulting in polydispersity and a wide range of buckling pressures."

Comment 2F

What if the pressure is not uniform across the sample? As one would expect in an flow. How would the metafluid then behave? Should this point be a concern for the programmability of the system?

The pressure-volume characteristic of a metafluid subjected to non-uniform pressure can be investigated using the same analysis presented in the paper. On the one hand, the fact that the total change in volume of the fluid is equal to the sum of the changes in volume of the individual capsule remains valid, so Eq. (2) of the main text is unaltered:

$$\Delta V = \sum_{i=1}^{N} \Delta V^{(i)}, \tag{R17}$$

where $\Delta V^{(i)}$ is the change in volume of the i-th capsule. On the other hand, if we denote as P_{ref} the pressure measured in the fluid at some reference location, we can say that the pressure at any other location $\mathbf{x}$ is equal to $P_{ref} + P_{offset}(\mathbf{x})$ where P_{offset} represents the non-uniform component of the pressure field. It follows that the pressure that acts on the outer surface of the i-th capsule located at $\mathbf{x}^{(i)}$ can be expressed as

$$P_{ext}^{(i)}\left(\Delta V^{(i)}\right) = P_{ref} + P_{offset}(\mathbf{x}^{(i)}). \tag{R18}$$

Expressing Eq. (R18) for all capsules within the metafluid and eliminating P_{ref} results in:

$$P_{ext}^{(1)}\left(\Delta V^{(1)}\right) - P_{offset}(\mathbf{x}^{(1)}) = P_{ext}^{(2)}\left(\Delta V^{(2)}\right) - P_{offset}(\mathbf{x}^{(2)}) = \ldots = P_{ext}^{(N)}\left(\Delta V^{(N)}\right) - P_{offset}(\mathbf{x}^{(N)}), \tag{R19}$$

which is equivalent to Eq. (1) in the main paper, if we define $P_{ext}^{(i)'}\left(\Delta V^{(i)}\right) = P_{ext}^{(i)}\left(\Delta V^{(i)}\right) - P_{offset}(\mathbf{x}^{(i)})$.

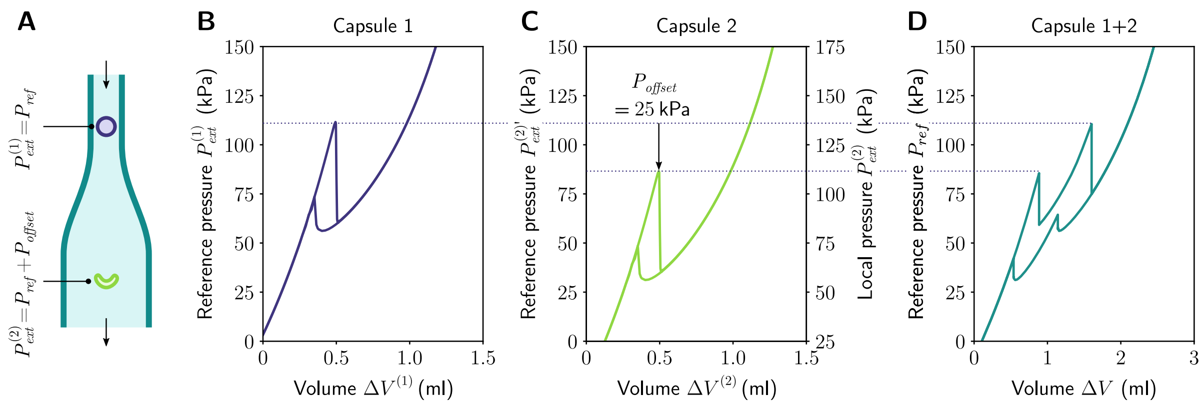

Fig. R9. Modeling metafluids with a non-uniform pressure distribution. **A**, Schematics of the system. **B**, $P_{ext}^{(1)} - \Delta V^{(1)}$ relation for capsule 1. **C**, $P_{ext}^{(2)'} - \Delta V^{(2)}$ relation for capsule 2. This is the same relation as between the local pressure $P_{ext}^{(2)}$ and ΔV shifted down with P_{offset}, which is 25 kPa in this example. **D**, Relation between the reference pressure P_{ref} and the total change in volume of the metafluid.

This indicates that metafluids that experience non-uniform pressure will have the same fundamental behavior as metafluids that experience uniform pressure. The only difference is that the pressure-volume curves of the individual capsules is transformed, such that for example capsules that experience a higher local pressure because of a positive $P_{offset}(\mathbf{x})$ will buckle at a lower value of P_{ref} than in the uniform pressure case. As an illustration, consider a metafluid with two identical capsules subject to flow in a channel with a non-uniform cross-section (Fig. R9A). Capsule 1 experiences an external pressure of $P_{ext}^{(1)} = P_{ref}$ while capsule 2 experiences a higher pressure $P_{ext}^{(2)} = P_{ref} + P_{offset}$ due to the decrease in flow speed. It follows from Eqs. (R17) and (R19) that the total P_{ref}-ΔV characteristic of such metafluid can be obtained by identifying the volumes $\Delta V^{(1)}$ and $\Delta V^{(2)}$ for which $P_{ext}^{(1)} = P_{ref} = P$ and $P_{ext}^{(2)'} = P_{ref} - P_{offset} = P$ (P denoting a given pressure - Figs. R9B and C) and adding up the resulting volumes (i.e. $\Delta V = \Delta V^{(1)} + \Delta V^{(2)}$ - Fig. R9D).

Comment 2G

In Fig3A, why not putting the dotted line plateau in between P_{hold} and P_{crush}?

We have made the requested change in Figure 3A by placing the dotted line plateau between P_{hold} and P_{crush}.

Response to Referee #3

Overall, although the concept of suspensions made from collapsing particles that lead to a highly nonlinear pressure-volume curve with enhance compressibility is not new (e.g. ref 27), many ideas put forward in this manuscript are novel and stimulating: the idea of using simple spherical shells, their manufacturing at the sub-millimeter scale and their use in applications such as gripping, logic, optics and rheology are all innovative and could lead to better integrated adaptive mechanical response. Embedding functionality in the fluid is complementary to the approach traditionally employed in mechanical metamaterials, where the functionality is integrated in elastic structures instead and could help applications in robotics and medicine applications.

We thank the reviewer for their thoughtful comments. We appreciate their recognition of the innovative aspects of our research.

Comment 3A

Could the author explain why the pressure-volume curve in the case of the sub-mm shells does not exhibit the clean plateau of the cm-shell? Is it due to polydispersity?

We kindly direct the Reviewer to our response to comment 2E.

Comment 3B

For the logic gate (fig 3C). What is the benefit of using such fluid instead of using valves? In this case it is not clear to me that the metafluid offers a better level of integration, since many mechanical apparatus are used in the setup.

First, we want to highlight that, in response to comment 2C by Reviewer 2, we have moved the logic gate section from the main text to the Supporting Information.

Regarding the advantages of utilizing metafluids for logic gates, we believe the primary benefit lies in the ease with which the fluid can be replaced, allowing for the reprogramming of circuit functionality. To emphasize this point we have added the following text to Section S2.1 of revised Supporting Information

> "These findings highlight a key benefit of employing metafluids in logic gates: the ease with which the fluid can be replaced, allowing for the reprogramming of circuit functionality. "

Additionally, metafluids offer the unique capability to shape the pressure-volume characteristics, such as creating a slanted plateau rather than a constant pressure plateau. This flexibility enables the creation of mappings between continuous variables, in contrast to the typical binary-to-binary transformations seen in conventional logic gates. This tunable continuous mapping between variables has the potential to facilitate the implementation of a fuzzification scheme, a crucial element in fuzzy logic systems. While the realization of a full fuzzy logic system within a fluidic network requires further research and development (a feat never achieved before), metafluids can play a pivotal role as an integral component in making this vision a reality.

Comment 3C

On the same figure 3c. I find that the setup and experimental protocol are not clearly illustrated and explained.

We have made modifications to the schematics that illustrate the structure of the logic gates (Figure S9 in the revised Supporting Information). We hope that these changes enhance clarity.

Comment 3D

On fig 3B. How would you model such metafluid at the continuum level? is the shape of the bulge different between the metafluid and ordinary incompressible fluids?

Firstly, it's essential to note that in the experimental results reported in Figure 3B of the main text, the occurrence of the bulge differs in terms of the volume of supplied fluid, denoted as ΔV, when using an incompressible fluid compared to the metafluid. However, it remains unaltered in terms of pressure and the change in volume for the flexible structure, denoted as ΔV_{flex}, regardless of the type of fluid used for inflation.

In Figure 3B of the main text, we present the pressure-volume curve resulting from inflating a latex tube with both an incompressible fluid (red line) and the metafluid (blue line). When a flexible structure is inflated with an incompressible fluid,

$$\Delta V(P) = \Delta V_{flex}(P). \tag{R20}$$

Differently, when the metafluid is used to inflate the flexible structure

$$\Delta V(P) = \Delta V_{flex}(P) + \Delta V_{caps}(P), \tag{R21}$$

where $\Delta V_{caps}(P)$ denotes the change of volume of the capsules suspended in the metafluid. It follows from Eqs. (R20) and (R21) that each point on the blue curve can be obtained by shifting the point on the red curve with the same pressure by $\Delta V_{caps}(P)$ along the horizontal axis. Due to the metafluid's pressure-volume characteristic, which features a pressure plateau in the vicinity of the pressure peak, all points beyond the peak of the red curve experience a considerably greater shift to the right compared to the points located before the peak.

Furthermore, we anticipate that the configuration of the bulge will remain unaffected by the choice of fluid for inflating the structure. This expectation is grounded in the fact that the overall energy of the system can be expressed as:

$$U_{tot} = U_{flex}(r) + U_{mf}(\Delta V_{caps}) - P\Delta V_{flex} \tag{R22}$$

where U_{flex} is the internal elastic energy of the flexible structure, which depends on the deformed shape of the structure walls described by the function r (13). Moreover, U_{mf} denotes the internal energy of the metafluid which, being a fluid medium, depends on

$$\Delta V_{caps} = \Delta V_{flex}(r) - \Delta V. \tag{R23}$$

Since for a fixed ΔV_{flex}, ΔV_{caps} does not depend on r, it can be considered as a constant. It follows that, when minimizing U_{tot} with respect to r for a given ΔV_{flex}, the same result is obtained regardless of the specific characteristics of U_{mf}. This underscores that the shape of the bulge remains unchanged, irrespective of the choice of inflating fluid. It is important to note that this conclusion holds true until the point at which the particles begin to jam within the metafluid, as beyond this point, the metafluid can no longer be described as a pure fluid.

Comment 3E

On fig 4DE: The rheology should be better described using theory or computation (e.g. Lattice Boltzmann could be a suitable technique here). Can the shear flow also deform the shell and hence affect their buckling? Do the particles migrate in the Poiseuille flow of fig 4d?

In terms of the flow aspect, there was no observed particle migration throughout the course of our experiments. Furthermore, as detailed in our response to Comment 2A, we carried out a fresh series of experiments aimed at providing a more comprehensive characterization of the rheological properties of the proposed metafluid.

In addition, we performed supplementary Finite Element simulations to explore the impact of shear flow on the buckling behavior of the capsules. To elaborate on these simulations, we have incorporated the following text into Section S3.1 of the revised Supporting Information

Influence of shear flow. When a metafluid flows, every point in the fluid experiences a shear stress τ_{ext} proportional to the viscosity of the fluid, η_0, and the spatial velocity gradient. If this shear load is sufficiently large, the relation between the hydrostatic pressure applied to a capsule and the resulting volume is modified. The reason is that the shear load deforms the sphere approximately into an oblate ellipsoid with the major axis rotated at 45 to the flow lines and that such a shape has a lower buckling pressure than a sphere (14). The change in shape of the capsule due to a simple shear stress τ_{ext} can be estimated analytically for a thin spherical shell made of an incompressible material and surrounded by an incompressible fluid (15). According to this model, the Taylor parameter D is given by

$$D = \frac{L-B}{L+B} = \frac{25}{12}\frac{\tau_{ext}}{G\eta_0}, \tag{R24}$$

where L and B are the major and minor radii of the ellipsoid, respectively. Since it has been shown that the critical buckling pressure ΔP_{cr}^{up} of oblate ellipsoids scales with the square of their aspect ratio B/L (16), it follows that

$$\frac{\Delta P_{cr}^{up}}{G} \propto \left(\frac{1 - \frac{25}{12}\frac{\tau_{ext}}{G\eta_0}}{1 + \frac{25}{12}\frac{\tau_{ext}}{G\eta_0}} \right)^2 . \tag{R25}$$

To validate the predictions of Eq. (R25), we perform a series of FE simulations in which we apply to the outer surface of the models a distributed load consistent with a uniform shear stress in the surrounding medium τ_{ext} prior to loading the capsule hydrostatically (Fig. R10A). Note that the models used for these simulations comprise three dimensional elements rather than axisymmetric ones. In Fig. Fig. R10B we report results for a spherical capsule with $\eta = 0.22$. These results indicate that Eq. (R25) accurately captures the drop in ΔP_{cr}^{up} as τ_{ext}/G is increased (Fig. R10C). They also show that the critical volumes on loading ΔV_{cr}^{up} follows a similar trend, while ΔV_{cr}^{down} is less sensitive to the shear load (Fig. R10D). For $\eta = 0.3$ and $G = 400$, which are typical values for the produced microcapsules, the total critical pressures P_{cr}^{up} and P_{cr}^{down} change by less than 2 if τ_{ext} remains below 620. Below this limit, these capsules can be considered to be unaffected by the flow field such that all modeling techniques presented in this paper for individual capsules and metafluids yield accurate results even though they assume hydrostatic loading.

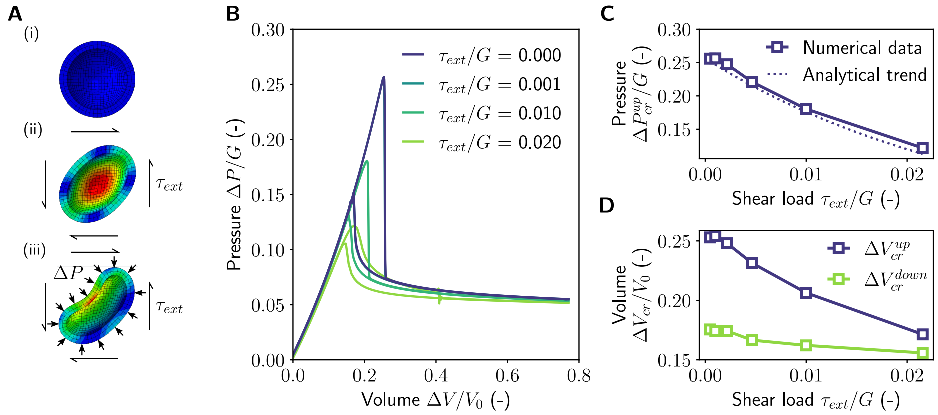

Fig. R10. Influence of a shear load on the pressure-volume characteristic. A, Illustration of the simulation procedure: (i) initial spherical shape of the capsule, (ii) a shear load τ_{ext} is applied on the outer surface, (iii) a uniform normal pressure ΔP is applied on top of τ_{ext}. **B**, Simulated pressure-volume characteristics for a shell with $\eta = 0.22$ subjected to a constant external shear load τ_{ext}. **C**, Relation between the normalized shear load τ_{ext}/G and the critical shell pressure $\Delta P_{cr}^{up}/G$ for a spherical capsule with $\eta = 0.22$. The dashed line represents the analytical scaling law provided by Eq. (R25). **D**, Relation between the normalized shear load τ_{ext}/G and the change in volume at which the instabilities under volume-controlled conditions are triggered upon loading (ΔV_{cr}^{up}) and unloading (ΔV_{cr}^{down}).

References

1. Bert Van Raemdonck, Edoardo Milana, Michael De Volder, Dominiek Reynaerts, and Benjamin Gorissen. Nonlinear inflatale actuators for distributed control in soft robots. Advanced Materials, page 2301487, 2023.
2. C. Quilliet, C. Zoldesi, C. Riera, A. van Blaaderen, and A. Imhof. Anisotropic colloids through non-trivial buckling. The European Physical Journal E, 27:13–20, 2008. ISSN 1292-895X. . URL https://doi.org/10.1140/epje/i2007-10365-2.
3. Gwennou Coupier, Adel Djellouli, and Catherine Quilliet. Let's deflat that beach ball. The European Physical Journal E, 42(9):129, Sep 2019. ISSN 1292-895X. . URL https://doi.org/10.1140/epje/i2019-11900-2.
4. John W Hutchinson. Buckling of spherical shells revisited. Proceedings of the Royal Society A: Mathematical, Physical and Engineering Sciences, 472(2195):20160577, 2016.
5. Ian D Johnston, Daniel K McCluskey, Christabel KL Tan, and Monica C Tracey. Mechanical characterization of bulk sylgard 184 for microfluidic and microengineering. Journal of Micromechanics and Microengineering, 24(3):035017, 2014.
6.Chung-Lin Wu, Hsueh-Chu Lin, Jiong-Shiun Hsu, Ming-Chuen Yip, and Weileun Fang. Static and dynamic mechanical properties of polydimethylsiloxane/carbon nanotube nanocomposites. ThinSolid Films, 517(17):4895–4901, 2009. ISSN 0040-6090. . URL https://www.sciencedirect.com/science/article/pii/S0040609009006154. 4th International Conference on Technological Advances oThin Films and Surface Coatings.
7. Lucia Stein-Montalvo, Douglas P. Holmes, and Gwennou Coupier. Delayed buckling of spherical shells due to viscoelastic knockdown of the critical load. Proceedings of the Royal Society A: Mathematical, Physical and Engineering Sciences, 477(2253):20210253, 2021. . URL https://royalsocietypublishing.org/doi/abs/10.1098/rspa.2021.0253.
8. Adel Djellouli, Philippe Marmottant, Henda Djeridi, Catherine Quilliet, and Gwennou Coupier. Buckling instability causes inertial thrust for spherical swimmers at all scales. Phys. Rev. Lett., 119: 224501, Nov 2017. . URL https://link.aps.org/doi/10.1103/PhysRevLett.119.224501.
9. S Chien, S Usami, and J F Bertles. Abnormal rheology of oxygenated blood in sickle cell anemia. J Clin Invest, 49(4):623–634, April 1970.
10. JMT Thompson. The rotationally-symmetric branching behaviour of a complete spherical shell. Proceedings of the Koninklijke Nederlandse Akademie van Wetenschappen, pages 295–311, 1964.11. Sebastian Knoche and Jan Kierfeld. Buckling of spherical capsules. Phys. Rev. E, 84:046608, Oct 2011. . URL https://link.aps.org/doi/10.1103/PhysRevE.84.046608.
12. John W. Hutchinson and J. Michael T. Thompson. Nonlinear Buckling Interaction for Spherical Shells Subject to Pressure and Probing Forces. Journal of Applied Mechanics, 84(6), 04 2017. ISSN 0021-8936. . URL https://doi.org/10.1115/1.4036355. 061001.
13. YB Fu and YX Xie. Stability of localized bulging in inflate membrane tubes under volume control. International Journal of Engineering Science, 48(11):1242–1252, 2010.
14. Dominique Barthes-Biesel. Motion and deformation of elastic capsules and vesicles in flw. Annual Review of flui mechanics, 48:25–52, 2016.
15. D Barthès-Biesel, JM Rallison, D Barthès-Biesel, and JM Rallison. The time-dependent deformation of a capsule freely suspended in a linear shear flw. Journal of Fluid Mechanics, 113:251–267, 1981.
16. K.M. Mushtari and K.Z. Galimov. Non-linear Theory of Thin Elastic Shells, chapter 13, pages 310–311. NASA TT F-. National Science Foundation, 1957. ISBN 9780608310121. URL https://archive.org/details/nasa_techdoc_19980231031/mode/2up.

Reviewer Reports on the First Revision:

Referees' comments:

Referee #1 (Remarks to the Author):

The authors addressed my previous criticism with attention by providing additional experimental and numerical results in the revised manuscript.

In particular, I like the convincing experiments showing that the “behaviour of the metafluid is mostly invariant” after being submitted to a hundred or so loading cycles.

I also appreciate that the authors provided more details about material parameters (the units are missing in the Rebuttal Letter, see Table R1, but are well written in the revised Supplementary Materials, see Table S1). However, I wouldn’t write that “K tending towards infinity” that is not correct even if elastomers are generally considered to be incompressible. If it’s true that, the bulk modulus K (~ 1GPa) of an elastomer is much larger than its shear modulus G (~ 1MPa), resulting in a Poisson’s ratio “approaching 0.5”, its bulk modulus K does not tend to infinity. The authors should rather give the values of this parameter K in the revised manuscript for the two materials studied here (PVS and PDMS).

Finally, I have to say that I’m not convinced by the arguments put forward by the authors to explain the improvement in optical properties with the randomly-oriented collapsed capsules.

The two mechanisms proposed by the authors (“lensing effect” and “reduction in the coverage area of the capsules in the collapse state”) seem to me to be used opportunistically. To analyse their experimental observations, the authors should conduct a more in-depth study of light scattering by these anisotropic particles in my opinion.

In conclusion, I still think that Djellouli et al. have reported a very interesting study about “Shell buckling for programmable metafluids” but I still doubt the relevance of publishing this work in a demanding journal like Nature.

Referee #2 (Remarks to the Author):

The authors have addressed my concerns thoroughly. The work is stimulating and could be impactful in soft matter. I see no reason to delay publication further.

Referee #3 (Remarks to the Author):

The authors have successfully addressed the comments of the reviewers. In particular, they have added a lot more information about the rheology of the metafluid, which is now well described and displays a surprising tunable shear-thinning behavior. I expect those metafluids to give rise to a lot more surprises in their mechanical properties beyond the hydrostatic load case and I hence expect the article to become a seminal contribution. I therefore recommend publication of the paper in its current form.

Author Rebuttals to First Revision:

Response to the Referees

Shell Buckling for Programmable Metafluids

Manuscript number: 2023-05-09013B

Adel Djellouli[a,1], Bert Van Raemdonck[b,1], Yang Wang[a,1], Yi Yang[a], Anthony Caillaud[a], David Weitz[a], Shmuel Rubinstein[c], Benjamin Gorissen[b,2], and Katia Bertoldi[a,2]

[a]J.A. Paulson School of Engineering and Applied Sciences, Harvard University, USA; [b]Department of Mechanical Engineering, KU Leuven and Flanders Make, Belgium; [c]The Racah Institute of Physics, The Hebrew University, Israel; [1]These authors contributed equally.; [2]These authors contributed equally.

Response to Referee #1

The authors addressed my previous criticism with attention by providing additional experimental and numerical results in the revised manuscript. In particular, I like the convincing experiments showing that the "behaviour of the metafluid is mostly invariant" after being submitted to a hundred or so loading cycles. I also appreciate that the authors provided more details about material parameters (the units are missing in the Rebuttal Letter, see Table R1, but are well written in the revised Supplementary Materials, see Table S1). In conclusion, I still think that Djellouli et al. have reported a very interesting study about "Shell buckling for programmable metafluids" but I still doubt the relevance of publishing this work in a demanding journal like Nature.

We appreciate the careful review of our manuscript and thank the Referee for acknowledging our efforts to address their previous comments. We are pleased to hear that the additional experimental and numerical results, particularly those demonstrating the consistent behavior of the metafluid after numerous loading cycles, have contributed positively to the manuscript.

Comment 1A

However, I wouldn't write that "K tending towards infinity" that is not correct even if elastomers are generally considered to be incompressible. If it's true that, the bulk modulus K (≈1GPa) of an elastomer is much larger than its shear modulus G (≈1MPa), resulting in a Poisson's ratio "approaching 0.5", its bulk modulus K does not tend to infinity. The authors should rather give the values of this parameter K in the revised manuscript for the two materials studied here (PVS and PDMS).

In response to this comment, we have modified the text in Supplementary Information to read as

> **Effect of bulk modulus.** It is well known that elastomers are nearly incompressible. Consequently, they are characterized by ν approaching 0.5 and high values of K/G (where ν, K and G represent the initial Poisson's ratio, bulk modulus colorred and bulk modulus, respectively). The classical buckling pressure of elastic thin spherical shells is given by (1)
>
> $$\Delta P_{cr}^{up} = \frac{2E}{\sqrt{3(1-\nu^2)}}\eta^2. \tag{R1}$$
>
> By introducing
>
> $$E = \frac{9KG}{3K+G} \text{ and } \nu = \frac{3K-2G}{2(3K+G)} \tag{R2}$$
>
> Eq. (R1) can be rewritten in terms of K/G as
>
> $$\frac{\Delta P_{cr}^{up}}{G} = 4\frac{\frac{K}{G}}{\sqrt{\left(\frac{K}{G}\right)^2+\frac{4}{3}\frac{K}{G}}}\eta^2. \tag{R3}$$
>
> As such, we anticipate that as K becomes larger, the value of ΔP_{cr}^{up} will increase asymptotically. To verify this point, we perform a number of FE simulations on a shell with $\eta = 0.22$ where we systematically vary the normalized bulk modulus K/G from e1 to e9 (note that $K/G \approx$ 5e3 for PDMS mixed in a base polymer to curing agent ratio of 10:1 (2) and $K/G \approx$ 3e3 when the ratio is 5:1 as in our micrometer scale capsules (3)). The results of S21 show that for $K/G >$ 1e3 the change in ΔP_{cr}^{up} is less than 0.1 as the K is varied from e3 to e9. Further, in S21B we compare the evolution of ΔP_{cr}^{up} as a function of K as predicted by Eq. (R3) with the data extracted from our FE simulations. Again, we find that the dependence of ΔP_{cr}^{up} on K is weak. Finally, in Fig. S21C we show the evolution of the critical volumes on volume controlled loading and unloading as a function of K and again find a weak dependence. Since approximating the material as incompressible introduces a negligible error (0.7 ‰ assuming $K/G =$ 1e3 and using Eq. (R3)), we use a perfectly incompressible material model in the simulations.

Comment 1B

Finally, I have to say that I'm not convinced by the arguments put forward by the authors to explain the improvement in optical properties with the randomly-oriented collapsed capsules. The two mechanisms proposed by the authors ("lensing effect" and "reduction in the coverage area of the capsules in the collapse state") seem to me to be used opportunistically. To analyse their experimental observations, the authors should conduct a more in-depth study of light scattering by these anisotropic particles in my opinion.

We value the input from the referees. Encouraged by their comments, we carried out additional simulations to systematically assess both transmittance and focusing efficiency across various incident angles. The findings from these additional simulations have been incorporated into the Supplementary Information and are reported here for completeness:

> "Further, we systematically quantified both the transmittance and focusing efficiency for incident angles $\theta \in [0, 180]$ (Fig. R1A). To measure the transmittance, we placed a power sensor at the focal length of the capsule, f (estimated from measurements of the spot diagram) for angles $\theta <= 90$ and at a distance $-f$ for $\theta > 90$ (to make sure to measure transmitted rays). The transmittance T was then obtained by integrating the power density recorded on the power sensor and dividing it by the power of the incident ray (fixed at 1 W),
>
> $$T = \frac{Power_{transmitted}}{Power_{incident}}. \quad \text{[R4]}$$
>
> To obtain the focusing efficiency, we integrated the power density recorded on the portion of the power sensor where it is above the half maximum, $Power_{FWHM}$, and divided it by the transmitted power **(? ?**)
>
> $$F = \frac{Power_{FWHM}}{Power_{transmitted}}. \quad \text{[R5]}$$
>
> As shown in Fig. R1B, we find that the average transmittance over all angles is ≈70%, with a maximum of 99% for $\theta = 180$(concave side facing the incident rays). We also find that the average focusing efficiency is above 40% for a wide range of angles, with a maximum of ≈60% for $\theta = 20$. "

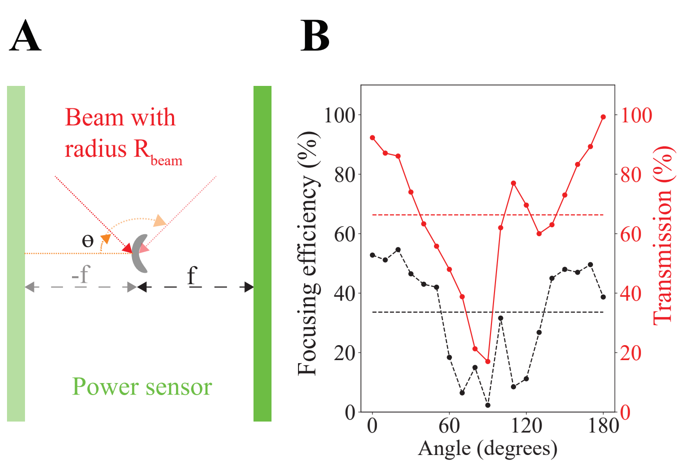

Fig. R1. Effect of incident angle. A Schematics of the simulations. **B** Transmittance and focusing efficiency as function of the incident angle θ. Dashed lines denote the average values across all angles.

Further, we have modified Fig. S28 of Supplementary Information (reported here as Fig. R2) for completeness) to highlight the decrease in coverage area for the array of randomly oriented collapsed shells. The snapshots clearly indicate that the light rays can penetrate more deeeply the array of randomly oriented collapsed capsules without being deviated by the shells.

Response to Referee #2

The authors have addressed my concerns thoroughly. The work is stimulating and could be impactful in soft matter. I see no reason to delay publication further.

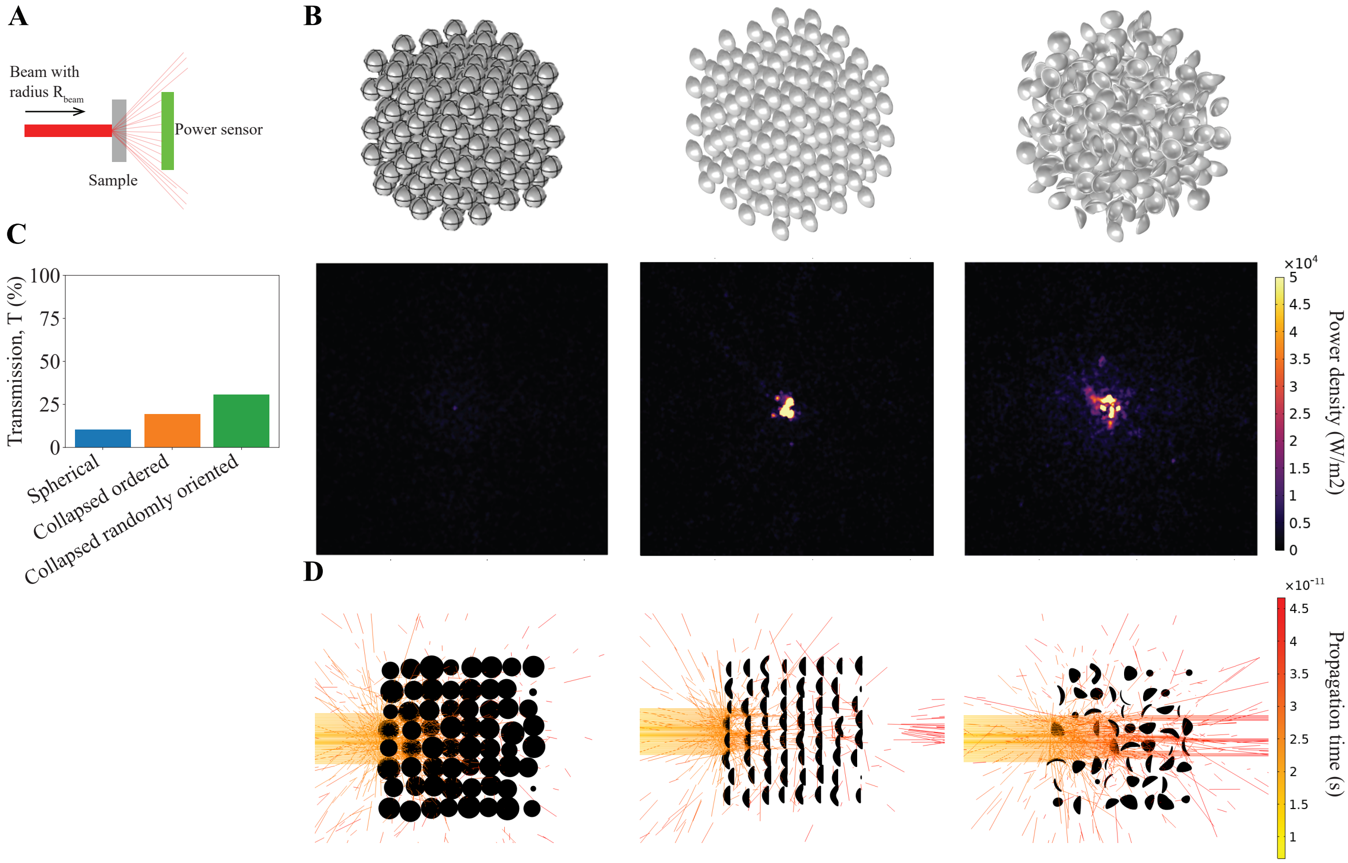

Fig. R2. Ray tracing simulations for a cluster of capsules. A Schematics of the simulation setup. **B** Heat map plots of the transmitted power density in the three different scenarios considered: cluster of spheres (left), cluster of collapsed capsules with their convex side facing the incident beam (middle) and cluster of randomly oriented collapsed capsules (right). **C** Transmitted power measured for the three different scenarios divided by the input power. **D** Numerical snapshots showing the propagation of the light rays across the three considered arrays.

We appreciate the positive feedback. It is rewarding to know that the Reviewer finds our work both stimulating and potentially impactful in the field of soft matter.

Response to Referee #3

The authors have successfully addressed the comments of the reviewers. In particular, they have added a lot more information about the rheology of the metafluid, which is now well described and displays a surprising tunable shear-thinning behavior. I expect those metafluids to give rise to a lot more surprises in their mechanical properties beyond the hydrostatic load case and I hence expect the article to become a seminal contribution. I therefore recommend publication of the paper in its current form.

We sincerely appreciate the positive feedback and recognition of our efforts in addressing the reviewers' comments. We share the Reviewer enthusiasm for the potential surprises in the mechanical properties of these metafluids beyond the hydrostatic load case.

References

1. John W Hutchinson. Buckling of spherical shells revisited. *Proceedings of the Royal Society A: Mathematical, Physical and Engineering Sciences*, 472(2195):20160577, 2016.
2. Ian D Johnston, Daniel K McCluskey, Christabel KL Tan, and Monica C Tracey. Mechanical characterization of bulk sylgard 184 for microfluidics and microengineering. *Journal of Micromechanics and Microengineering*, 24(3):035017, 2014.
3. Tae Kyung Kim, Jeong Koo Kim, and Ok Chan Jeong. Measurement of nonlinear mechanical properties of pdms elastomer. *Microelectronic Engineering*, 88(8):1982–1985, 2011. ISSN 0167-9317. . URL https://www.sciencedirect.com/science/article/pii/S0167931710005964. Proceedings of the 36th International Conference on Micro- and Nano-Engineering (MNE).

Reviewer Reports on the Second Revision:

Referees' comments:

Referee #1 (Remarks to the Author):

I appreciate that the authors modified the text in Supplementary Information about the effect of bulk modulus (comment 1A). I am also pleased that they provided additional numerical results in the revised Supplementary Materials in relation to my comment 1B although I'm not convinced by it.

By first considering individual capsules, the authors report new numerical results about the lensing capability of single collapsed capsules depending on the incident angle, ranging from 0° to 180°. Although it is regrettable that they were not performed on a single capsule in its initial spherical state, these simulations clearly show that both the transmittance and focusing efficiency are improved when the collapsed capsules are aligned with the incident beam.

On the other hand, the authors show that the transmission across an array of capsules is much higher when the capsules are randomly oriented compared to the case when they are all aligned with the incident beam. To justify this counter-intuitive observation, the authors added new simulations in the Supplementary Materials about the propagation of light rays across 2D arrays of capsules. By simple looking at snapshots, the authors note that the "coverage area" is decreased when the capsules are randomly oriented which explains why the transmission is higher in this later case. In my opinion, this argument is not convincing and used opportunistically. Furthermore, the results shown in the main text (Fig. 3C) lead the reader to believe that both improved transmission and contrast are due to lensing effects only (fig. 3C (i)).

Author Rebuttals to Second Revision:

Response to the Referees

Shell Buckling for Programmable Metafluids
Manuscript number: 2023-05-09013B

Adel Djellouli[a,1], **Bert Van Raemdonck**[b,1], **Yang Wang**[a,1], **Yi Yang**[a], **Anthony Caillaud**[a], **David Weitz**[a], **Shmuel Rubinstein**[c], **Benjamin Gorissen**[b,2], **and Katia Bertoldi**[a,2]

[a]J.A. Paulson School of Engineering and Applied Sciences, Harvard University, USA; [b]Department of Mechanical Engineering, KU Leuven and Flanders Make, Belgium; [c]The Racah Institute of Physics, The Hebrew University, Israel; [1]These authors contributed equally.; [2]These authors contributed equally.

Response to Referee #1

I appreciate that the authors modified the text in Supplementary Information about the effect of bulk modulus (comment 1A). I am also pleased that they provided additional numerical results in the revised Supplementary Materials in relation to my comment 1B although I'm not convinced by it. By first considering individual capsules, the authors report new numerical results about the lensing capability of single collapsed capsules depending on the incident angle, ranging from 0° to 180°. Although it is regrettable that they were not performed on a single capsule in its initial spherical state, these simulations clearly show that both the transmittance and focusing efficiency are improved when the collapsed capsules are aligned with the incident beam. To justify this counter-intuitive observation, the authors added new simulations in the Supplementary Materials about the propagation of light rays across 2D arrays of capsules. By simple looking at snapshots, the authors note that the "coverage area" is decreased when the capsules are randomly oriented which explains why the transmission is higher in this later case. In my opinion, this argument is not convincing and used opportunistically. Furthermore, the results shown in the main text (Fig. 3C) lead the reader to believe that both improved transmission and contrast are due to lensing effects only (fig. 3C (i)). .

We thank the reviewer for their thoughtful comments on our manuscript and appreciate their acknowledgment of the modifications made in response to previous comments.

Regarding the absence of simulations of a single capsule in its initial spherical state, we would like to highlight that characterizing the focusing efficiency of a shell presents challenges. In our simulations, we place the power sensor at the focal plane of the "lens" to assess focusing efficiency. However, for a spherical shell, which lacks a distinct focal plane (as it is not a lens), determining focusing efficiency becomes inherently challenging. The results of simulations for a spherical shell would be highly dependent on the arbitrary location of the power sensor.

Concerning the results presented in Fig. 3C(i), we want to clarify that these are included solely to emphasize the distinct scattering behaviors between spherical and collapsed capsules. This point is explicitly stated in the main text, and our intention is to illustrate the differences rather than attribute improved transmission and contrast solely to lensing effects. We hope this clarification addresses any potential misinterpretation.