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Droplet lift-off from hydrophobic surfaces from impact with soft-hydrogel spheres

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ABSTRACT

Droplet impacts on superhydrophobic surfaces may result in complete bouncing, with the absence of contact hysteresis and viscous dissipation leading the droplet to fully rebound off the surface. This rebound usually happens in the retraction phase, when the droplet retracts back after reaching a maximum spread diameter. Here, we present experimental evidence of a novel bouncing phenomenon where a sessile droplet on a hydrophobic surface bounces off the surface in its spreading phase when a soft deformable hydrogel sphere axisymmetrically impacts the droplet. We term this as ‘Lift-Off’ and propose a simple force balance based on the deformation characteristics of the hydrogel sphere to explain the out-of-plane jump of the droplet during spreading. We observe three different impact regimes, and propose their dependency on a modified elastic ‘Mach’ number (Ma^*) with $Ma^* = 0.2$ corresponding to the onset of lift-off. We also report on the unique acoustic signatures of lift-off cases, associated with the capture of air-bubbles through the air-borne retracting droplet rim. Concerning the novel lift-off results, this work may have potential applications for drainage and surface cleaning, non-stick surface coating, industrial mixing and plant disease spreading.

Droplet impacts are ubiquitous in nature^{1–6}, and have been extensively studied for over a century⁷ because of their immense importance in various industrial and chemical processes^{8–15}. Droplet impacts on superhydrophobic surfaces have been of particular interest, as these non-wetting impacts may exhibit complete rebound of the impacting droplets due to low viscous dissipation and contact line hysteresis of the surface^{16–23}. Upon impact, a droplet expands to reach a maximum diameter, then retract and rebound off the surface, with these mechanisms governed through the balance of inertia and capillarity^{20,22,24}. In binary drop impacts a droplet impacts a sessile droplet on a hydrophobic surface^{25–30}. These droplets might coalesce, and follow the ‘expansion-maximal deformation-retraction-bouncing’ pattern similar to a single droplet impact (supplemental figure 5). An unexplored variation to these binary impact cases involves the substitution of soft spheres for droplets. Figure 1a shows such an experimental setup where a soft deformable hydrogel sphere (diameter $D = 2R$) impacts a large sessile water droplet with an impact velocity u . The droplet resembles a flattened puddle with height $2\kappa^{-1} \sin(\theta_c/2)$, as its maximum azimuthal diameter ($d = 2r$) is greater than the capillary length $\kappa^{-1} \sim \sqrt{\sigma/(\rho g)}$ ³¹ (σ and ρ are the surface tension and density of water, g is acceleration due to gravity). The hydrophobicity of the surface (static contact angle $\theta_c \sim 141^\circ$, figure 1a) compels the droplet to take an oval shape on the surface, common for such large puddles ($d > \kappa^{-1}$), with the base contact length l of the droplet always following the $l < d$ rule. Upon impact with the surface, the soft hydrogel sphere significantly deforms, as shown in the image sequence of figure 1c. We quantify this deformation using stretch λ and strain ϵ ^{32–34} (supplemental figure 6(a,b)). We also take into account the growing base length L (figure 1(b, c)) of the hydrogel sphere in contact with the surface as it forces the droplet radially outwards from its pinned position. If $L > l$ in this spreading phase, the droplet that is attached to the sphere bounces off the surface.

We call this droplet bouncing mode ‘Lift-Off’, as the droplet is lifted off the surface through the deformation of the impacting soft sphere. One such case is presented in figure 1d, where a soft sphere ($D = 16$ mm) impacts a sessile large droplet (volume $V = 0.15$ mL) with impact velocity $u = 1.17$ m/s (figure 1d, $t = 0$ ms). In the initial moments the water rises up along the sphere surface as it plunges through the droplet. The sphere deforms upon impacting the aluminum substrate and the droplet starts to spread radially along the surface obtaining a maximum spreading velocity of v . The droplet lifts-off entirely from the surface at $t = 5.75$ ms. The droplet hang-time in air is notable ($O(10)$ ms), as the attached droplet moves upward with the rebounding soft sphere, all the while spreading radially to reach a maximum radius r_{max} ($t = 5.75 - 17.5$ ms). This droplet rebound mechanism is significantly different than conventional bouncing^{18,22,24,35} seen in single or drop-on-drop impact cases, where the droplet retracts after reaching r_{max} and then rebounds off the surface. For a sessile droplet with the same amount of water as 1d, if the sphere impact speed u is sufficiently high, we observe suppression of these lift-off characteristics. The

Figure 1. | Experimental setup and sphere-droplet impact dynamics. **a**, Schematic of the experiment: a soft hydrogel sphere (diameter D) impacts a droplet resting on a hydrophobic aluminum substrate (contact angle $\theta_c \sim 141^\circ$). The droplet takes on a flattened shape on the surface resembling a large puddle with a maximum height of $h_o = 2\kappa^{-1} \sin(\theta_c/2)$. The length of the droplet base is l (blue bars, $l < d$ where d is the maximum azimuthal diameter of the droplet). A microphone captures the acoustic signature of the impact event. **c**, On impact, the hydrogel sphere ($D = 16$ mm) deforms, with the sphere base length (L , red bars) increasing over time ($t = 0 - 3.8$ ms) and reaching a maximum value and then retracting to rebound off the surface. L exhibits a power-law trend as a function of non-dimensional velocity based time as shown in **b**. Equation of the best fit line (red dashed line) is $0.8(ut/D)^{-0.31}$. **d**, If L surpasses l when the hydrogel sphere impacts a resting droplet, the droplet simultaneously spreads radially ($t = 0 - 5.75$ ms) and lifts off vertically during this spreading leaving no water remnant on the surface. h_{max} is measured at r_{max} for consistency, even though the droplet continues to rise higher ($t = 25.5$ ms). **e**, Impact velocity of the hydrogel sphere plays an important role in determining whether the droplet would lift-off, as greater impact velocity results in dominant radial spreading and suppression of the droplet rebound. **f**, If maximum deformed base length of the hydrogel sphere (L_{max}) is less than l ($t = 8.5$ ms), the droplet fails to completely lift-off on sphere-impact, leaving a water puddle on the surface which connects to the rebounding hydrogel sphere in the form of a water column ($t = 8.5 - 28$ ms). **g**, This behavior is similar to the impact of a rigid sphere ($D = 16$ mm) on a droplet. Here also, we see $L_{max} < l$ similar to no lift-off case in **f**. The droplet does not completely separate from the substrate in this case either, indicating that deformation of the soft sphere is essential to droplet lift-off. Supplementary movies S1-S5 show these impact events in their entirety.

droplet rapidly spreads radially along the surface as a thin water sheet in this case (figure 1e, $u = 1.53$ m/s), reaching r_{max} before breaking up into multiple droplets due to inertia of the spreading overcoming capillarity. We call these impact cases ‘Sheet’, denoting the formation and expansion of the thin water sheet on the surface after impact. The sheet cases also show a small lift-off of water from the surface at the very initial moments (figure 1e, $t = 6.5$ ms), but they are short-lived ($O(1)$ ms) compared to the lift-off cases. Varying the amount of water in the droplet and lowering the sphere impact velocity result in less deformation of the sphere base ($L_{max} < l$). The droplet still spreads radially after sphere impact with a lift-off of the rim, but a liquid column connecting the water attached to the rebounding sphere with a droplet remnant on the surface forms (figure 1f), preventing total lift-off. These ‘No Lift-Off’ cases are similar to that of a rigid sphere impacting a sessile droplet. In figure 1g a rigid plastic sphere of minimal deformation ($L_{max} \ll l$) lands on a droplet forming a liquid column after rebound, similar to particle impact with shallow pools³⁶. A consistent observation from all these different types of impact is that a large deformation ($L_{max} > l$) enables the sphere to displace the entire droplet contact region, which is essential for complete lift-off.

We propose that this large sphere deformation applies a downward force on the droplet enabling the droplet to lift-off. The deformation characteristics of the soft hydrogel sphere are investigated to validate this hypothesis. The hydrogel spheres used in this study are commercially available cross-linked Acrylamide-chain spheres that experience strain under impact loading, as defined in figure 2a. Compression tests reveal that the hydrogel spheres show a Hertzian response, with the indentation

Figure 2. | Hydrogel sphere deformation leads to lift-off of resting droplet. **a**, Schematic of the deformation of the impacting soft hydrogel sphere. The initial diameter D increases on impact, leading to sphere height reduction from H_o . Strain ε is defined as the ratio of diameter deformation ($D' - D$) (D' is the instantaneous deformed diameter) and corresponding deformed sphere height, H . The indentation is defined as $\delta = (H_o - H)$. **b**, The deformation of the hydrogel spheres leads to radial base propagation, with the curved sides of the elongating sphere applying a normal force, F , on the droplet. ' θ ' shows the angle this curved side of the sphere creates with the surface (measured when $L_{max} = l$). The vertical and horizontal components of F leads to rebound and radial propagation of the droplet respectively. **c**, The two force components affects the droplet in different ways. The horizontal component pushes the droplet rim radially outwards which glides over the thin air layer on the hydrophobic surface (3.75 ms). The vertical force component pushes the rim down, and the rim bounces off the air layer which acts as a spring (4.75 ms). Finally, we see **complete** lift-off (5.75 ms). Images showing the same drop event shown in figure 1a. **d**, The angle θ plays an important role in controlling lift-off. With increasing velocity ' θ ' becomes greater. At lower θ values $F \cos \theta$ dominates, leading to lift-off cases with greater lift-height h_{max} . h_{max} is largest at $\tan \theta \sim 1$. The red and blue colored markers indicate droplet volumes of $V = 0.15$ mL and 0.30 mL respectively.

of the sphere $\delta = H - H_o$ under compression following the slope of $3/2$ (Supplemental figure 7). This indicates that these soft gel spheres act as elastic materials, and we can use the Hertz theory for deformation of an elastic sphere to determine the Young's modulus (Supplemental Eq. 7)^{32,34,37}, which in turn can be used to approximate the shear modulus G for the spheres (assuming Poisson's ratio $\nu = 0.5$ under the incompressibility condition). G has values that range in between 6 KPa to 8 KPa, quantifying the softness of the spheres (see supplemental section 0.5 for detailed analysis of G). The characteristics deformation behavior of the hydrogel spheres translates into a growing base length L over time. The resulting continuous change of sphere surface curvature (figure 1c) acts as a force F on the droplet as drawn in figure 2c. This change in curvature can be tracked by measuring the angle (θ) the sphere surface curvature makes with the substrate. θ is measured when $L_{max} = l$ since this displacement condition needs to be fulfilled for droplet lift-off. The force F applied on the droplet by this changing curvature can be broken up into components, with the horizontal component $F \sin \theta$ contributing to the radial spreading of the droplet. The vertical component $F \cos \theta$ acts normal to the impact surface, pushing the droplet downwards. Since the surface is hydrophobic with a Cassie-Baxter state³⁸, the droplet does not get pinned to the substrate under this normal force. The droplet is instead caught between the sphere and substrate boundaries, and is forced to move up (lift-off) and out (radial spread). The strength of lift-off is $F \cos \theta$, which is a function of impact speed u (figure 2d, color-bar). Quantitatively, the effect of θ can be seen in figure 2d, where max lift-off occurs when $F \cos \theta = F \sin \theta$. It is evident that lift-off height decreases for higher values of θ , since $F \sin \theta$ far exceeds $F \cos \theta$. This hints at the possibility of suppression of lift-off at high sphere impact speeds, as confirmed experimentally in figure 1e.

Although this force F is difficult to directly quantify, its features provide a physical understanding of the lift-off mechanism in the droplet spreading phase. We can quantify different features of sphere-droplet impacts using elastic deformation, impact kinetic energy and droplet spreading. For a large maximal spreading factor $\lambda_{max} \gg 1$, where $\lambda = D'/D$ with D' being the azimuthal diameter of the impacting sphere during any given moment of deformation, the maximum bulk elastic energy can be expressed as $E_{bulk}^{max} = (\pi D^3/6)G\lambda_{max}^2$ ^{32,33}. Balancing this expression with the impact kinetic energy of the hydrogel sphere

75 $E_k = (1/2)\rho_R(\pi D^3/6)u^2$ (where ρ_R is the hydrogel density), we find

$$76 \quad \lambda_{max} \sim \frac{u}{\sqrt{G/\rho_R}} \sim \frac{u}{C_s} = Ma \quad (1)$$

77 where $C_s = \sqrt{G/\rho_R}$ is the speed of shear wave in hydrogel sphere material and Ma is the elastic Mach number for the hydrogel
 78 sphere material. Ma characterizes the deformation of the hydrogel sphere. The impact kinetic energy of the sphere translates
 79 into radial spreading of the droplet with velocity v . If we use a simplified version of the sphere impact kinetic energy $\rho_R u^2 R^3$
 80 (where $R = D/2$) and the droplet spreading energy $\rho v^2 r^2 h_o$ (where ρ is the density of water, v is the spreading velocity of the
 81 droplet, $r = d/2$ and h_o is the droplet height), we obtain $u \sim \alpha v (r/R)^{3/2} \sqrt{h_o/r}$, where $\alpha = 1/0.4825$ is a fitting parameter.
 82 Figure 3a shows the experimentally measured u and v data for different impact combinations of impact velocity ($u = 0.76 -$
 83 3 m/s) and different droplet sizes ($V = 0.15 - 0.30$ mL), and they are in good agreement with the result of the energy balance.
 84 This energy balance is appropriate for low viscosity fluids, where the spreading results from pure transfer of kinetic energy into
 85 surface energy. Substituting u in Eq. 1 results in the modified Mach number (Ma^*),

$$86 \quad Ma^* \sim Ma \left(\frac{R}{r}\right)^{3/2} \sqrt{\frac{r}{h_o}}, \quad (2)$$

87 where $Ma^* = v/C_s$. Ma^* includes all the input parameters involved in the sphere-droplet impact events. Figure 3b plots Ma^*
 88 versus L_{max}/l for all impact experiments analyzed, and produces a regime diagram that shows two experimentally observed
 89 conditions that need to be fulfilled for lift-off. The first one is the previously mentioned displacement condition: (i) $L_{max}/l > 1$.
 90 The second condition is the ‘Mach’ condition or the deformation condition: (ii) $Ma^* > 0.2$, which provides the experimental
 91 condition of sphere deformation controlling the different outcomes for the droplet, especially the onset of droplet lift-off.

92 Using Eq. 2, we can also make a prediction about the time it takes for the droplet to lift-off from the surface after hydrogel
 93 sphere impact. The time to lift-off or τ_c is analogous to contact time of conventional single droplet rebound, which in the latter
 94 case is proportional to $\sqrt{\rho r^3/\sigma}$, an increasing function of droplet radius r that does not depend on droplet impact velocity^{29,35}.
 95 Herein, lift-off happens earlier in the droplet spreading phase, and is approximated to be $\tau_c \sim r/u$. Rearranging Eq. 2,

$$96 \quad \tau_c \sim \frac{r}{u} \sim \frac{R^{3/2}}{v\sqrt{h_o}}, \quad (3)$$

97 a detailed derivation given in the supplemental section 0.6. Experimental measurements follow the proposed scaling as shown
 98 in figure 3c.

99 We can also estimate the maximum spreading ratio (r_{max}/r) of a droplet after the soft sphere impact. For single or
 100 drop-on-drop conventional bouncing cases in low viscosity fluids, the spreading ratio is a function of droplet Weber number,
 101 $r_{max}/r \sim We^{1/4}$ where $We = \rho u^2 r / \sigma$ ^{20,29}. We can arrive at this expression by applying volume conservation to the droplet
 102 spreading after impact. In hydrogel sphere-droplet impact cases, the transfer of kinetic energy from sphere to droplet drives
 103 spreading. The initial volume of the droplet can be scaled as $r^2 h$, where the droplet thickness h can be obtained through
 104 $\sqrt{\sigma/(\rho g)}$. After the sphere impacts the droplet, the droplet undergoes deformation, with r and h changing over time. Clanet, et
 105 al.²⁰ proposed that for a single droplet impact, the typical acceleration experienced by the spreading droplet scales as u^2/r ,
 106 where u is the droplet impact speed. This would alter the formulation for the droplet thickness to $h = \sqrt{\sigma r/(\rho u^2)}$. Following
 107 the same argument, and using the droplet spreading velocity v in place of u , we can scale the droplet maximum spread as
 108 $(r_{max}^2 - R^2) \sqrt{\sigma r/(\rho v^2)}$. We subtract sphere radius R from the droplet maximum radius, since after impact the droplet takes on
 109 a torus shape around the sphere. Subsequently, we obtain the spreading ratio,

$$110 \quad \frac{r_{max}}{r} \sim \left[\left(\frac{R}{r}\right)^2 + We^{1/2} \left(\frac{h_o}{r}\right) \right]^{1/2}. \quad (4)$$

111 where We is the droplet spreading weber number ($We = \rho v^2 r / \sigma$). Detail derivation for Eq. 4 is given in the appendix section 0.7.
 112 Figure 3d shows a comparison of the experimentally measured spreading factor data to values predicted through Eq. 4. The
 113 comparison reveals the proposed relationship works well over a range of different droplet volumes. This only holds for liquid
 114 droplets of low viscosity whereas higher viscosity has been shown to inhibit the spreading of droplet after sphere impact²⁰.

115 Figure 4a presents an image sequence of the retraction dynamics of a lifted-off droplet in flight after it has reached maximum
 116 spreading ratio r_{max}/r . At this point, the droplet spreading velocity $v \sim 0$, and the capillarity of water dominates, causing the
 117 droplet rim to move inwards ($t = 14 - 26$ ms). As the droplet remains attached to the upwards moving rebounding sphere, the
 118 thin water sheet that connects the rim of the droplet with the sphere also moves upward. These two motions create a hollow
 119 liquid column over time, with the inbound droplet rim at the bottom. The surface tension in the rim forms multiple fingers

Figure 3. | Energy balance and regime diagram. **a**, Energy balance between the sphere impact and droplet spreading reveals maximum droplet spreading velocity v to be a function of impact velocity u , and sphere and droplet sizes respectively. Plotting the experimentally measured v against the calculated functional form shows a one to one relation, using a fitting parameter of 2.072. **b**, This leads to the formulation of a modified Mach number Ma^* , which plotted against L_{max}/l shows $Ma^* = 1$ is the tentative onset of lift-off. This regime plot also reveals another condition of lift-off, $L_{max}/l > 1$. **c**, The lift-off time (τ_c) of the droplet, measured from the moment of sphere impact on droplet to the first moment of droplet rim leaving the surface, plotted as a function of Ma^* . The experimentally measured values of τ_c scales with the lift-off times found from Eq. 3. **d**, The spreading ratio of the droplet after soft sphere impact can be predicted using the scaling law proposed through Eq. 4. The experimentally measured spreading factor values exhibit the same predicted slope. In all the plots, the red, blue, and green marker colors denote the droplet volume, $V = 0.15, 0.45$ and 0.30 mL respectively. Uncertainty bands are marked with a 95% confidence.

120 which later collapse inward ($t = 26 - 33$ ms) leading to the hollow column closing from below, trapping a few air bubbles
 121 ($t = 33 - 38$ ms). These bubbles coalesce and form a large bubble which oscillates with a distinctive frequency ($t = 41$ ms,
 122 figure 4b). We capture the sound of bubble entrapment and oscillations using a synchronized microphone (Earthworks QTC-40,
 123 bandwidth 5 Hz - 40 KHz) sampling at 96 KHz. The acoustic signal reveals a clear peak and high-frequency oscillation
 124 around 41 ms, indicating that this novel lift-off bouncing mode has distinct acoustic characteristics (figure 4c,d). The spectrum

Figure 4. | Bubble capture and collapse in mid-air through droplet retraction in lift-off cases. **a**, Image sequence showing the retraction phase of the droplet. The hydrogel sphere impacts a droplet (volume, $V = 0.30$ mL) at time $t = 0$ with an impacting velocity of 1.17 m/s. The droplet bounces off and reaches the maximum spreading radius at $t = 14$ ms. Afterwards, the rim starts to retract inwards with the upwards motion of the rebounding sphere, a reverse cone develops with several spherical fingers at the edge of the rim ($t = 26$ ms - 33 ms). This hollow cone collapses on itself ($t = 38$ ms) trapping a significant amount of air inside the resultant liquid jet ($t = 38$ - 41 ms). **b**, The air inside coalesces into a large bubble (white box), that oscillates at resonance frequency. **c**, The oscillation is captured in the microphone as an acoustic signal. Acoustic signal captured for a no lift-off case is also plotted for comparison. As the no lift-off case show no bubble capture (figure 1f), no discernible sounds are measured. 0 ms marks the time of sphere impact. **d**, A power spectrum indicates a resonance frequency $f_s \sim 4$ kHz.

shows the resonance frequency of the bubble to be $f_s \sim 4$ KHz. Using the Minnaert frequency formula for air bubbles³⁹, we calculate the approximate volume equivalent droplet radius to be 0.85 mm which is close to the measured droplet volume equivalent radius of 1.1 mm from the image (figure 4b). This further strengthens the argument that lift-off has unique acoustics characteristics. It is to be noted here that this signature acoustic behavior of lift-off cases are generally dominant at the low impact velocity lift-off cases. As impact velocity increases, the lift-off cases transition to the sheet cases, where instead of having one dominant bubble oscillation multiple oscillations and noise throughout the impact event follow from the capillary breaking of the sheet into multiple droplets (see supplemental figure 9).

In summary, we have presented experimental observations along with the theoretical analysis of a new type of droplet rebound mechanism in binary impact cases for a deformable hydrogel sphere impacting a sessile droplet on a hydrophobic surface. This novel lift-off of water droplet results from the deforming characteristics of the hydrogel sphere. This type of impact can also produce two more regimes: ‘No Lift-Off’ and ‘Sheet’. We propose a modified Ma^* number to quantify different regimes, and propose two conditions for lift-off: (i) $L_{max}/l > 1$ & (ii) $Ma^* > 0.2$. In addition to these fundamental observations, our findings show bubble entrapment through an air-borne retracting droplet rim. The uses for hydrogel spheres are growing for chemical and microfluidics processes^{40,41}, such that this water-scooping technique along with air bubble entrapment and acoustic signature in receding phase may be useful in certain scenarios, e.g. mixing and emulsification, surface cleaning. It may

also have potential implications in plant disease spreading through droplet bouncing⁶ with increasing use of hydrogel spheres as crop protection agents (CPA)⁴².

Methods

0.1 Preparation of hydrogel spheres and hydrophobic surface.

The soft hydrogel spheres used for this study are commercially available growing spheres from **Educational Innovations (GB-710)**. The dehydrated hydrogel particles were submerged in 100% de-ionized water, and were left to grow undisturbed for 24 hours. The mean final mass of the spheres used for the impact experiments was 2.5285 ± 0.2386 gm. An aluminum plain surface was used as the metal substrate, made hydrophobic by applying Glaco-Mirror Coat Zero superhydrophobic coating, which yielded a static contact angle of $\theta_c \sim 141^\circ$. A polystyrene sphere (also from **Educational Innovations**) of diameter $D = 16$ mm, mass = 2.10 gm and shear modulus $G = 750$ MPa (data cited from manufacturer's manual) was used for the solid sphere impact experiments.

0.2 Binary drop experiments

The hydrogel spheres were dropped from different heights, ranging from 30 mm - 400 mm. The free falling spheres impact the droplets and then the aluminum substrate below with velocities in 0.75 m/s - 3 m/s range. The droplets were deposited on the surface using a pipette, with the volume of water in the droplet carefully controlled. Two droplet volumes were used for most cases, 0.15 ± 0.05 mL and 0.30 ± 0.05 mL, and for a few cases a 0.45 ± 0.05 mL droplets were also used. A Phantom v2511 high-speed camera was used to record the impact events at 31,000 frames per second, with an LED array worked as the backlight. Images from the video were processed in MATLAB.

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243 **Competing interests**

244 The authors declare no competing interests.

Supplementary information

0.3 Uncertainty

The measurement uncertainty plotted through error-bars in the figures 3(a-d) are calculated using the Taylor series method (TSM)⁴³. For brevity, only the detailed breakdown of the uncertainty analysis for the modified Ma^* number calculation is shown below.

The expression for the modified Mach number can be written as $Ma^* \sim v/C_s$, which through simplification becomes,

$$Ma^* \sim \frac{v}{C_s} = \sqrt{\frac{\rho_R}{G}} v,$$

where v is the maximum droplet radial spread velocity, C_s is the material shear wave velocity, ρ_R is the hydrogel density and G is the total shear modulus of the hydrogel sphere material. Applying TSM to calculate the uncertainty relating to Ma^* ,

$$u_{Ma^*}^2 = \left(\frac{\partial Ma^*}{\partial v}\right)^2 U_v^2 + \left(\frac{\partial Ma^*}{\partial G}\right)^2 U_G^2, \quad (5)$$

where U_v^2 is the error relating to v and U_G is the error in shear modulus G . Now,

$$\frac{\partial Ma^*}{\partial v} = \sqrt{\frac{\rho_R}{G}}, \quad \frac{\partial Ma^*}{\partial G} = -\frac{v\sqrt{\rho_R}}{2G^{3/2}}$$

The error U_v originates from the experimental measurement of maximum droplet radial spread velocity v . To calculate v , we followed the spline fitting method laid out by⁴⁴, and followed the error calculation procedure presented there to obtain U_v (found to be in the order of $O(10^{-2})$). It should also be noted that this error calculation in the spline fitting of data also produces absolute errors for the dynamic radial spread values measured over time, which was used to quantify the measurement for figure 3d. U_G is obtained from the confidence interval found from the power law fitting of the shear modulus data measured by the Rheometer ($U_G = 0.085$). Substituting these values in Eq. 5,

$$u_{Ma^*} = \sqrt{\frac{\rho_R}{G} U_v^2 + \frac{\rho_R v^2}{4G^3} U_G^2} \quad (6)$$

Eq. 6 is the uncertainty approximation of the Ma^* .

0.4 Stretch λ & maximum strain ϵ_{max}

When the hydrogel bead impacts the underlying metal surface below, it undergoes deformation which can be defined through the parameter stretch (λ). Supplemental Figure 6a shows how λ varies over time for three different impact velocities. The value of λ is important, because the Ma and by extension the Ma^* formulation holds for $\lambda_{max} > 1$. For the impact velocities covered in this study, λ_{max} is always greater than 1. Figure 6b shows the non-linear scaling of maximum strain ϵ_{max} with non-dimensional contact time of the soft hydrogel sphere, ut_c/D . The contact time t_c is the time required for the sphere to rebound, measured from the moment of first impact with the surface. Data is presented for all the drop heights H , in the range of 30 mm - 400 mm.

0.5 Measurement of shear modulus, G

A custom made compression rig was built to measure the Young's modulus of the soft hydrogel spheres^{34,45}. The spheres were placed between a rigid plate connected to a micrometer and a scale. By adjusting the micrometer, the indentation of the spheres and the corresponding forces were measured. Figure 7a shows the data on a log-log scale, and reveals that the data follows the

Figure 5. Binary droplet impact. Image sequence of binary impact of water droplets. A 2.85 mm diameter sessile water droplet rests on a hydrophobic surface ($\theta_c = 141^\circ$). Another water droplet (diameter 2.54 mm) impacts the sessile droplet with an impact velocity $u = 0.7$ m/s. Upon impact, the droplets coalesce, and spreads radially (0 - 7.75 ms). After reaching maximal spreading, the coalesced single droplet starts to retract (10 - 20.2 ms), and eventually rebounds off the surface (35.5 ms).

Figure 6. Stretch (λ) & strain (ϵ). **a.** The deformation of the hydrogel bead after impact can be characterized using the stretch (λ) parameter, where $\lambda = (D'/D)$. D is the maximum undeformed sphere azimuthal diameter and D' is the maximum deformed sphere diameter at any given moment during deformation. Data for three different drop heights are plotted, and the maximum stretch in all these cases are greater than one, $\lambda_{max} > 1$. **b.** After impact, the soft hydrogel sphere undergoes significant strain (ϵ). Maximum strain value ϵ_{max} scales with the non-dimensional contact time ut_c/D , where t_c is the time the sphere remains in contact with the surface, from impact to rebound. The equation of the best of fit line: $0.3318\epsilon_{max}^{1.84}$.

3/2 power law typical for Hertz theory for a linear isotropic elastic sphere. The experimental fit (green dashed line) can then be used to determine the Young's modulus of the sphere material by employing the relation below³⁷,

$$F(\delta) = \frac{4Y}{3\sqrt{2}(1-\nu^2)} D_o^{1/2} \delta^{3/2} \quad (7)$$

where Poisson's ration $\nu = 0.5$, assuming the incompressibility condition. The shear modulus G for the spheres can be calculated from the relation $G = Y/2(1 + \nu)$, which is necessary for the calculation of the Mach (Ma) number. For different sphere diameters, the G values in this experiment varied from 6000 to 8000 KPa.

Another method of calculating the G values of the soft hydrogel spheres would be through investigating the bi-axial deformation behavior of the soft spheres in terms of modal frequency. The spheres would undergo cyclical axial deformation, which is plotted in figure 7b. The frequency of the deformation cycle can be used to find the shear modulus G through the characteristics equation⁴⁶,

$$G = \frac{\rho \omega_n^2 R^2}{(2.665)^2} \quad (8)$$

where $\omega_n = 2\pi/T$, the natural angular frequency of deformation for the sphere (T is the period of the oscillation). Figure 7 provides two different candidates for T , the values T_1 and T_2 showing consecutive periods of oscillation for the impacting sphere. Using both values in Eq. 8, we find the range $5244 < G < 9500$ Pa. We can conservatively assume that the G is in range, as it also agrees well with our previously measured values from Eq. 7.

0.6 Lift-off time, ' τ_c '

The lift-off time τ_c is defined as the time the droplet takes to lift-off of the surface, calculated from the moment when the sphere first impacts the droplet to the moment the droplet rim leaves the surface. We can derive τ_c mathematically by rearranging

Figure 7. Measuring the elastic properties of the deformable sphere. **a**, The hydrogel spheres are put under compression in a custom made setup. Forces measured corresponding to differing indentations δ (figure 2a) of the sphere are plotted on log-log scale. The red and blue marker colors denote two different sizes of spheres used in the study. Data follow a $3/2$ power law consistent with Hertz theory for a linear isotropic elastic sphere, indicating the Hertz theory can be used to calculate the Young's modulus Y . Shear modulus G is obtained through the Poisson's ratio relationship. **b**, The soft hydrogel spheres go through cyclical deformation upon impact. The frequency of the deformation oscillation can be used to approximate shear modulus G from Eq. 8.

Eq. 1,

$$\begin{aligned}
 Ma^* &= Ma \left(\frac{R}{r}\right)^{3/2} \sqrt{\frac{r}{h_o}} \\
 \frac{u}{\alpha C_s} \left(\frac{R}{r}\right)^{3/2} \sqrt{\frac{r}{h_o}} &= Ma^* \\
 \frac{u}{r} \frac{R^{3/2}}{C_s \sqrt{h_o}} &= Ma^* \alpha \\
 \tau_c^{-1} &= \alpha Ma^* \frac{C_s \sqrt{h_o}}{R^{3/2}} \\
 \tau_c &= \frac{R^{3/2}}{\alpha v \sqrt{h_o}}
 \end{aligned} \tag{9}$$

where (r/u) scales the lift-off time τ_c , $Ma^* = v/C_s$ and α is the fitting parameter used in the energy balance scaling of u & v .

0.7 Derivation of Eq. 4: spreading ratio ' r_{max}/r '

When a hydrogel sphere impacts the resting droplet, the droplet becomes attached to the sphere surface and spreads radially to reach maximum spread resulting from the competition between capillarity and inertia, after which it would retract back on itself. The droplet spreading velocity reach a maximum spreading velocity v , and then reach 0 when it reaches maximum spreading radius r_{max} . During spreading, the droplet thickness h , which is initially h_o , should also change. Usually, h is defined as a function of capillary length $\kappa^{-1} = \sqrt{\sigma/(\rho g)}$. Clanet, et al.²⁰ showed that during spreading, a droplet would experience acceleration scaled as v^2/r . The thickness h then scales with a modified capillary length $h \sim \sqrt{\sigma r/(\rho v^2)}$. Also, the presence of the soft sphere alters the spreading geometry of the droplet, which in its spreading phase resembles a torus. To calculate the effective maximum spread radius of the droplet, we subtract the sphere diameter R . Taking these modifications into account and

Figure 8. Droplet spread velocity. **a**, A typical lift-off regime droplet spread velocity profile. A 16 mm hydrogel sphere impacts a sessile droplet ($V = 0.30$ mL) with an impact velocity of 0.89 m/s. The droplet spreads with a velocity v' , reaching a maximum $v'_{max} = v \sim 0.7$ m/s at $t = 8$ ms. After reaching the maximum, v' decreases over time and crosses $v' = 0$, indicating the onset of the retraction phase ($t = 16$ ms). The black dashed vertical line separates the two phases: spreading and retraction. **b**, Application of energy balance leads to a one to one relationship between sphere impact velocity u and droplet spreading velocity v , with a fitting parameter of 2.072. For solid sphere impacts (red filled diamonds), the droplet spread velocity shows a slightly different scaling relation, suggesting a modification of the fitting parameter is needed to obtain the exact one to one relation between u and v in this case.

using volume conservation, we find,

$$r^2 h_o \sim (r_{max}^2 - R^2) \sqrt{\frac{\sigma r}{\rho v^2}}$$

$$r^4 h_o^2 \sim (r_{max}^2 - R^2)^2 \frac{\sigma r}{\rho v^2}$$

$$\left(\frac{r_{max}^2 - R^2}{r^2}\right)^2 \sim \frac{\rho v^2 r}{\sigma} \left(\frac{h_o}{r}\right)^2$$

$$\frac{r_{max}^2 - R^2}{r^2} \sim We^{1/2} \left(\frac{h_o}{r}\right)$$

$$\frac{r_{max}^2}{r^2} \sim \frac{R^2}{r^2} + We^{1/2} \left(\frac{h_o}{r}\right)$$

$$\frac{r_{max}}{r} \sim \left[\frac{R^2}{r^2} + We^{1/2} \left(\frac{h_o}{r}\right)\right]^{1/2}$$

(10)

We note that the substitution $h \sim \sqrt{\sigma r / (\rho v^2)}$ only holds when $v^2/r > g$. For no lift-off cases and the low sphere impact velocity lift-off cases, $v^2/r \sim g$, and it is not until later when the $v^2/r > g$ condition is met. We conjecture this as the reason why the scaling is less accurate in the early stages (no lift-off cases), and later becomes better at predicting the spreading ratio as $v^2/r > g$.

Figure 9. Acoustic characteristics of sheet regime. **a**, Image sequence of a hydrogel sphere ($D = 16$ mm) impacting a sessile droplet ($V = 0.30$ mL) with a velocity of 1.98 m/s, resulting in the distinctive sheet case as in figure 1e. **b**, The subsequent audio signal is captured by the synchronized microphone. Significant oscillations can be seen at different times of the impact event, the vertical lines corresponds to the same colored boxes in the image sequence of **a**. **c**, FFT shows the frequency spectrum of the signal with a broadband noise.

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