LETTER

Drop trampoline

A Letters Journal Exploring the Frontiers of Physics

October 2018

EPL, **124** (2018) 24003
doi: 10.1209/0295-5075/124/24003

www.epljournal.org

Drop trampoline

Pierre Chantelot, Martin Coux, Christophe Clanet and David Quéré

PMMH, UMR 7636 du CNRS, ESPCI Paris, PSL Research University, Sorbonne Université, Université Paris Diderot - 75005 Paris, France and
LadHyX, UMR 7646 du CNRS, Ecole polytechnique - 91128 Palaiseau, France

received 26 August 2018; accepted in final form 15 October 2018
published online 22 November 2018

PACS 47.55.dr – Interactions with surfaces

Abstract – Rigid superhydrophobic materials have the ability to repel millimetric water drops, in typically 10 ms. Yet, most natural water-repellent materials can be deformed by impacting drops. To test the effect of deformability, we perform impacts of non-wetting drops onto thin ($\sim$10 μm), circular PDMS membranes. The bouncing mechanism is markedly modified compared to that on a rigid material: the liquid leaves the substrate as it is kicked upwards by the membrane. We show that the rebound is controlled by an interplay between the dynamics of the drop and that of the soft substrate, so that we can continuously vary the contact time by playing on the membrane's characteristics and reduce it up to 70%.

Liquids and soft solids can interact in many ways. For instance, a drop can deform a soft solid close to the contact line [1,2] or bend slender and thin structures [3,4]. In more dynamical situations, substrate deformation can induce self-propulsion [5,6], prevent splashing [7,8] or allow liquid penetration in soft solids [9]. It can also lead to improvement of the water or ice repellency of superhydrophobic materials [10,11]. Such materials have the ability to reflect impacting drops and we focus here on the way their flexibility affects this property. The contact time τ of millimetric water drops on a rigid, repellent solid is on the order of 10 ms, which can be large enough to induce freezing [12], significant heat transfer [13] or contamination by surfactants [14]. Different techniques have been proposed to reduce τ. Decorating the substrate with macrotextures (such as ridges) was found to divide τ by a factor of typically 2 [15,16], a reduction also obserbved using soft membranes, as recently shown by Weisensee *et al.* [17]. This effect occurs at large impact velocity, in a regime difficult to explore due to splashing, which might explain the scattered nature of the results. Vasileiou *et al.* also stressed the ability of soft membranes to reflect viscous drops —a point of obvious practical interest— but did not provide neither a specific study on the contact time nor a model to account for its reduction [11]. In order to discuss systematically the ability of soft solids to enhance water repellency, we choose to texture liquids rather than solids, that is, to use liquid marbles as a model of non-wetting drops [18]. This allows us to show that the interplay between flexible substrates and non-wetting impacts leads to the possibility of continuously tuning the contact time, which we model.

The experiment is sketched in fig. 1(a). Our substrates are polydimethylsiloxane (PDMS) sheets with thickness $h = 20\,\mu$m (Silex) clamped between two plexiglas rings. The clamped sheets are placed on a frame with radius a (a = 7.5, 10, 17.5, 25 mm) and tension is adjusted by weighting the membrane with a mass m. Liquid marbles are made by coating distilled water (density $\rho = 1000\,\mathrm{kg/m^3}$ and surface tension $\gamma = 72\,\mathrm{mN/m}$) with lycopodium grains (diameter $\sim 30\,\mu$m) treated with fluorodecyl-trichlorosilane. Because the grains occupy less than one monolayer at the liquid/air interface, the marbles keep a surface tension ($\gamma \simeq 57 \pm 8\,\mathrm{mN/m}$) comparable to that of water [18,19]. They non-wet any susbtrate on which they are placed as contact only occurs between the grains and the solid. Liquid marbles show the same remarkable mobility as non-wetting drops. They can resist impacts of moderate velocity, a property enhanced on a repellent substrate. To that end, the membranes are made water-repellent by spraying a solution of hydrophobic nanobeads dispersed in acetone (Ultra Ever Dry, UltraTech International). After evaporation of the solvent, the PDMS is coated by a few layers of nanobeads and it exhibits the high contact angles ($\sim 160°$) and low hysteresis ($\sim 6°$) typical of superhydrophobic materials. Water drops ($R = 1$ mm and $R = 1.8$ mm) made from calibrated needles roll on a horizontal groove covered by lycopodium grains, until they get coated. Then we release them above

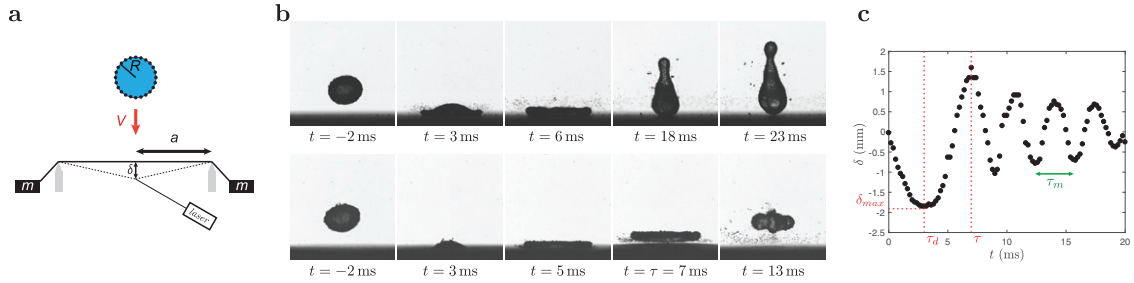

Fig. 1: (Colour online) (a) A liquid marble (radius R, velocity V) impacts a circular PDMS membrane with radius a and thickness $20\,\mu$m put into tension by a mass m. The membrane is superhydrophobic and its deflection δ is measured by the deviation of an oblique laser sheet. (b) Top: a liquid marble ($R = 1.8$ mm) impacts a rigid superhydrophobic substrate at $V = 0.75$ m/s. The drop leaves the substrate after 22.5 ms. See the corresponding movie `Movie1_trampo.avi` in the supplemental material. Bottom: same liquid marble impacting a flexible membrane ($a = 10$ mm, $m = 12.23$ g) at $V = 1$ m/s. The drop is kicked off, with a pancake shape, after 7 ms, a reduction of 70% compared to the rigid case. See the corresponding movie `Movie2_trampo.avi` in the supplemental material. (c) Deflection δ of the center of the membrane for the impact shown in (b), from which we obtain the maximal deflection δ_{max} and its time τ_d. Later, the free oscillations of the membrane give access to its fundamental period τ_m.

the substrate through a hole at the end of the groove. The impact velocity V can be varied between 0.5 and 1.5 m/s by adjusting the height from which these marbles fall. We record side views of the impact and monitor the membrane deflection δ through the observation of a laser sheet in oblique incidence, using two fast video cameras (Phantom V7) working at typically 10000 frames per second. The vertical position of the membrane is directly proportionnal to the displacement of the laser sheet.

Marbles are repelled differently by flexible or rigid materials (fig. 1(b)). In the latter case (top sequence), they spread, recoil and take off with an elongated shape, here after 22.5 ms. We also notice excess grains expelled from the marbles during impact, without affecting the integrity of these objects. The contact time τ of a marble with radius $R = 1.8$ mm is roughly independent of the impact velocity V and plateaus at $\tau_0 = 22.7 \pm 0.8$ ms (see fig. 1 in the supplementary material `Supplementarymaterial.pdf` (SM)), a value comparable to that of a non-wetting drop. In contrast (bottom sequence), a marble impacting a flexible membrane deforms its substrate, which transiently renders it invisible in our side views. As the membrane recovers its horizontality, we observe that the spread marble takes off with a flattened shape, which implies a reduction of contact time. τ in fig. 1(b) is 7 ms, that is, about one third of τ_0, a reduction even larger than that reported in [11,17]. Recoiling takes place later, while the drop is in the air. Figure 1(c) shows the time evolution of the deflection δ of the center of the membrane. Firstly, the membrane sinks down to its maximal deflection δ_{max} at time τ_d (see fig. 2 in the SM). Then, the substrate moves back and goes above the horizontal, which kicks the marble and makes it take off (at time τ) at the membrane's uppermost position. Later, it freely oscillates, allowing us to measure its natural period, here $\tau_m = 3.45 \pm 0.10$ ms. These free oscillations are faster than the first oscillation forced by impact, showing that our system has a characteristic time τ intermediate

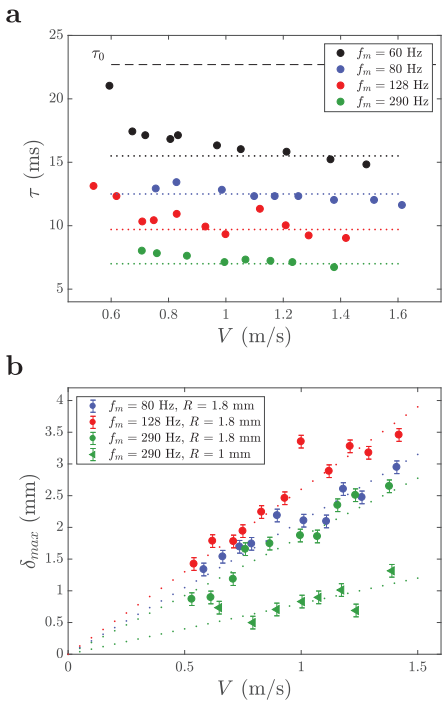

Fig. 2: (Colour online) (a) Contact time τ of liquid marbles with radius $R = 1.8$ mm bouncing off flexible membranes of frequency f_m as a function of impact velocity V. The dashed line shows the contact time τ_0 on a rigid substrate. (b) Maximal deflection δ_{max} at the center of the membrane as a function of V. Dotted lines are linear fits.

between τ_m and τ_0, the respective response times of the membrane and of the drop.

The time τ_m (and corresponding frequency $f_m = 1/\tau_m$) can be varied by tuning the membrane geometry (through the radius a) and tension (through the mass m, see fig. 3 in the SM). We show in fig. 2(a) how the contact time τ varies as a function of the impact velocity V for various frequencies f_m. For each value of f_m, the contact time is roughly independent of the impact velocity V, apart

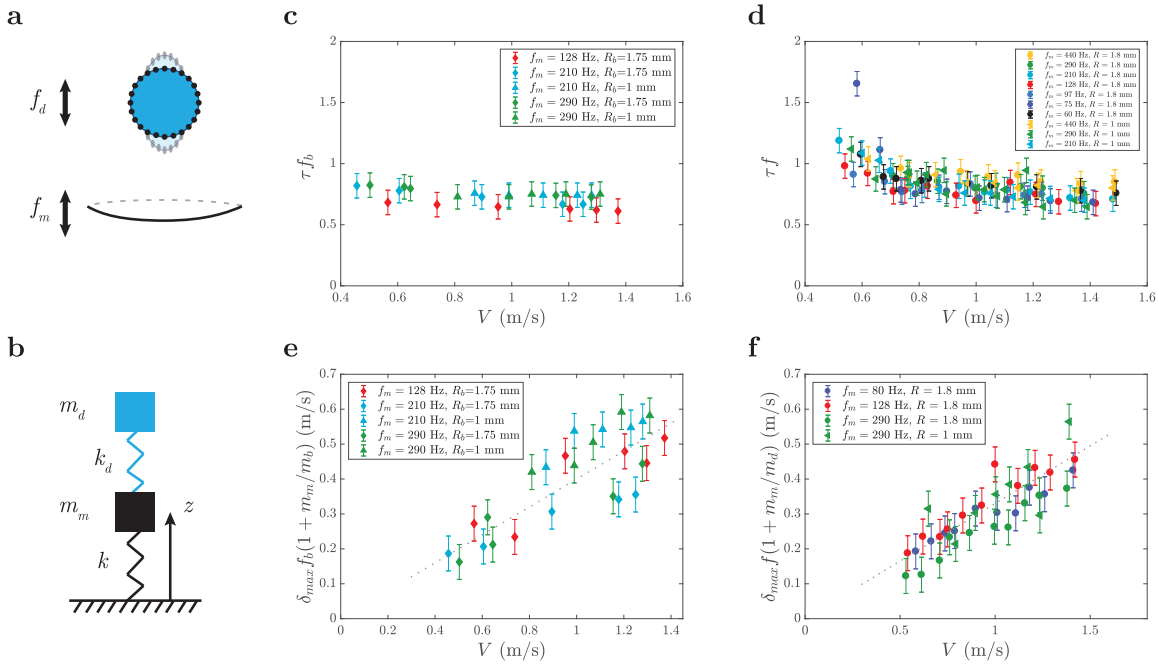

Fig. 3: (Colour online) (a) Drop and membrane are modelled as oscillators with respective frequencies f_d and f_m. (b) We assume that the system behaves as two oscillators in series during contact. (c) Normalized contact time τf_b for solid beads impacting a flexible membrane at velocity V. $f_b = \sqrt{\frac{k}{m_m+m_d}}$ is the frequency of a membrane with mass $m_d + m_m$. See the corresponding movie `Movie3_trampo.avi` in the supplemental material. (d) Normalized contact time τf for liquid marbles impacting a flexible membrane at velocity V, where $f = \sqrt{f_d f_m}$ is extracted from eq. (1). (e) Maximum deflection δ_{max} of the membrane as a function of the impact velocity V for various membrane frequencies and bead radii. Collapse of data is obtained by multiplying δ_{max} by the quantity $f_b(1+\frac{m_m}{m_b})$ as suggested in the text. (f) Same plot for marble impact. δ_{max} is now multiplied by $f(1+\frac{m_m}{m_d})$. In both cases, we observe a linear behavior, as predicted in the text. Dotted lines in (e) and (f) are linear fits with slopes 0.4 ± 0.1 and 0.33 ± 0.03, respectively.

from a weak increase at low V also observed for drops on rigid substrates [20]. More importantly, we confirm our main observation: the contact time τ on soft membranes is reduced compared to τ_0, the plateau value on a rigid substrate indicated in fig. 2(a) with a dashed line. Specifically, τ decreases as we increase the frequency of the membrane, showing the influence of the substrate on the timescale at which the liquid is repelled. The marble size R also influences the contact time, small drops being shed faster than large ones, as shown in fig. 4 in the SM where τ is observed to vary slower than $R^{3/2}$, the usual inertio-capillary behavior [21]. The response of the substrate can be characterized spatially, and we plot in fig. 2(b) the maximal deflection δ_{max} as a function of V. δ_{max} varies linearly with V, and its value is typically millimetric. When R is fixed (filled circles, $R = 1.8\,\mathrm{mm}$), there is no obvious relationship between δ_{max} and f_m. At fixed f_m ($f_m = 290\,\mathrm{Hz}$, green circles and triangles), δ_{max} increases with R, a logical consequence of the change in liquid mass.

Our aim is to understand how the liquid and the membrane cooperate in an original bouncing mechanism. Our analysis holds for $\tau_m < \tau_0$, the only regime where we expect contact time reduction. We model the solid/liquid system as coupled oscillators (fig. 3(a) and (b)). On the one hand, the marble can be viewed as a spring of stiffness k_d, mass m_d, and oscillating frequency $f_d = \frac{1}{2\pi}\sqrt{\frac{k_d}{m_d}}$. Lord Rayleigh [22] calculated the frequency of freely oscillating drops, and showed that it writes: $f_d = \sqrt{\frac{8\gamma}{3\pi m_d}}$, which provides the stiffness $k_d = \frac{32\pi}{3}\gamma$ of the spring. On the other hand, the membrane can be modelled as a spring of stiffness k, mass m_m and fundamental frequency $f_m = \frac{1}{2\pi}\sqrt{\frac{k}{m_m}}$. We assume that the droplet-membrane system behaves during contact as oscillators in series, as sketched in fig. 3(b). Then the position z of the membrane obeys a 4th-order differential equation, as derived in the SM:

$$\frac{\mathrm{d}^4 z}{\mathrm{d}t^4} + \frac{k_d}{m_m}\left(1 + \frac{m_m}{m_d} + \frac{k}{k_d}\right)\frac{\mathrm{d}^2 z}{\mathrm{d}t^2} + \frac{k k_d}{m_d m_m} z = 0. \quad (1)$$

Equation (1) has two natural limits. (1) On a rigid substrate ($k \to \infty$), it reduces to a second-order differential equation: $\ddot{z} + 4\pi^2 f_d^2 z = 0$. The contact time on rigid repellent materials is simply proportional to the Rayleigh period $1/f_d$ [21]. (2) A rigid bead ($k_d \to \infty$) hitting a flexible membrane is described by the equation $\ddot{z} + 4\pi^2 f_b^2 z = 0$, with $f_b = \frac{1}{2\pi}\sqrt{\frac{k}{m_d+m_m}}$, that is, the frequency of a membrane of stiffness k and mass m_d+m_m. We performed

experiments with polypropylene beads ($R_b = 1\,\mathrm{mm}$ and $R_b = 1.75\,\mathrm{mm}$, $\rho_b = 900\,\mathrm{kg/m^3}$) with mass m_b, and our data plotted in fig. 3(c) show that the reduced contact time τf_b collapses on a single curve, confirming that f_b is the frequency of the bead-membrane system. However, this added mass argument does not capture the contact time reduction observed for drops, as shown in fig. 5 in the SM.

As for the impact of a water drop on a flexible substrate, we can notice that eq. (1) provides two natural frequencies, that is, $f_* = \frac{1}{2\pi}(\frac{k_d}{m_m}(1 + \frac{m_m}{m_d} + \frac{k}{k_d}))^{1/2}$ and $f = \frac{1}{2\pi}(\frac{kk_d}{m_d m_m})^{1/4}$. As shown in the SM, we have $f_* > f$ whatever the values of the physical parameters, which suggests that the dynamics of the system is set by $1/f$, the longer timescale. When we rescale the contact time τ by the frequency f and plot it as a function of the impact velocity V (fig. 3(d)), data for various f_m (such as in fig. 2(a)) and various R indeed collapse. Apart from the increase observed at low V [20], contact time is found to plateau at a value $\tau \sim 0.75f$. The frequency f turns out to be the geometric mean of that of the drop and of the membrane, $f = \sqrt{f_m f_d}$, a formula capturing how the two objects conspire to generate fast bouncing. Interestingly, the frequency f scales as $R^{-3/4}$, a behavior very different from that on a rigid substrate (where it varies as $R^{-3/2}$), in agreement with our data in fig. 4 in the SM. Knowing the frequency f yields a simple prediction for δ_{max}. Before impact, the membrane is immobile and the droplet with mass m_d moves at speed V; during the first oscillation, drop and membrane both oscillate at the frequency f. Conserving the momentum provides the following scaling: $m_d V \sim (m_m + m_d)\delta_{max} f$. Figure 3(e) and 3(f) represent $\delta_{max} f_b(1 + \frac{m_m}{m_b})$ and $\delta_{max} f(1 + \frac{m_m}{m_d})$ as a function of V. For both solid and liquid marbles, we observe the predicted linear relationship, with respective slopes 0.4 ± 0.1 and 0.33 ± 0.03. These slopes are smaller than 1, suggesting that part of the initial momentum is not injected into membrane oscillations but also in membrane stretching and dissipation in air, and for liquids in internal motion.

Making impacting drops interact with soft solids modifies the outcome of collisions and can lead to enhanced repellency expressed, as by a systematic reduction of contact time. The timescale of such impacts depends on the dynamics of both membrane and drop, so that we can continuously adjust the contact time between $0.3\tau_0$ (a very low value) and τ_0, by playing on the membrane characteristics and the drop radius. It would also be interesting to model the effect of damping on the bouncing time and on the elasticity of the shocks. We expect two limiting cases: either the damping impedes the rebound or it enhances the liquid reflection. The latter case is rather counter-intuitive: for instance, glycerol will display a solid-like behavior during impact and will be repelled faster and more efficiently than water by a soft non-wetting membrane. Characterising the effect of an increasing drop viscosity would deserve a separate study.

* * *

We thank Direction Générale de l'Armement (DGA) for contributing to the financial support. We also thank Hadrien Bense, Lucie Domino and Benoît Roman for insightful comments.

REFERENCES

[1] Carré A., Gastel J.-C. and Shanahan M. E., *Nature*, **379** (1996) 432.
[2] Marchand A., Das S., Snoeijer J. H. and Andreotti B., *Phys. Rev. Lett.*, **109** (2012) 236101.
[3] Bico J., Roman B., Moulin L. and Boudaoud A., *Nature*, **432** (2004) 690.
[4] Py C., Reverdy P., Doppler L., Bico J., Roman B. and Baroud C. N., *Phys. Rev. Lett.*, **98** (2007) 156103.
[5] Style R. W., Che Y., Park S. J., Weon B. M., Je J. H., Hyland C., German G. K., Power M. P., Wilen L. A., Wettlaufer J. S. *et al.*, *Proc. Natl. Acad. Sci. U.S.A.*, **110** (2013) 12541.
[6] Karpitschka S., Pandey A., Lubbers L. A., Weijs J. H., Botto L., Das S., Andreotti B. and Snoeijer J. H., *Proc. Natl. Acad. Sci. U.S.A.*, **113** (2016) 7403.
[7] Howland C. J., Antkowiak A., Castrejón-Pita J. R., Howison S. D., Oliver J. M., Style R. W. and Castrejón-Pita A. A., *Phys. Rev. Lett.*, **117** (2016) 184502.
[8] Pepper R. E., Courbin L. and Stone H. A., *Phys. Fluids*, **20** (2008) 082103.
[9] Tagawa Y., Oudalov N., El Ghalbzouri A., Sun C. and Lohse D., *Lab Chip*, **13** (2013) 1357.
[10] Vasileiou T., Gerber J., Prautzsch J., Schutzius T. M. and Poulikakos D., *Proc. Natl. Acad. Sci. U.S.A.*, **113** (2016) 13307.
[11] Vasileiou T., Schutzius T. M. and Poulikakos D., *Langmuir*, **33** (2017) 6708.
[12] Mishchenko L., Hatton B., Bahadur V., Taylor J. A., Krupenkin T. and Aizenberg J., *ACS Nano*, **4** (2010) 7699.
[13] Shiri S. and Bird J. C., *Proc. Natl. Acad. Sci. U.S.A.*, **114** (2017) 6930.
[14] Song M., Ju J., Luo S., Han Y., Dong Z., Wang Y., Gu Z., Zhang L., Hao R. and Jiang L., *Sci. Adv.*, **3** (2017) e1602188.
[15] Bird J. C., Dhiman R., Kwon H.-M. and Varanasi K. K., *Nature*, **503** (2013) 385.
[16] Liu Y., Moevius L., Xu X., Qian T., Yeomans J. M. and Wang Z., *Nat. Phys.*, **10** (2014) 515.
[17] Weisensee P. B., Tian J., Miljkovic N. and King W. P., *Sci. Rep.*, **6** (2016) 30328.
[18] Aussillous P. and Quéré D., *Proc. R. Soc. London, Ser. A: Math., Phys. Eng. Sci.*, **462** (2006) 973.
[19] Planchette C., Lorenceau E. and Biance A.-L., *Soft Matter*, **8** (2012) 2444.
[20] Chevy F., Chepelianskii A., Quéré D. and Raphaël E., *EPL*, **100** (2012) 54002.
[21] Richard D., Clanet C. and Quéré D., *Nature*, **417** (2002) 811.
[22] Rayleigh L., *Proc. R. Soc. London*, **29** (1879) 71.