Droplet Asymmetric Bouncing on Inclined Superhydrophobic Surfaces

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ABSTRACT: A droplet impacting on inclined surfaces yields more complex outcomes than on normal impact and the effect of the inclining angle on the impact dynamics is still in controversy. Here, we show that a drop impacting on inclined superhydrophobic surfaces exhibits an asymmetric rebound with a distinctive spreading and retraction along the lateral and tangential directions. Meanwhile, there is an obvious contact time reduction with the increase of the inclining angle and impact velocity. We demonstrate that the contact time reduction is attributed to the asymmetric drop spreading and retraction, which endows a fast drop detachment. Simple analyses are presented to interpret this phenomenon, which is in a good agreement with the experimental results.

INTRODUCTION

Since the discovery of the unique properties of lotus leaves, superhydrophobic surfaces have received considerable attention owing to their distinct properties rendered by their large contact angle and small contact angle hysteresis.1−4 The complete rebound of an impacting droplet from superhydrophobic surfaces is relevant for many practical applications, including anti-icing,5−7 dropwise condensation,8,9 and antifouling.10−16 Various research studies have been conducted to reveal the effect of surface morphology on the impact outcomes.17−21 On horizontal superhydrophobic surfaces, an impinging drop first undergoes an inertia-dominant lateral spreading, then retracts on the liquid-repellent surface to minimize the surface energy, and finally takes off in the vertical direction. Normally, the drop retains a symmetrically circular shape during the whole process. The contact time, defined as the time span between the moment the drop comes into contact with the surface and the moment of detachment, keeps constant and is independent of the impact velocities.22

Note that the contact time minimization is highly desirable in various practical applications.7,14 Recent research studies have witnessed a dramatic increase in the mechanism of reducing the liquid−solid contact time. Bird et al. first introduced center-assisted recoil to accelerate the retraction process to shorten the contact time.23 During the droplet impacting on a macrotextured ridge with a diameter (a) of hundreds of micrometers, the liquid on the peak of the ridge retained a thin film and retracted as $V_{ret} = \sqrt{2\gamma/\rho(h-a)}$, where $h$ is the film thickness, and $\rho$ and $\gamma$ are the density and surface tension of the impacting droplet, respectively. The droplet retracted faster along the ridge than perpendicular to it and tended to fragment and decreased the distance and time required for recoil, resulting in an overall reduced contact time.

According to the scaling law22 of contact time $\tau \approx \sqrt{\rho_0^2/\gamma}$, splitting an impacting drop into smaller ones (a reduced drop radius $r_0$) could shorten the contact time. This is verified by introducing microstructures to promote splashing at large impact velocities.18 Similar research using asymmetric retraction to reduce the contact time was further implemented by Liu et al. and Gauthier et al.24,25 In their experiments, a spreading drop retracted faster in one direction than perpendicular to it while retaining an integrated shape.

Alternately, a superhydrophobic surface with a point-like defect endows ring bouncing, during which the spreading liquid film is punctured by the defect and the impacting drop...
adopts an unusual ring shape as it leaves the substrate.\textsuperscript{26} Upon impact, the punctured film de-wets outwards until it collides with the outer receding rim moving in the opposite direction. The collision drives the drop upwards and takes off as a ring, leading to reduced contact time. In 2014, Liu et al. made an exciting breakthrough in sculpturing superhydrophobic surfaces to promote “pancake bouncing” to shorten the contact time by \(\sim 80\%\).\textsuperscript{27} The so-called “pancake bouncing” results from the rectification of capillary energy stored in the penetrated liquid into upward motion sufficient to lift the droplet. Moreover, on truncated pyramidal post arrays with the edge length increasing continuously and linearly from top to bottom in the vertical direction, robust pancake bouncing occurred over a wide range of impact velocities.\textsuperscript{28}

There is already a wealth of knowledge about normal drop impact and rebound characteristics on a superhydrophobic surface. However, inclined impact exists in most of the practical applications and the droplet bouncing outcomes differ greatly from those on normal impact.\textsuperscript{29–35} Moreover, the contact time variation along with the inclining angle is still a controversy. Yeong et al. found that although inclined and normal impacts behaved similarly at low Weber numbers, inclined impact shows pronounced spreading asymmetry at high Weber numbers and the contact time is independent of surface inclination.\textsuperscript{34} Here, Weber number \(\left( W_e \right)\) is a dimensionless quantity, defined as \(W_e = \rho v_0^2 D_0/\gamma\), with \(v_0\) being the impacting velocity at the moment of the drop touches the substrate. On oblique impact, normal Weber number \(\left( W_{e_o} \right)\) is defined as \(W_{e_o} = \rho(v_0 \cos \alpha)^2 D_0/\gamma\), where \(\alpha\) is the surface inclining angle, \(D_0\) is the drop diameter, and \(v_0 \cos \alpha\) relates to the normal impact velocity. In contrast, Antonini et al. showed that surfaces placed obliquely promoted the drop rebound than that placed horizontally, rendering a reduced contact time by \(\sim 40\%).\textsuperscript{35} Zhang et al. achieved similar results and attributed the contact time reduction to the emptying of capillary energy stored in the penetrated liquid into the underneath microstructures, endowing an upward motion adequate to lift the drop faster.\textsuperscript{36}

In this work, we show that inclined superhydrophobic surfaces reduce the contact time via modifying the spreading and retraction process of impacting drops. During impact, the drop promotes a larger spreading in the tangential direction than that in the lateral direction, followed by a fast retraction in the lateral direction and finally leaves the surface in an elongated shape. Simple analysis was put forward to reveal the asymmetric bouncing mechanism.

\section*{Experimental Section}

In the experiment, superhydrophobic surfaces were fabricated using an electrochemical machining technique on an aluminum substrate with a size of \(40 \, \text{mm} \times 80 \, \text{mm} \times 4 \, \text{mm}\). For the sample preparation, the aluminum substrate was polished using \#800, \#1500, and \#2000 abrasive papers successively and then ultrasonically cleaned in ethanol and deionized water for 10 min, to remove the surface impurities. After drying by compressed air, the aluminum substrates as the anode and the lead plate as the cathode were put into 0.1 M sodium chloride electrolyte at 0.5 A/cm\(^2\) current density for 6 min. The as-fabricated substrate was then thoroughly rinsed with deionized water, followed by drying in a nitrogen stream. Finally, the substrate was immersed in 0.5 mM \(\eta\)-hexane solution of trichloro-\((1H,1H,2H,2H\text{-perfluoroctyl})\)silane for \(\sim 50\) min, followed by heat treatment at 150 °C in air for \(\sim 30\) min to render the surface superhydrophobic.

As evidenced in the scanning electron micrography (SEM) images in Figure 1a,b, the surface is uniformly coated by loosely clumped textures (Figure 1a) with a rugged block roughness (Figure 1b) of a size of \(\sim 2\) \(\mu\)m. By averaging five individual measurements, the advancing and receding contact angles (\(\sim 4\) \(\mu\)L, with a diameter of 1.97 mm) on the modified surface were measured to be 158.5\(^\circ\) ± 1.0\(^\circ\) and 154.3\(^\circ\) ± 1.0\(^\circ\), respectively (Figure 1c), using an OCA25 Standard Contact Angle Goniometer, suggesting that the complex structures required for excellent superhydrophobicity have been obtained by electrochemical etching. Droplet impact experiments were conducted in an ambient environment, at room temperature with 60% relative humidity. Deionized water droplets of \(\sim 12\) \(\mu\)L (\(D_0 = 2.85\) mm) were released from a syringe pump equipped with a fine needle at a flow rate of 2 \(\mu\)L/s. The distance of the needle above the surface was adjusted to change the initial impact velocity. Two synchronized high-speed cameras (Photon SA4) were used to capture the droplet impacting dynamics both from the side and normal views (the shooting direction is parallel with the normal direction as shown in Figure 1d) at a frame rate of 10 000 fps. The dynamic change of impacting drops was analyzed using Imagej software (Version 1.46, National Institutes of Health).

\section*{Results and Discussion}

Figure 2a shows the image sequences of droplets impinging on the horizontal superhydrophobic surface at \(W_e = 37\) (Video S1, the first row). The impacting drop holds a circular symmetry during the whole spreading and retraction process and finally bounces off the surface at \(\sim 17.2\) ms (\(= 2.7\sqrt{\rho_0^3/\gamma}\)), which is consistent with previous research.\textsuperscript{22} However, the impacting drop exhibits an asymmetric bouncing dynamics on tilting surfaces (see Figure 1d for the clarification of directions). For example, on the superhydrophobic surface with an inclining angle of \(\alpha = 60\text{°}\), as exhibited in Figure 2b, the impacting drop initially spreads isotropically at \(W_e = 37\), but the spreading becomes anisotropic as the drop starts to retract. Specifically, when the liquid in the lateral direction started to retract at \(\sim 3.9\) ms, the liquid in the tangential direction continues to
spread. Once the liquid in the lateral direction has fully contracted at \( \sim 12.6 \text{ ms} \), the drop leaves the surface in an elongated shape along the tangential direction (Video S1, the second row). The contact time is reduced by \( \sim 30\% \) compared to that on the nontilting surface.

Figure 3 plots the variation of the contact time with Weber numbers at different surface inclining angles. It generally shows that the contact time decreases with the increase of the inclining angle and Weber number. To better understand the dependence of contact time on the inclining angle, we decouple the contact time into spreading time \( (t_s) \) and retraction time \( (t_r) \), as displayed in Figure 3b. It is obvious that the spreading time is almost independent of the inclining angle. However, the retraction time decreases strongly as the inclining angle increases.

To interpret the effect of the inclining angle on contact time reduction, we first consider the effect of the inclining angle on the droplet spreading process. A droplet in the oblique impact is subject to a combination of downward motion along the tilting surface with an impinging velocity of \( v_0 \sin \alpha \) together with motion normal to the surface with an impinging velocity of \( v_0 \cos \alpha \). A droplet impacting on the tilting surface process a tangential component of momentum that enables the drop to spread larger along the tangential direction. Figure 4a shows the nondimensionalized drop spreading diameter \( (D^*) \) as a function of time in the lateral and tangential directions at \( We_n = 37 \) and surface tilting angle \( \alpha = 60^\circ \). To quantify the spreading asymmetry, we define \( Q \) as the ratio of the maximum values of the drop spreading diameters in the tangential and lateral directions. Figure 4b shows the variation of \( Q \) as a function of the inclining angle at different \( We_n \). It is obvious that a large inclining angle at high Weber number leads to a

Figure 2. (a) Selected snapshots showing a drop \((D_0 = 2.85 \text{ mm})\) impacting on a horizontal superhydrophobic surface at \( We_n = 37 \) from both the side and normal views. The impacting drop holds a circular symmetry during the whole spreading and retraction process and finally bounces off the surface at \( \sim 17.2 \text{ ms} \). (b) Selected snapshots showing a drop \((D_0 = 2.85 \text{ mm})\) impacting on the inclined surface \((\alpha = 60^\circ)\) at \( We_n = 37 \) from both the side and normal views. When the liquid in the lateral direction started to retract at \( \sim 3.9 \text{ ms} \), the liquid in the tangential direction continues to spread; once the liquid in the lateral direction has fully contracted at \( \sim 12.6 \text{ ms} \). More details are shown in Video S1.

Figure 3. (a) Variation of the contact time with surface inclining angle at different normal Weber numbers \( We_n \). (b) Spreading time \( t_s \) (left) and retraction time \( t_r \) (right) as a function of \( We_n \) at different inclining angles. Open symbols indicate \( t_s \) and closed symbols indicate \( t_r \).
pronounced spreading asymmetry. Note that in normal impact ($\alpha = 0^\circ$), $Q$ is equal to unity, suggesting that the asymmetric spreading is regulated by the surface inclining angle.

A simple hydrodynamic analysis is employed to explain the contact time reduction with the droplet deformation on inclined superhydrophobic surfaces. Note that drop spreading on superhydrophobic surfaces is mainly governed by inertia, only retraction is considered here. After the drop spreads to its maximal extension, the liquid film subsequently de-wets the surface. The retraction force is primarily driven by a reduction of surface energy, which forms a rim to collect the liquid and move inwards. On a nontilting surface, the impacting drop retracts in a circular symmetric shape and the surface energy of the central film of radius $r$ can be expressed as $E_s \approx \pi r^2 \gamma (1 - \cos \theta)$, where $r$ is the radius of the liquid film and $\theta$ is the surface apparent contact angle. On inclined surfaces, however, owing to the tangential component of impacting velocity, the central film can be approximated as an ellipse with a short axis $x$ in the lateral direction and a long axis $y$ in the tangential direction, as shown in Figure 2b. The surface energy of the elliptical liquid film can be approximated as $E = \pi xy \gamma (1 - \cos \theta)$. Therefore, the retraction force in the lateral direction becomes $F = \partial E / \partial x \approx \pi xy \gamma (1 - \cos \theta)$.

As the viscous effect can be neglected because the Ohnesorge number in the experiments is $\sim 0.002$ which is far less than 1, the retraction dynamics is dominated by a competition between the surface tension of the central film and the inertia of the rim. The momentum conservation for the liquid film in the lateral direction can be written as $\frac{d}{dt} \left( m \frac{dx}{dt} \right) = F$, where $m$ is the mass of the rim. As the major $y$ changes little as the lateral retraction starts, we can get $\frac{dm}{dt} = \pi y h \frac{dx}{dt}$ with $h$ being the film thickness. Then the retraction speed is $V_x = \frac{dx}{dt} \approx \sqrt{\gamma (1 - \cos \theta) / ph}$. Clanet et al. have proposed a widely used scaling model to predict the maximum spreading diameter $D_{\text{max}}$ as $D_{\text{max}} \approx 0.9D_0 W_e^{0.25}$. However, the relationship between $D_{\text{max}}$ and $W_e$ tends to slightly vary among different studies depending on the surface tested. Here, we measured the maximum spreading diameter of drops impacting on the horizontal superhydrophobic surface and obtained the scaling of $D_{\text{max}} \approx$...
0.79DxWeα0.29 through the regression fitting, as shown in Figure 5a. Yeong et al. have built a theoretical model for the maximum spreading in the tangential direction as \( D_{\text{max}} = D_{0}(0.79\rho \gamma \cos \alpha)^{0.28} + 0.0033 \tan^2 \alpha \). Following this model, we determined the value of the scaling factor \( \epsilon = 0.0033 \) using the experimental data in the range of 0–45°. To verify the scaling factor \( \epsilon \), droplet impact experiments on the tilting surface with \( \alpha = 60^\circ \) was conducted, as displayed by the open markers in Figure 5b, suggesting that the model by Yeong et al. can be used to describe the relationships among \( D_{\text{max}} \), \( We_\alpha \), and \( \alpha \) in our experiments. Finally, we obtained the maximum spreading diameter in the tangential direction as \( D_{\text{max}} = D_{0}(0.79\rho \gamma \cos \alpha)^{0.28} + 0.0033 \tan^2 \alpha \). In the lateral direction, the droplet maximum spreading is governed by inertia, which is close to that on the horizontal impact and confirmed in the previous research, following the scaling of \( D_{\text{max}} \approx D_{\text{max}} \approx 0.79DxWeα0.29 \). According to volume conservation, the film thickness is determined as \( h = D_0/6D_{\text{max}}D_{\text{max}} \). Hence, the retraction time in the lateral direction is \( t_r = D_{\text{max}}/2V \approx f (\rho \gamma \cos \alpha)/\gamma \), where \( f \) is a function of \( We_\alpha \) and the inclination \( \alpha \), expressed as \( f = (0.79 + 0.0033 \tan^2 \alpha \rho \gamma)^{0.28} \), which decreases as \( We_\alpha \) (5 < \( We_\alpha \) < 70) and \( \alpha \) (0 < \( \alpha < 90^\circ \)) increase. Meanwhile, the model shows a good agreement with the experimental data (Figure S1). Therefore, the contact time is reduced on the inclined surfaces with a large Weber number or a large inclination, as evidenced in Figure 5a. This might be explained by a much more prominent asymmetric spreading at large Weber numbers or a large inclination, which endows a fast retraction and a shortened contact time.

**CONCLUSIONS**

In this study, we systematically investigate the droplet impact dynamics on inclined superhydrophobic surfaces. During impact, after the drop spreads to its maximum extension in the lateral direction, it continues to spread in the tangential direction. However, the drop retraction in the lateral direction is much faster than that in the tangential direction. At the end of retraction in the lateral direction, the drop detaches from the surface directly in an asymmetric shape, endowing a shortened contact time. We show that the contact time decreases with the increase of impact velocity and inclining angle. A simple theoretical model is constructed to explain this phenomenon. We envision that the asymmetric bouncing on inclined surfaces not only extends our fundamental understanding of drop impact dynamics but also provides a reference for design superhydrophobic surfaces for various applications.

**ASSOCIATED CONTENT**

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acsomega.9b01348.

Comparison of the retraction time \( t_r \) as a function of \( f \) between experimental measurement and theoretical prediction (PDF)

Water drop impact dynamics on the horizontal (the first row) and inclined (the second row) superhydrophobic surfaces at \( We_\alpha = 37 \) from both the side and normal views (AVI)

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**Notes**

The authors declare no competing financial interest.

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