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Contactless prompt tumbling-rebound of drops from a sublimating slope

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22 **ABSTRACT**

23 *We have uncovered a drop rebound regime, characteristic of highly viscous liquids impacting onto tilted*
24 *sublimating surfaces. Here the drops rather than showing a “slide, spread, recoil and rebound” behavior,*
25 *exhibit a “prompt tumbling-rebound”. As a result, glycerol surprisingly rebounds faster than three orders of*
26 *magnitude less viscous water. When a viscous drop impacts on a sublimating surface, part of its initial linear*
27 *momentum is converted into angular momentum: Lattice Boltzmann simulations confirmed that tumbling*
28 *owes its appearance to the rapid transition of the internal angular velocity prior to rebound to a constant*
29 *value, as in a tumbling solid body.*

30 **I INTRODUCTION**

31 Despite its illusory simplicity, the interaction between a liquid drop and a solid surface during impact is a
32 fascinating fluidics problem, combining a variety of phenomena at multiple temporal and spatial scales [1–
33 5]. These include splash [6–10], phase-change induced surface levitation [11–15], skating on a film of
34 trapped air [16–18] and rebounding [19–22]. Recently [11], it was demonstrated that drops can rebound
35 after impact on an extremely cold solid carbon dioxide surface (at -79°C , well below the limit of even
36 homogeneous nucleation of water), because of the formation of a sublimated vapor layer acting both as
37 impact cushion and thermal insulator, enabling drops to hover and rebound without freezing. A sublimating
38 surface is different from aerodynamically assisted surface levitation [23–25] and from the Leidenfrost
39 effect [12–14,26–28], in the sense that it is independent from liquid properties, such as boiling
40 temperature, and there is no loss of drop mass due to its own boiling (as in the Leidenfrost phenomenon).
41 Of course, in both cases an intervening layer is generated between the drop and the substrate. Sublimating
42 surfaces can thus be used to study the contactless interaction of virtually any liquid, such as the highly
43 viscous liquids used here. Also, they enable the study of phenomena expected from a superhydrophobic
44 surface (SHS) with extreme performance [29] (very high contact angles and very low hysteresis), providing
45 further motivation for the fabrication and subsequent study of such surfaces. In the present fundamental
46 study, we demonstrate and explain the existence of a new “prompt tumbling-rebound” mechanism, in

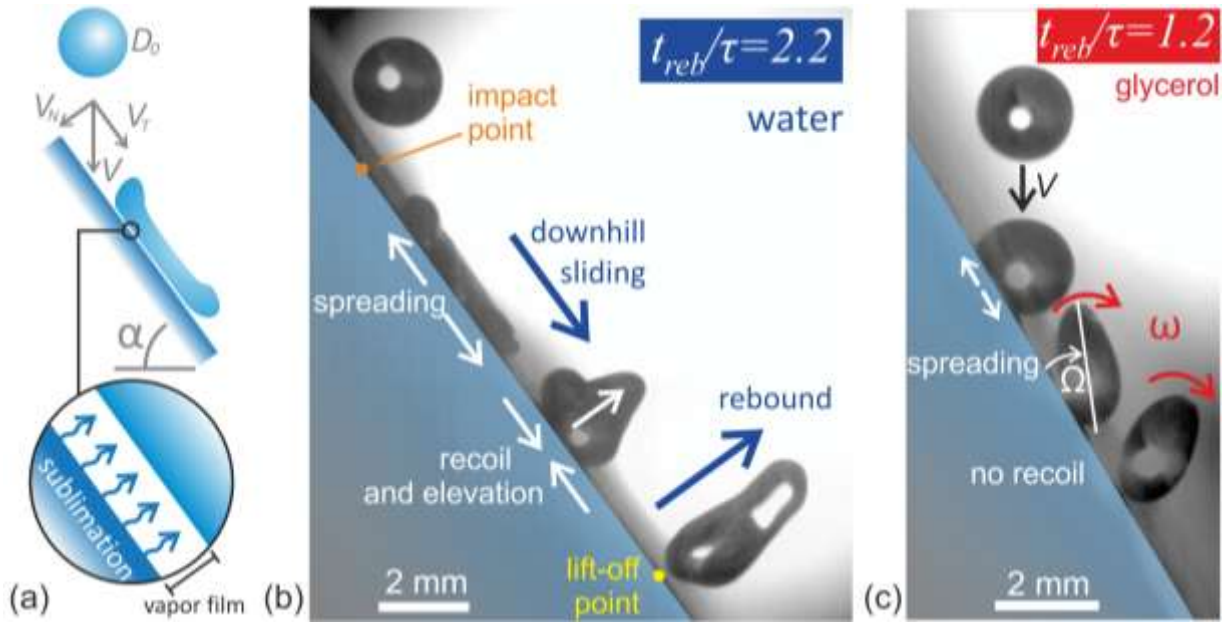
47 which a small conversion of translational to rotational kinetic energy, at non-axisymmetric impact
48 conditions, promotes fast drop rebound despite high viscosities.

49 By focusing on non-axisymmetric impact conditions at increasing viscosity, we observe a transition from an
50 expected “slide, spread, recoil and rebound” mechanism (see Figure 1a and b, and Video 1 in the
51 Supplemental Material) to a “tumbling” behavior, enabling the prompt rebound of highly viscous liquids
52 such as glycerol (see Figure 1c and Videos 2 and 3 in the Supplemental Material). Here, after an initial,
53 viscosity-limited and impact-driven deformation phase, the drops simply tumble off the surface,
54 rebounding faster than three orders of magnitude less viscous (water) drops. As confirmed by 3D numerical
55 simulations based on the entropic lattice Boltzmann method (ELBM, see Video 4 in the Supplemental
56 Material), the behavior at high viscosities is aided by the contactless nature of the impact and was not
57 observed on classical superhydrophobic surfaces, where an increase of viscosity can protract the rebound
58 time [30], or even prevent rebound. In experiments on micropillar-based tilted surfaces, we observed that
59 viscous drops stay initially pinned at the impact point and eventually roll down the surface at a speed of
60 approximately 10^{-2} m/s (see section SM1 for details on superhydrophobic surface preparation and Video 5
61 in the Supplemental Material showing viscous drop behavior). **In addition, drop pinning may occur on
62 textured superhydrophobic surfaces as a result of drop impalement at high impact speed [22,30,31]:
63 impalement is not an issue sublimating surfaces.**

64 A representative schematic of a plausible scenario of contactless drop impact on a tilted sublimating
65 surface is shown in Figure 1a: after impact, the drop slides down the slope, and simultaneously spreads.
66 The presence of the CO₂ vapor layer due to substrate sublimation significantly reduces friction between the
67 drop and the solid substrate, playing a similar role as that of the lubricating melted liquid water layer in ice
68 skating [32]. The drop subsequently bounces off the surface after a certain rebound time, t_{reb} , defined as
69 the time lag the impact and the lift-off. The corresponding downhill distance travelled by the drop, L_{slide} ,
70 was measured from the impact point to the lift-off point (see Figure 1b).

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74 **Figure 1:** Drop impact onto a tilted carbon dioxide sublimating surface: schematic (a) and image sequence of water (b)
 75 and glycerol (c) drop impacts. Impact conditions are: $\alpha = 55^\circ$, $We = \rho V^2 D_0 / \sigma = 86$,
 76 $Oh = \mu / \sqrt{\rho \sigma D_0} = 2.7 \cdot 10^{-3}$ for water, and $\alpha = 60^\circ$, $We = 121$, $Oh = 3.2$ for glycerol. Image sequence was
 77 obtained by overlapping of 4 images: see Video 1 and 2 in the Supplemental Material for full sequence. The non-
 78 dimensional rebound time is $t_{reb}/\tau = 2.2$ for water, and $t_{reb}/\tau = 1.2$ for glycerol. Also indicated are the normal,
 79 V_N , and tangential, V_T , components of impact velocity, as well as the substrate tilt angle, α .

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II METHODS AND MATERIALS

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A Experimental tests

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Drop impact studies were performed at room temperature ($T \approx 23^\circ\text{C}$) on a carbon dioxide disk at -79°C ,
 corresponding to the CO_2 sublimation temperature at 1 atm. The surface was first kept horizontal to study
 normal impacts as a basis for comparison, and was subsequently tilted up to an angle of 75° to study
 oblique impacts. Most of the experiments were conducted using water, glycerol, and water-glycerol
 mixtures, spanning over three orders of magnitude of viscosity (from ~ 1 to $\sim 10^3$ mPas), and surface tension
 $63 < \sigma < 72$ mN/m. Additional normal impact experiments were performed with glycols and silicon oils,
 with surface tension down to 20 mN/m (see details in section SM2 in the Supplemental Material). The drop
 impact velocities and drop diameters were $0.8 < V < 3.2$ m/s and $1.3 < D_0 < 2.1$ mm, respectively. The

91 corresponding non-dimensional number ranges were for the Weber number, $20 < We = \rho V^2 D_0 / \sigma < 610$,
92 for the Reynolds number, $1.45 < Re = \rho V D_0 / \mu < 5490$, and for the Ohnesorge number,
93 $2.7 \cdot 10^{-3} < Oh = \mu / \sqrt{\rho \sigma D_0} < 4.92$.

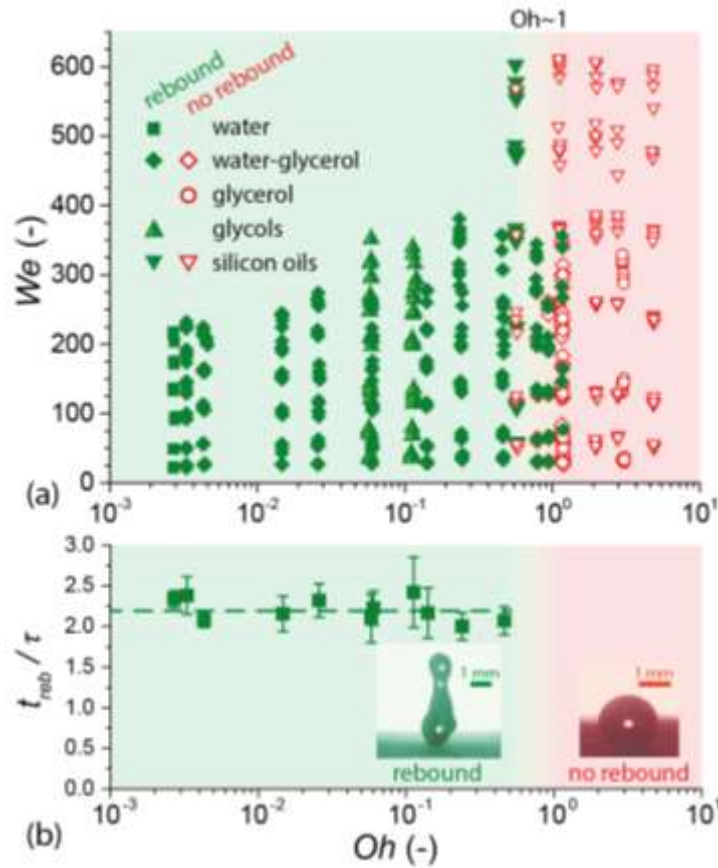
94 B Numerical simulations

95 To provide insight for the appearance of tumbling, we used the ELBM modeling approach [33], employing
96 the Navier-Stokes equations for a two-phase fluid, where a van der Waals-type equation of state and
97 Korteweg's stresses are implemented in the kinetic lattice Boltzmann setting of discrete velocity
98 populations [34–36]. The impacting liquid was modeled as a drop on a superhydrophobic surface [37] with
99 the contact angle $\theta = 180^\circ$ and partial slip at the wall – see Ref. [38] for details on application of boundary
100 conditions for the lattice Boltzmann populations at the wall. Partial slip at the wall was imposed using the
101 slip coefficient, k , obtained from the experimental measurements, presented below. The tangential
102 velocity at the wall nodes was made equal to kV_t' , where V_t' is the tangential velocity at the neighboring
103 node within the fluid, in the direction perpendicular to wall, thus locally enforcing the partial slip observed
104 in the experiments. This procedure helps us circumvent explicit modeling of the sub-micron gas layer
105 trapped between the drop and the substrate. The validity of the numerical simulations is confirmed *a*
106 *posteriori* from the good prediction of the rebound time, angular velocity and drop shape, in comparison to
107 the experiments.

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109 **III RESULTS AND DISCUSSION**110 **A Normal axisymmetric impacts**

111 The identification of a transition in the drop dynamics at high viscosity, i.e. high Oh , can be understood by
 112 first looking at the behavior of drops during normal impact on horizontal surfaces (Figure 2) and then
 113 oblique impact on tilted surfaces (Figure 3 and Figure 4Figure 4). Figure 2a illustrates the rebound/no-
 114 rebound behavior on the $We - Oh$ plane, for normal drop impact on a horizontal sublimating substrate. It
 115 is found that a transition from rebound to no-rebound occurs at $Oh \approx 1$, with low viscosity drops always
 116 rebounding for $Oh < 0.6$, and high viscosity drops unable to lift-off for $Oh > 1.2$. In the in-between
 117 transition regime, either outcome is possible. The transition regime at $Oh \approx 1$ can be explained by the fact
 118 that in this range the viscous effects become of the same order as the surface energy effects, and cause a
 119 rapid dissipation of the initial kinetic energy of the impacting drop: viscous effects thus inhibit the
 120 conversion of kinetic energy into surface potential energy, and back to kinetic energy, as typically occurs for
 121 relatively low viscosity liquids, such as water, and prohibit rebound. As shown in the inset picture in Figure
 122 2b, at high Oh the highly viscous drops bead up to a quasi-spherical shape at rest, and eventually roll
 123 away at the slightest perturbation, because of the absence of wetting and lateral adhesion forces due to
 124 the sublimating substrate. In the rebound regime, the rebound time follows the conventional scaling
 125 $t_{reb}/\tau = a = \text{const}$, where $\tau = (\rho D_0^3 / 8\sigma)^{0.5}$ [19] and $a = 2.2 \pm 0.2$ [20], with a being constant and
 126 independent of the Ohnesorge number (see also more details in Section SM3 in the Supplemental
 127 material).



128

129 **Figure 2:** (a) Rebound/no rebound map for normal drop impact on a horizontal sublimating surface (603 impact tests
 130 with different liquids - see legend). (b) Non-dimensional rebound time, t_{reb}/τ , as function of the Ohnesorge number,
 131 Oh .

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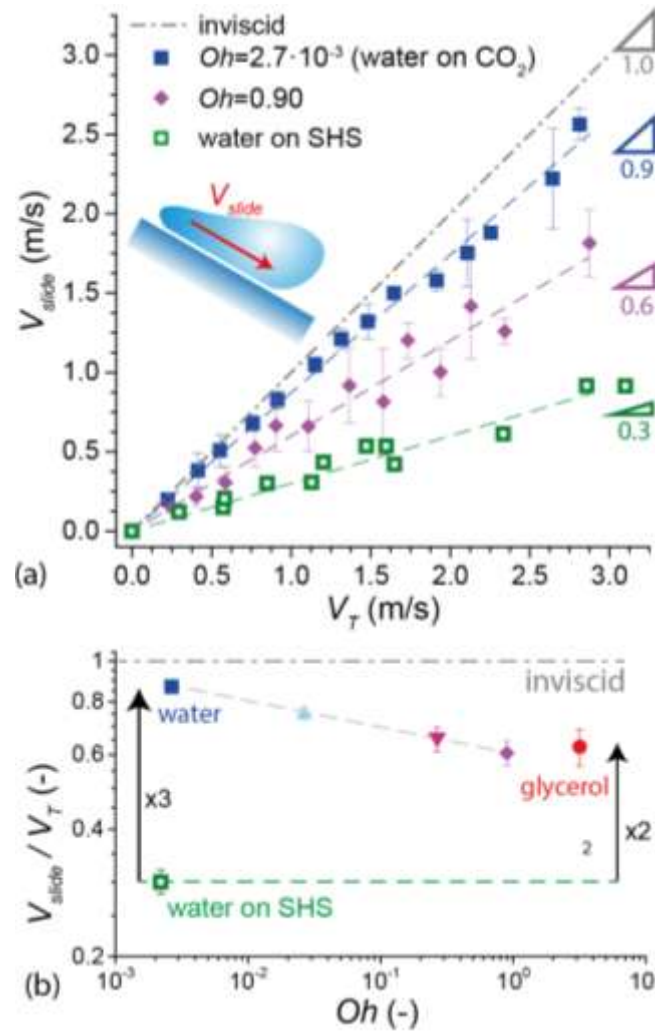
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134 B Oblique non-axisymmetric impacts

135 For non-axisymmetric impacts on tilted sublimating surface, however, experiments reveal a qualitative
 136 change of the rebound pattern, showing that highly viscous glycerol drops can also rebound, and do this
 137 even faster than water drops. The characteristic sliding velocity of the drop, V_{slide} , computed as the ratio
 138 L_{slide}/t_{reb} , is plotted in Figure 3a as function of the tangential component of the impact velocity, V_T . In the
 139 ideal inviscid case, assuming zero lateral adhesion forces [39,40] to cause drop deceleration and negligible
 140 acceleration due to gravity, the drop will continue to travel on the substrate at $V_{slide} \approx V_T$. A linear scaling

141 $V_{slide} = kV_T$ holds for all tested liquids, with k depending on the Ohnesorge number (see Figure 3b). For a
 142 millimetric water drop ($Oh = 2.7 \cdot 10^{-3}$), $k = 0.9$, remarkably close to the ideal inviscid case, $k = 1$, and
 143 three-fold higher than for water drops impacting on a superhydrophobic surface ($k = 0.3$, as reported
 144 previously [17]). The enhanced sliding on a sublimating surface, even with respect to a superhydrophobic
 145 surface, is a consequence of the contactless regime, in which lateral adhesion forces are absent. The value
 146 of k decreases with increasing Ohnesorge number (see Figure 3b), and subsequently plateaus remaining
 147 approximately constant at the value of $k \approx 0.6$ for $Oh > 1$, as shown by experiments. Values of $k < 1$
 148 denote that frictional losses in the vapor layer trapped between the drop and the substrate, despite being
 149 smaller than on superhydrophobic surfaces, are not negligible. As confirmed by the entropic lattice
 150 Boltzmann method, viscous losses occur mainly in the first (spreading) phase of the impact, over a
 151 timescale t_{fr} , as a result of the drop rapid deformation and wall friction. Thus, the friction force can be
 152 estimated as $F_{fr} \approx m(V_T - V_{slide})/t_{fr} = mV_T(1 - k)/t_{fr}$, which we will use below to estimate the drop
 153 angular velocity during tumbling. **A thorough understanding of the dependence of k on the Ohnesorge**
 154 **number, beyond the clear trend shown by the experimental data, would require the accurate**
 155 **reconstruction of the vapor layer flow at the liquid-solid interface through numerical simulations, a**
 156 **challenge that goes beyond the goals of the present study, and can certainly represent a motivation for**
 157 **future works.**

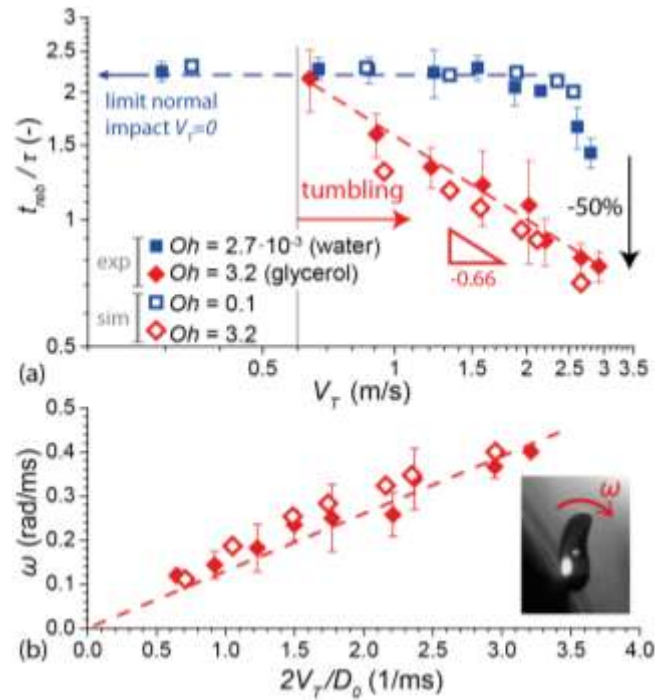
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160 **Figure 3:** (a) Drop average sliding velocity, V_{slide} , as function of impact tangential velocity, V_T . V_{slide} is calculated as
 161 the ratio L_{slide}/t_{reb} . Legend reports values of the corresponding non-dimensional Ohnesorge number, Oh . (b) Ratio
 162 $k = V_{slide}/V_T$ as function of Oh . Symbol legend: experimental data for impacts on the sublimating substrate (■
 163 water, ▲ water/glycerol 40/60, ▼ water/glycerol 15/85, ◆ water/glycerol 7/93, ● glycerol); □ water drop impact on a
 164 superhydrophobic surface (SHS, data from [41]).

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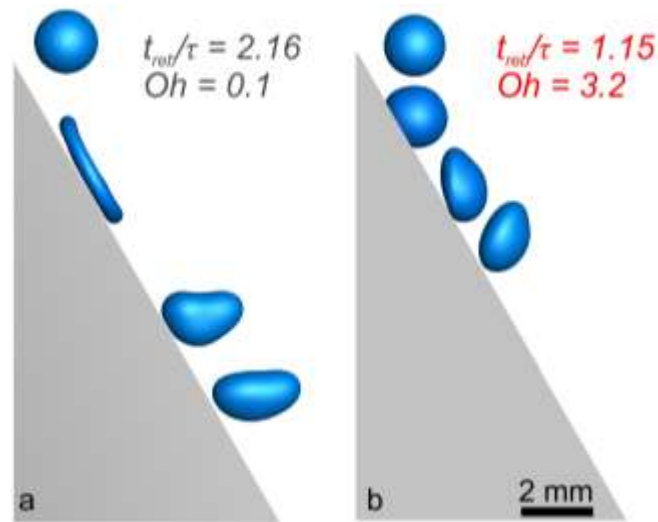
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167 **Figure 4:** (a) The non-dimensional rebound time, t_{reb}/τ , as function of the tangential velocity, V_T : both
 168 experimental data for water and glycerol and numerical results are included. Legend reports values of Oh . (b) The
 169 angular velocity, ω , of highly viscous glycerol drop scales linearly with $2V_T/D_0$. Solid symbols: experiments; open
 170 symbols: simulations. The fitting line is $\omega = 0.13 (2V_T/D_0)$.

171

172 Figure 4a shows the variation of the rebound time as function of the tangential velocity for water and
 173 glycerol. Unsurprisingly, for water ($Oh = 2.7 \cdot 10^{-3}$), the rebound time remains constant for a wide range of
 174 tangential impact velocity, up to $V_T \approx 2$ m/s, and $t_{reb}/\tau = 2.2$ still holds, meaning that the spreading and
 175 recoiling process of the drop is not affected by the simultaneous downward sliding. In other words, the
 176 usual picture of the inertia-capillarity interplay during the conventional rebound still holds and the viscosity
 177 (if low) does not play a prominent role. Only for $V_T > 2$ m/s, the impact deviates from the classical
 178 axisymmetric behavior: as demonstrated recently by Bird *et al.* [20] in the context of engineered
 179 superhydrophobic surfaces, the non-axisymmetric spreading and recoiling (see Video 7 in the Supplemental
 180 Material) can lead to a reduction of drop rebound time down to $t_{reb}/\tau \approx 1.5$ for the maximum tested
 181 tangential velocity

182 ($V_T \approx 3 \text{ m/s}$). A comparison of the drop shape evolution resulting from ELBM simulations between the
 183 low-viscosity regime ($Oh = 0.1$, see Figure 5a and corresponding Video 8 in the Supplemental Material)
 184 and the high-viscosity regime ($Oh = 3.2$), highlights the same trend and confirms the limited role of
 185 viscosity on drop dynamics in the regime $Oh < 1$.



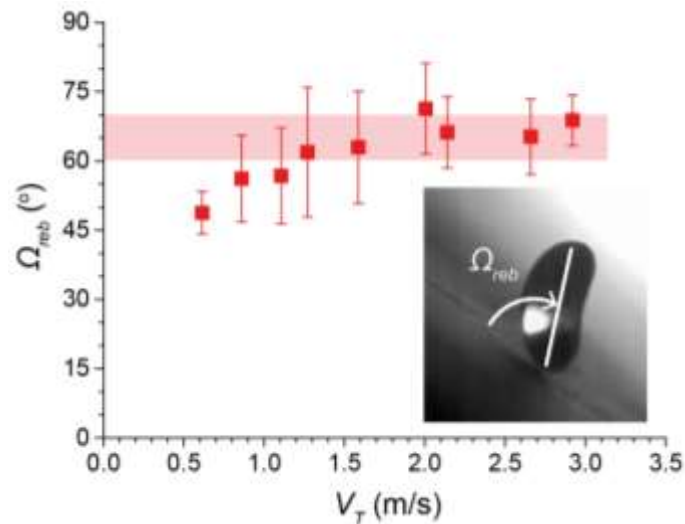
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 187 **Figure 5:** Evolution of simulated drop impacting on a sublimating slope: (a) a drop in the low viscosity regime
 188 ($Oh = 0.1$, $We = 121$, $\alpha = 60^\circ$), and (b) a drop in the high viscosity regime ($Oh = 3.2$, $We = 121$, $\alpha = 60^\circ$).
 189 The rebound time in the simulation was identified through matching the experimentally observed orientation of the
 190 drop at the rebound with the corresponding simulated image.

191

192 However, for highly viscous glycerol drops ($Oh = 3.2$) a fundamentally different rebound mechanism is
 193 identified, as highlighted previously in Figure 1c. The drop rebound starts at $V_T \approx 0.6 \text{ m/s}$ with a rebound
 194 time of $t_{reb}/\tau \approx 2.2$ (see Figure 4a), similar to that of water at the same V_T , and is significantly reduces
 195 down to a minimum of $t_{reb}/\tau \approx 0.7$ at the highest tested tangential velocity, $V_T = 3 \text{ m/s}$. The glycerol
 196 rebound time is thus half that of water above $V_T = 3 \text{ m/s}$, despite the fact that glycerol drops do not
 197 rebound for $V_T < 0.6 \text{ m/s}$ and even with the three orders of magnitude higher viscosity of glycerol
 198 compared to water (Figure 4a).

199 The reason for faster rebound at high viscosity is that, after some spreading and limited sliding, the drop
 200 tumbles off without recoiling. The drop rapidly detaches from the surface by rotating almost as a rigid
 201 body. The transition to a different rebound regime at high viscosities can be understood by comparing the
 202 drop relaxation time, $t_{rel} \propto \mu D_0 / 2\sigma$ [42], to the characteristic oscillation time, τ , whose ratio is
 203 proportional to Oh . Indeed, at high viscosities, when $t_{rel} / \tau \sim Oh > 1$, the longer t_{rel} delays drop recoiling
 204 and leads to sustaining the drop rotational energy during tumbling. Also, the distance travelled by the drop
 205 through sliding, $L_{slide} = V_{slide} t_{reb}$, is significantly reduced for high Oh , since both V_{slide} and t_{reb} are
 206 reduced with increasing Oh , as shown in Figure 3 and Figure 4, respectively. Figure 4b shows that the
 207 average drop angular (spinning) velocity, ω , scales linearly with the ratio $2V_T / D_0$ (see section SM5 in the
 208 Supplemental Material for more details). The average angular velocity, ω , was defined and measured as
 209 the ratio of the angle formed by the major axis of the flattened drop and the substrate at the moment of
 210 drop lift-off (Ω_{reb}), and the rebound time, t_{reb} . Indeed, since the balance of the drop angular momentum
 211 gives $F_{fr} D_0 / 2t_{fr} \approx I\omega / t_{fr}$, on the basis of the above estimation for F_{fr} , we obtain $\omega \approx (1-k)(2V_T / D_0)$,
 212 confirming the linear correlation. Figure 6 shows the value of Ω_{reb} as function of the impact tangential
 213 velocity, V_T : Ω_{reb} is practically constant in the range $\sim 60-70^\circ$ for $V_T > 1.3\text{m/s}$. As such, an increase of
 214 angular velocity, ω , corresponding to a faster spinning, is responsible for a significant reduction of the
 215 rebound time, t_{reb} , for very viscous liquids with $Oh > 1$ (glycerol, $Oh = 3.2$), compared to the three orders
 216 of magnitude less viscous water.

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219 **Figure 6:** Values of the drop tilting angle at the moment of rebound, Ω_{reb} , as function of tangential component of
 220 velocity, V_T , for a highly viscous glycerol drop ($Oh=3.2$).

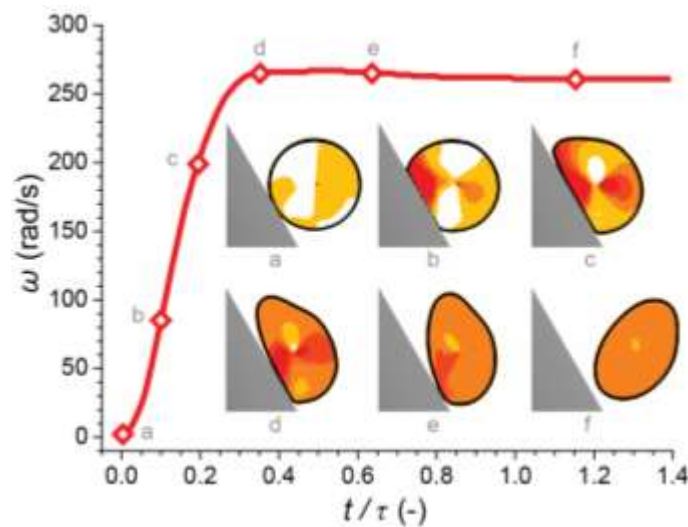
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222 The occurrence of tumbling can be better understood within the framework of vorticity generation from a
 223 boundary (shear) layer. Injection of vorticity through a shear layer was studied, in particular by [43] in a
 224 direct numerical simulation of a drop normal impact onto a flat surface. Transition to tumbling under shear
 225 is a common scenario also away from boundaries [44]. In the present experiment, the tilted slope provides
 226 an off-center impact condition and thus can cause generation of the angular momentum. Hence, when a
 227 viscous drop impacts on the sublimating surface, the linear momentum of the drop is partially converted
 228 into angular momentum, providing the drop with a spin that facilitates take off causing tumbling. Note that
 229 the energy associated with the rotation, $R \sim mD_0^2\omega^2/8$ is small compared to the tangential component of
 230 translational kinetic energy $K \sim mV_T^2/2$. Since $\omega = 0.13 (2V_T/D_0)$ (Figure 4b), then $R/K \sim 10^{-2}$; that
 231 is, only about one percent of the drop initial kinetic energy is converted into rotational energy.

232 C Vorticity generation and evolution

233 To better identify the origin of the tumbling effect, the local angular velocity was calculated from the
 234 numerical simulations as $\vec{\omega}_{loc} = |\vec{r} \times \vec{v}|/r^2$, where \vec{r} is the position vector relative to the center-of-mass of

235 the drop, and \vec{v} is the relative fluid velocity at that location. The component of the vector $\vec{\omega}_{loc}$, orthogonal
 236 to the plane of symmetry of the impacting drop, is overwhelmingly dominant, i.e. two orders of magnitude
 237 larger than the other two orthogonal components, since the rotation axis is essentially perpendicular to the
 238 plane of symmetry. From this dominant component, the average instantaneous angular velocity over the
 239 drop volume, $\omega(t)$, was computed: its evolution is illustrated in Figure 7 (see also corresponding Video 9),
 240 together with the characteristic snapshots of the local angular velocity spatial distribution on the symmetry
 241 plane; ω rapidly reaches a maximum value well before rebounding, with the rotational motion initiated
 242 near the impact zone and then becoming uniform. Thereafter, ω remains practically constant through the
 243 liquid, with the drop exhibiting the behavior of a solid ready to tumble. Put differently, the oblique impact
 244 breaks the axisymmetry of the drop at the beginning of the sliding, and the gain in angular momentum is
 245 thus initiated by the off-center flow reversal. If the relaxation time is large enough ($Oh > 1$), the angular
 246 momentum diffuses through the entire drop before it can recoil, and the tumbling takes place.

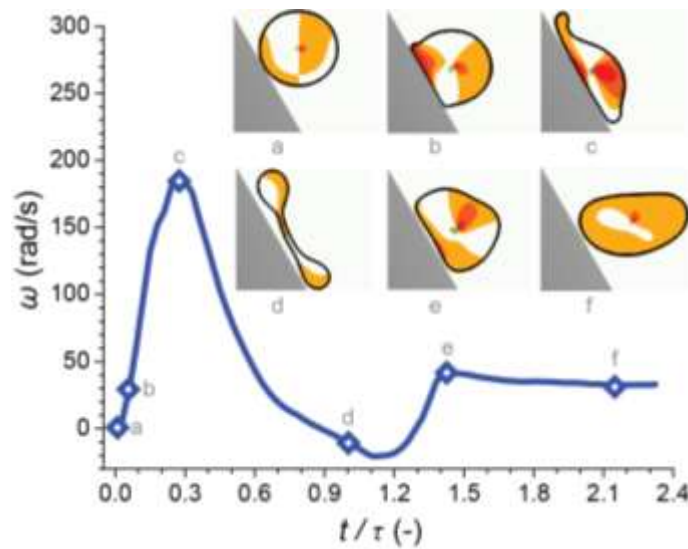


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248 **Figure 7:** History of the average angular velocity, ω , of highly viscous liquid drop with $Oh = 3.2$, $We = 121$,
 249 $\alpha = 60^\circ$ during tumbling (simulation). Propagation of the dominant angular velocity component of $\vec{\omega}_{loc}$
 250 inside the drop is shown in the insets (a-f), at different stages of tumbling. Stages a-c: Initial rise of the angular velocity shortly
 251 after the impact; Stages d-f: Rotation of the drop acting essentially as a solid. Drop rebounds (f) at $t/\tau = 1.15$.
 252 Shading/color increase corresponds to the increase of the clock-wise rotation.

253

254 For comparison, Figure 8 shows the history of the averaged angular velocity, ω , in the low viscosity regime
 255 ($Oh = 0.1$), together with the characteristic snapshots of the distribution of the local angular velocity
 256 during impact (see also Video 10). Unlike in the case of high viscosity, the angular velocity gained initially in
 257 the impact (phase a-c in Figure 8) is not sustained at a constant value during the extension and sliding
 258 (phase c-d), and no solid-like tumbling is observed. At the final stage close to the rebound (f), a small gain
 259 of the average angular velocity is due to the merging of the dumbbell-like shape of the drop at the
 260 intermediate stage, with non-axisymmetric collapse of the rim from (d) to (e). In other words, in the low
 261 viscosity regime the fluid slips away and the rotation is not sustained.



262
 263 **Figure 8:** History of the average angular velocity ω of low viscosity liquid drop with $Oh = 0.1$, $We = 121$, and slope
 264 $\alpha = 60^\circ$ (simulation). Propagation of the dominant angular velocity component of $\vec{\omega}_{loc}$ inside the drop is shown in
 265 the insets (a-f), at different stages of drop impact. Drop rebounds (f) at $t/\tau = 2.1$. Shading/color increase
 266 corresponds to the increase of the clock-wise rotation.

267

268 IV CONCLUSIONS

269 Taken together, our results demonstrate and explain the existence of a new, prompt tumbling-rebound
 270 regime for non-axisymmetric drop impact on surfaces under slip conditions, here readily realized with the
 271 help of a sublimating slope. To this end, sublimating surfaces present themselves as an interesting, easy to
 272 use platform for the study of unexplored, liquid-surface interactions, especially in the limit of small friction,

273 brought about by the absence of direct contact between the liquid and the solid surface. Similar effects
274 could be possible also on superhydrophobic surfaces, if they are fabricated to reach extreme performance.
275 Preliminary simulations for contact angles $170^\circ < \theta < 180^\circ$ indicate that tumbling-rebound could take
276 place, opening an interesting direction of future work, e.g. to promote repellence of viscous supercooled
277 drops in icing conditions [30,45].

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279

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285

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