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Acoustic responses of underwater superhydrophobic surfaces subjected to an intense pulse

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10 Underwater stability of an air layer trapped in a micro-structure, plastron, is critical in drag reduction application. Here we investigate the wetting state and plastron stability upon 11 underwater superhydrophobic surfaces (SHS) under an intense acoustic drive. Flat surfaces 12 and SHS are subjected to short acoustic pulses of different intensities. At low amplitude, 13 the comparison between the results of various surfaces shows that plastron behaves like a 14 water-air interface, whose presence can be detected from the phase of the reflected acoustic 15 waves. At moderate intensity, a wetting transition towards a completely wetting state is 16 observed and shown to be triggered by a sufficiently large acoustic radiation pressure. This 17 wetting transition is well captured by a simplified model by balancing radiation pressure 18 with the critical capillary pressure for the interface sliding. Cavitation clouds appear under 19 strong excitation; their sizes and position greatly depend on the surface acoustic boundary 20 condition. For a SHS in a Cassie-Baxter state (with an air layer), cavitation clouds appear 21 at specific locations (from the solid surface) corresponding to the pressure anti-node of the 22 transient standing wave generated by the reflection. This study unprecedentedly demonstrates 23 the capability of acoustic waves to monitor and characterize plastron stability with low and 24

- 25 moderate amplitudes, respectively.
- 26 Key words: acoustics, cavitation, contact lines, wetting and wicking

27 1. Introduction

28 Superhydrophobic surfaces (SHS) are bio-inspired surfaces composed of hydrophobic

- 29 patterned micro/nano-structures (Quéré 2008). The combination of roughness and hydropho-
- 30 bicity makes such surfaces water-repellent, an attracting property for various applications
- 31 (Quéré 2008). When immersed in water, SH surfaces can trap an air layer, also called
- 32 plastron, inside the microstructure (Bobji *et al.* 2009), opening the avenue for drag reduction

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applications since such surfaces exhibit significant effective slipping lengths (Rothstein 33 2010). However, the stability of the air layer against pressure fluctuations, induced by 34 potentially turbulent flow (Seo et al. 2018; Castagna et al. 2018) or against gas dissolution 35 (Poetes et al. 2010), is a crucial problem in the development of such SH coatings. Indeed, 36 the air layer can be pushed inside the microstructure, and the superhydrophobic state breaks 37 38 down (Moulinet & Bartolo 2007; Reyssat et al. 2007), i.e., a wetting transition occurs from 39 a (gas-trapping) Cassie-Baxter to a (completely wetting) Wenzel state, and the surface drag coefficient increases dramatically (Karatay et al. 2013). Although substantial efforts have 40 been invested in developing robust SHS (Verho et al. 2012; Xiang et al. 2017), plastron 41 stability still limits the useful applications of SHS. 42

To better understand superhydrophobic breakdown, the wetting transition has been ex-43 44 tensively studied in quasi-static configurations, such as in evaporating droplet (McHale et al. 2005; Jung & Bhushan 2007; Tsai et al. 2010; Bussonnière et al. 2017), with 45 46 surfactant additives (Shardt et al. 2019; Aldhaleai & Tsai 2022), under different drop volume (Yoshimitsu et al. 2002) and pressure (Lafuma & Quéré 2003), or in dynamic situations, e.g., 47 48 drop impact (Bartolo et al. 2006) or under surface vibrations (Bormashenko et al. 2007). For immersed SHS, wetting transition was mainly studied under static pressure (Poetes et al. 49 2010; Lv et al. 2014). However, in the main application of underwater SHS (drag reduction), 50 pressure fluctuates due to the flow, which can become turbulent (Seo et al. 2018). 51

Acoustic waves have the ability to impose precise pressure over an extensive frequency 52 range. Moreover, sound waves strongly interact with bubbles (e.g., a plastron can be seen 53 54 as an elongated bubble) in liquid. Therefore, an acoustic method can be a well-suited tool for submerged SHS characterisation. Promising plastron acoustic monitoring was obtained 55 by using longitudinal waves coming from the solid, from beneath the surface, to probe 56 57 the wetting state by monitoring the back-scattered waves (Saad et al. 2012; Dufour et al. 2013; Li et al. 2014). However, this method requires calibration or precise knowledge of the 58 59 morphology of the SH coating. Furthermore, elastic acoustic waves are not in contact with the air-water interface and, therefore, prohibit direct interaction with the gas-liquid interface. 60 In (Sudeepthi *et al.* 2020), drop wetting transitions were triggered by surface acoustic waves 61 through indirect interaction, i.e., through drop oscillations. Other authors studied submerged 62 SHS as potential acoustic meta-materials (Tong et al. 2020; Feng et al. 2019). However, the 63 strong interaction between acoustic waves coming from the liquid phase and a SHS remains 64 unexplored. Such an acoustic method is expected to be versatile since the technique uses 65 only one portable transducer to excite and receive waves, similar to non-destructive testing. 66 The acoustic technique would be hence well suited to characterize SHS in applications or 67 in complex environments (e.g., underwater) whereby classical optical techniques, such as 68 confocal microscopy (Papadopoulos et al. 2013) or light interference (Moulinet & Bartolo 69 70 2007) which require a microscope and transparent substrate, cannot be used. 71

In this paper, we propose to fill the gap by exploring experimentally the behavior of immersed superhydrophobic surfaces subjected to short acoustic pulses of varying intensity. Different SHS and flat surfaces are exposed to acoustic pulses using the experimental setup described in 2. At low acoustic amplitude, various surfaces show different acoustic boundary conditions described in Section 3.1. Cavitation behavior is characterized at high amplitude in Section 3.2. The wetting transition is observed at an intermediate intensity and discussed in Section 3.3.

78 2. Experimental setup

79 The experimental setup shown in Figure 1 consists of a High Intensity Focused Ultrasound

80 (HIFU) transducer (Sonic Concepts H101, 1.1 MHz) which focuses short acoustic pulses onto

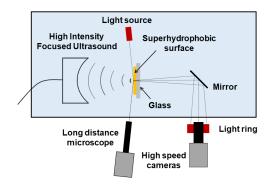


Figure 1: Schematic of the experimental setup. Short acoustic pulses are emitted by a High Intensity Focused Ultrasound transducer and focused upon the tested surface held by a glass coverslip. The dynamic is monitored from the side and from the bottom by two high speed cameras.

81 an immersed surface with a circular focal area of 3 mm in diameter. The acoustic excitation comprises 3 cycles of 1.1 MHz. To calibrate the pressure, we first map the acoustic field at 82 a constant low amplitude using a needle hydrophone (Onda HNR-0500, bandwidth 0.25-10 83 MHz) mounted on 3D axis displacement stage and use the open source code solving nonlinear 84 propagation (Khokhlov-Zabolotskaya-Kuznetsov equation) developed by Joshua Soneson 85 (Soneson 2009; Maruvada et al. 2015) to fit the experimental results. The hydrophone is then 86 kept at the focal point (for low amplitude) and at the secondary lob (for high amplitude), and 87 the amplitude is varied. Combining the acoustic mapping, pressure amplitude measurements, 88 and the nonlinear simulation, we finally obtain the calibrated pressure field for a specific 89 supply voltage of the acoustic drive (see Appendix A). 90 Four surfaces were used in this study to compare different behavior under acoustic excita-91 tion: glass, flat PDMS, and two PDMS SH surfaces. The SH and flat PDMS surfaces were 92 first placed on a glass surface and then introduced at the focal point. The superhydrophobic 93 surfaces consist of micro-scale cylindrical pillars of 5.5 μ m in diameter (D) and 5 μ m in 94 height (H) organized in a square lattice. SHS 1 and 2 have a periodicity P of 14.1 μ m and 95 7.6 μ m, respectively. Superhydrophobic surfaces were made by pouring liquid PDMS into 96 a Si mold, vacuumed, cured, and finally peeled off as the samples. The PDMS surfaces 97 were 3 mm thick. Once introduced at the focal point, surface dynamics is monitored by two 98 high-speed cameras (Photron AX200 and AX100), while the HIFU transducer records the 99

back scattering sound at low amplitude. A long range microscope is mounted on a camera to record from the side, while the second camera is used with a mirror to image the dynamic

102 from the bottom (see Fig. 1).

103 **3. Results and Discussion**

104

3.1. Initial wetting state

The initial wetting state for the different surfaces is compared in Figure 2 using the bottom view and acoustic reflection at small amplitude. Glass, flat PDMS, and SH1 surfaces appear black from the bottom view, indicating that the light coming from the bottom is not reflected, and hence the solid surfaces are fully wet. The bottom view of the SH2 surface is much brighter, indicating that the bottom light is reflected by air-water interface, and thus an air layer is trapped inside the micro-structure. The stability of the Cassie-Baxter state has been predicted by Bico *et al.* (Bico *et al.* 2002) based on energy analysis. They show that an air

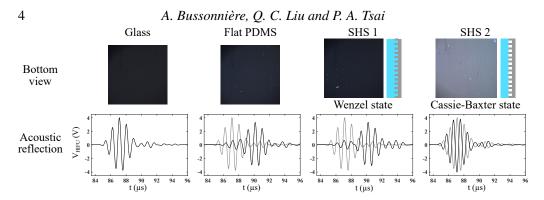


Figure 2: Comparison of the initial wetting state of the different solid surfaces submerged in water. The first row of images shows the bottom view by a high-speed camera, while the second row of data are the back-scattered sound detected by monitoring the voltage of the transducer. For comparison, the acoustic reflection from the glass surface solely (bottom left) is added in grey for the other surfaces.

112 layer will be stable if the contact angle of the constituting material on flat surface θ_f obeys:

113
$$\theta_f > \theta_c$$
, with $\cos \theta_c = \frac{\Phi - 1}{R - \Phi}$, (3.1)

114 where $R = 1 + \pi DH/P^2$ is the roughness, and $\Phi = (\pi D^2)/(4P^2)$ is the solid fraction. Our 115 SH1 (SH2) has a roughness R = 1.43 (R = 2.5) and a solid fraction of $\Phi = 0.12$ ($\Phi = 0.41$), 116 leading to a critical contact angle $\theta_c = 134^o$ ($\theta_c = 106^o$). The water contact angle on flat 117 PDMS is $\theta_f = 110 - 115^o$ (Mata *et al.* 2005). In agreement with the theoretical prediction, 118 the θ_f -value for the flat PDMS is smaller than θ_c for the SH1 surface so water invades the 119 microstructure. For the SH2 surface $\theta_f > \theta_c$, therefore, a stable air layer is trapped between 120 the micropillars, as observed in the experiments.

The back-scattered sound of the different surfaces is then analyzed at a small amplitude. 121 The HIFU transducer voltage averaged over ten pulses is shown in Figure 2. For all surfaces, 122 echoes are detected at 85 μ s, corresponding to the time required for an acoustic wave to travel 123 to the focal point and return to the transducer. The reflected pulse has the same shape as the 124 incoming pulse for the glass surface. For the flat PDMS and the SHS 1, the back-scattered 125 126 pulse is more complex and consists of a first small amplitude reflection followed by a second higher reflection similar to the glass case. More surprisingly, the back-scattered sound from 127 the SHS 2 is only made of a strong pulse with a π -phase change, compared to the incoming 128 pulse. 129

These observations can be explained using linear acoustics. The pressure reflection 130 coefficient (r) of acoustic plane waves on an interface is given by $r_{i,j} = (Z_j - Z_i)/(Z_i + Z_j)$, 131 with $Z_i = \rho_i c_i$ the acoustic impedance, ρ_i and c_i the density and sound velocity in the 132 media *i*. The subscript i(j) represents the media supporting the incident (transmitted) pulse. 133 134 The different interfaces present in our experiments have the following reflection coefficient: $r_{w,g} = 0.8$ for water/glass interface, $r_{w,p} = -0.18$ for water/PDMS, $r_{w,a} = -1$ for water/air 135 and $r_{p,g} = 0.85$ for PDMS/glass (Xu et al. 2020). For the water-glass interface, $r_{w,g}$ is 136 positive and close to 1, and the back-scattered pulse is expected to be of high amplitude 137 without phase change as observed in Figure 2. The flat PDMS and SH1 surfaces are held by 138 a glass plate, and the incoming pulse will first encounter the water/PDMS interface leading 139 to a first weak reflection since $r_{w,p}$ is small. A non-negligible part of the pulse is transmitted 140 to the PDMS and then reflected by the PDMS/glass interface, generating the second strong 141 142 echo since $r_{p,g}$ is high.

143 Finally, the back-scattered sound coming from the SH2 surface is solely made of a strong

pulse with π -phase change, in agreement with a reflection on a water/air interface as $r_{w,a}$ = 144 -1. This confirms the presence of an air layer inside SH2 microstructure as optically observed, 145 demonstrating that acoustics can be of great use to probe plastron stability. The absolute 146 value of the reflection coefficient of the water/air interface is higher than the coefficient of 147 water/glass ($|r_{w,a}| > |r_{w,g}|$). The amplitude of the back-scattered signal from SHS 2 should 148 therefore be higher than the one from the glass surface, but the measured reflections are of 149 150 similar amplitude (see Figure 2). This discrepancy might be ascribed to the fact that the SH2 interface is not a pure water/air interface but a mixture of water/air and PDMS pillars (41% 151 152 of the surface is PDMS), which modifies the reflection coefficient.

153

3.2. Cavitation behavior

The solid surfaces were subjected to high-intensity pulses of different amplitudes to 154 155 characterize their cavitation behavior. The cavitation is monitored by side-view images 156 [see Figure 3 (b)-(c)] taken during the acoustic excitation when cavitation clouds are visible. The area of such cavitation cloud (A_{cav}) is measured and compared for the different surfaces 157 under various excitation amplitudes in Figure 3 (a). Although the data are scattered, which is 158 expected for cavitation experiments, it can be seen that only a few small cavitation bubbles 159 appear on flat PDMS and on the SHS 1. In contrast, significant cavitation is observed on 160 the glass surface and the SHS 2. The flat glass surface exhibits cavitation at low acoustic 161 excitation, while cavitation cloud appears later for SHS 2. 162

The ability of surfaces or liquids to cavitate under relatively small pressure excitation, well above the spinodal pressure of water (~ -140 MPa) (Zheng *et al.* 1991), is determined by the presence of micro/nanobubbles trapped in surface defects or impurities, called cavitation nuclei (Harvey *et al.* 1944). Stabilisation of such nuclei on a surface requires a hydrophobic geometrical defect called crevice (Atchley & Prosperetti 1989; Bussonniere *et al.* 2020). When subjected to low enough negative pressure (i.e., cavitation threshold), such nuclei become mechanically unstable, and cavitation bubbles appear.

The different cavitation responses are therefore either due to a difference in nuclei 170 population and/or in local negative pressure. The first nuclei source in the experiments 171 comes from tap water, which contains many impurities and thus a large number of nuclei. 172 However, the same water was used in all experiments, and hence these nuclei could not 173 explain the cavitation differences observed. The second source of nuclei comes from the 174 different solid surfaces. PDMS is naturally hydrophobic, which should favor nuclei trapping 175 176 compared to hydrophilic glass. However, this soft material is expected to have little, if no, sharp defect (crack-like) due to its polymerization from a liquid form, which can explain that 177 almost no cavitation on flat PDMS was detected (Figure 3). Similarly, SHS 1 in a Wenzel 178 wetting state, although being micro-textured with cylindrical pillars, is not expected to have 179 180 sharp crevices, which can explain the small cavitation activity observed. Glass, on the other hand, can have small cracks potentially leading to significant cavitation activity as observed 181 182 on glass.

The other important factor for cavitation is the local negative pressure, which is greatly 183 influenced by the acoustic boundary condition. The acoustic wave reflected by a surface with 184 a factor of r interferes with the incident wave resulting in a local minimum pressure, which 185 can greatly differ from the incident minimum pressure, P_{min}^{inc} . For example, at the interface between water and a surface *j*, the minimum pressure is $P_{j}^{wall} = (1+r_{w,j}) P_{min}^{inc}$. The minimum pressure is therefore amplified on the glass surface ($P_{glass}^{wall} = 1.8 P_{min}^{inc}$), reduced on flat PDMS 186 187 188 and SHS 1 ($P_{pdms,shs1}^{wall} = 0.8 P_{min}^{inc}$), or almost cancelled on SHS 2 ($P_{shs2}^{wall} \approx 0$). This simplified 189 analysis is in agreement with the experiments where cavitation clouds appear on the surface 190 for glass [Figure 3 (b)], whereas no cavitation is observed upon the surface of SHS 2 (Figure 191

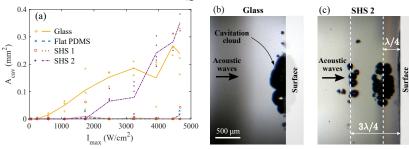


Figure 3: (a) Area of the cavitation cloud (A_{cav} , captured from the side-view) as a function of the maximum incident acoustic intensity (I_{max}) for the different solid surfaces. Points in (a) represent the area of one experiment, while the solid lines represent the average values. (b)-(c) Side-view images of the cavitation cloud on glass surface [in (b)] and on the superhydrophobic surface 2 [in (c)] appearing for an incident maximum intensity of 3.9 kW/cm².

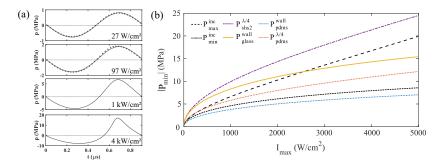


Figure 4: (a) Incident waveform at the focal plan for different maximum incident intensity (I_{max}). The simulation results (solid lines) using the nonlinear acoustic code (Soneson 2009) are compared to the experimental measurements (cross points) at low intensity. (b) Evolution of the minimum and maximum incident pressure ($P_{min,max}^{\text{inc}}$) and of the minimum local pressure ($P_{surface}^{\text{position}}$) at specific position for a surface with the maximum incident intensity (I_{max}).

3 (c)). Note that the small bubbles in Figure 3 (b) top are actually on the surface, although
 appearing away from it due to the slight tilt between the camera and the surface.

Interference between the incident and reflected waves also occurs away from the surface. 194 If a continuous excitation was used, a standing wave in the liquid would have been generated. 195 However, here, we use short pulses with only 2 periods with a significant amplitude. Wave 196 interference therefore only occurs in the surface vicinity, in a region of approximately one 197 wavelength (λ). Local minimums generated by the transient standing wave depend on the 198 199 reflection coefficient sign. If r > 0, the pressure anti-nodes are located on the surface, at $\lambda/2$ and λ away for the surface. Differently, if r < 0 the minimum pressures are located at $\lambda/4$ 200 and $3\lambda/4$ away from the surface. The above analysis may be consistent with the experimental 201 data, which reveals that cavitation cloud appears at $\lambda/4$ and $3\lambda/4$ for the SHS2 experiments. 202 However, with the analysis one would have expected appearance of cavitation at λ and $\lambda/2$ 203 in the experiments with the glass surface and not only on the surface. 204

To understand this discrepancy for the glass surface, one needs to analyze the high acoustic intensity used in the experiments. Under such strong excitation, the acoustic propagation is nonlinear, and the acoustic waveform departs from a simple cosine function, as shown in Figure 4 (a). The solid lines represent the results from the nonlinear acoustic simulations. The simulation results are in good agreement with the experimental data (crosses) at low values of I_{max} . Note that the measurements at the focal point can only be performed at low intensity to prevent deterioration of the hydrophone by cavitation. As detailed in the appendix A, the acoustic field at high intensity was calibrated by measuring the pressure at the second pressure lob (after the focal region). As I_{max} increases, the waveform become asymmetric, and the maximum incident pressure P_{max}^{inc} increases faster than the absolute minimum pressure $|P_{min}^{inc}|$, as shown in figure 4 (b).

Such asymmetry has important implications in the local minimum generated in the 216 interference region. For r > 0, the local minimums located at $\lambda, \lambda/2$ and on the wall are equal 217 to $P_j^{\lambda} = P_j^{\lambda/2} = P_j^{\text{wall}} = (1 + r_{w,j}) P_{\min}^{\text{inc}}$. However, for r < 0, the reflected wave is inverted (with a phase change by π), and the local minimum pressures are $P_j^{3\lambda/4} = P_j^{\lambda/4} = P_{\min}^{\text{inc}} - r_{w,j}P_{\max}^{\text{inc}}$. 218 219 Figure 4 (b) shows the minimum pressures generated by the combination of nonlinear 220 221 acoustic and the reflection for the different surfaces. As the reflection from SHS2 at low intensity (Figure 2) has the same amplitude as the glass surface, we use $r_{w,shs2} = -0.8$ 222 to better estimate the reflection. Due to the reflection of the strong maximum incident 223 pressure, minimum pressures in SHS2 experiments reach the lowest level, which explains 224 the biggest cavitation cloud observed. Cavitation with SHS2 appears at I = 2.5 kW/cm², 225 i.e., at $P_{shs2}^{1/4} \approx -16$ MPa, which corresponds to the threshold for cloud cavitation in water. Pressure in the glass experiment only reaches $P_{glass}^{wall} \approx -15$ MPa at the maximum intensity. 226 227 This explains why no clouds are observed in water for the glass cases. Cavitation, in this case, 228 229 occurs only on the glass interface due to the presence of surface nuclei. It should be noted that the nonlinear waveform reflected with a phase shift by π is unstable and will recover to 230 a sine wave at the transducer, shown by Tanter et al. (2001). 231

3.3. Wetting transition

The stability of the superhydrophobic state is also studied by using the bottom camera and the 233 acoustic reflection at a small amplitude. Figure 5 (a) shows the images from the bottom taken 234 after the exposition of the SHS 2 to different intense acoustic pulses. Note that the surface is 235 moved between each high impulsion experiment, so each trial is performed on a fresh area 236 of the SHS. For intensities smaller than ≈ 0.1 kW/cm², the air layer remains unchanged, as 237 shown by the brownish color. Starting at 0.25 kW/cm², a change in color is observed, and 238 clear interference color pattern appears at 0.59 W/cm^2 . These colors arise from the white 239 light interference inside the air layer of the SHS and indicate that the air-layer thickness has 240 been decreased at the focal region due to the acoustic pulse. For $I_{max} > 1.1$ kW/cm², a 241 dark disk appears in the center surrounded by interference fringes. The radius of this disk 242 increases with the acoustic intensity, and its evolution with I_{max} is reported in Figure 5 (c). 243

This black disk might correspond to a local superhydrophobic breakdown (i.e., wetting 244 transition) since SHS 1, which didn't trapped an air layer, appears black in Figure 2. To 245 verify this, the acoustic reflection signals at low amplitude from the SHS 2 surface before 246 (shown in dashed blue) and after (shown in red) an intense acoustic pulse of 2.5 kW/cm² 247 are compared in Figure 5 (b). Before, the reflection corresponds to an air-water reflection 248 as shown in Figure 2. The reflection after the high intensity pulse is drastically changed 249 and is similar to the reflection of SHS 1 of Figure 2. This observation confirms that high 250 251 intensity pulses trigger a local wetting transition, corresponding to the dark area where water penetrates inside the microstructure. It should be note that this transition occurs at amplitude 252 smaller ($I_{max} = 1.1 \text{ kW/cm}^2$) than the appearance of cavitation cloud ($I_{max} = 2.5 \text{ kW/cm}^2$), 253 ruling out a transition driven by cavitation. 254

The transition from a Cassie-Baxter to a Wenzel wetting state is usually understood by looking at the local force balance of the air-water interface in contact with the microstructure. A small pressure increase in the liquid phase is balanced by a capillary pressure arising from

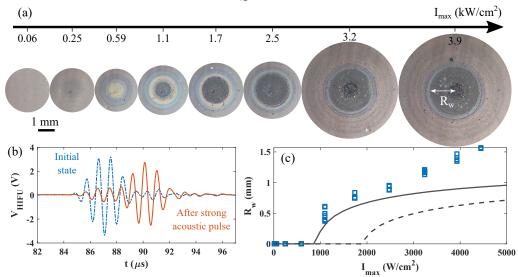


Figure 5: (a) Bottom view of the SHS 2 after exposition of different high intensity acoustic pulses showing the apparition of interference fringes and a wetted region in black of radius (R_w) . (b) Acoustic reflection of the SHS 2 of a small amplitude pulse before (blue) and after (red) the exposition of a high intensity pulse of $I_{\text{max}} = 2.5 \text{ kW/cm}^2$. (c) Evolution of the wetted radius (R_w) observed experimentally as a function of the maximum acoustic intensity (I_{max}) . The solid and dashed lines correspond the wetted radius predicted by the sliding and touch-down models, respectively.

the bending of the interface between pillars (Quéré 2008). At higher liquid pressure, the
interface can either touch the bottom of the SHS (i.e., touch-down scenario) or slide on the
pillar when the local contact angle on pillars reaches the advancing one (i.e., sliding scenario)
(Moulinet & Bartolo 2007; Reyssat *et al.* 2007). These different scenarios are associated with
two critical liquid pressures:

$$p_t^c \approx \frac{2\gamma H}{(\sqrt{2}P - D)^2},\tag{3.2}$$

for the touch-down, with γ the air-water surface tension, and θ_a the advancing contact angle; for the sliding (Moulinet & Bartolo 2007):

266
$$p_s^c = \frac{2\Phi}{1-\Phi} |\cos\theta_a| \frac{2\gamma}{D}.$$
 (3.3)

Previously, we showed that the oscillating acoustic pressure at the SHS 2 interface almost 267 vanishes due to the acoustic boundary condition, and thus the high oscillating acoustic 268 pressure (at 1.1 MHz) cannot explain the wetting transition. Nonetheless, acoustic waves 269 are known to apply radiation pressure on interfaces (Borgnis 1952) and are commonly used 270 to trap objects in acoustofluidics (Bruus 2012). Such radiation forces arise from a change 271 of media momentum induced by the acoustic scattering or reflection of an interface or an 272 object (Westervelt 1957; Baresch et al. 2013). These radiation forces originate from the 273 difference of the time-averaged acoustic energy density, i.e., radiation pressure, across the 274 interface. It is a cumulative process, and these forces hence occur on a longer timescale than 275 276 the fast-oscillating acoustic pressure. If we assume that the SHS 2 behaves as a pure water-air interface and an incident angle of 90° , the radiation pressure applied on the interface is given 277

278 by (Borgnis 1952):

279

$$p_{\rm rad}(r) = \frac{2I(r)}{c},\tag{3.4}$$

with *I* the incident acoustic intensity, *c* sound speed in water, and *r* the radial coordinate on the surface centered on the focal axis. The acoustic intensity, and thus the radiation pressure, depends on *r* due to the HIFU geometry as shown in Figure 6 (b). The wetting transition would occur if $p_{rad} > p_{s,t}^c$, and a critical radius r_c can be extracted from the simulated intensity field whereby $p_{rad}(r_c) = p_{s,t}^c$. The predictions for r_c using the sliding and touchdown models presented above are compared with the experimental data in Figure 5 (c) for $\theta_a = 110^\circ$.

1

This simplified model predicts the beginning of the transition around $I_{\text{max}} = 900 \text{ W/cm}^2$ 287 for the sliding scenario, which is in good agreement with the experimental data. Moreover, 288 the wetted radius R_w agrees well with the critical radius predicted in the range 1-2.5289 kW/cm² of I_{max} . The good agreement indicates that the wetting transition occurs where the 290 acoustic beam is more intense and also validates the wetting transition driven by radiation 291 pressure. It should be noted that the interference pattern starts to appear at a lower intensity 292 (see Figure 5 (a) for $I_{\text{max}} = 0.59 \text{ kw/cm}^2$). This may suggest that the interface starts to slide 293 at lower radiation pressure than predicted but does not reach the microstructure bottom. A 294 more comprehensive analysis involving time dependence, similar to the one done for free 295 296 surface in (Issenmann et al. 2011), is needed to better understand the interface motion under radiation pressure. In addition to developing such a complex model, a study of the influence 297 of pulse duration and a better acoustic calibration would be needed to fully capture the 298 interface dynamic in the future. 299

For larger intensity, the water penetration area is underestimated by the sliding model. 300 301 This deviation coincides with the appearance of an intense cavitation cloud, which can reflect acoustic wave and also create strong pressure during its collapse. Therefore, the local acoustic 302 field is expected to greatly differ from the simple combination of an incident and reflected 303 wave, thereby modifying the radiation pressure on the surface. Note that the local acoustic 304 field can be inferred from the imprint left on the SH surface. In Figure 5 (a), concentric grey 305 circles surrounding the wetted area appear and are spaced by $\approx \lambda/4$. However, such circles 306 cannot be explained by the secondary lobes of the incident beam, spaced by $\approx \lambda$ (see Figure 307 6 (b)), which tends to support a strong modification of the acoustic field by the cavitation 308 cloud. 309

310 4. Conclusions

We investigate the acoustic responses of superhydrophobic surfaces subjected to short intense pulses. Experiments at low amplitude reveal that when a SHS is able to stabilize an air layer (in a Cassie-Baxter state) the acoustic waves are reflected with a π phase-shift similar to an air-water interface. In a Wenzel state, a patterned surface behaves as a flat surface of the same material, and the microstructure has no influence on the acoustic reflection.

At high amplitude, intense cavitation is observed for the glass surface and SHS in (Cassie-316 Baxter) gas-trapping state, while almost no cavitation is detected for flat PDMS surface and 317 PDMS SHS in a Wenzel state. This difference is mainly attributed to the acoustic boundary 318 condition. For hard material, such as glass, the pressure on the solid surface is nearly doubled, 319 leading to the appearance of a cavitation cloud on the surface. For the flat and wetted patterned 320 PDMS surfaces (i.e., flat and SHS in Wenzel), the pressure on the surface is slightly lowered 321 322 due to the close match in acoustic impedance with water. For SHS in a Cassie-Baxter state, the interference between the reflected and the incident wave results in a significant negative 323

pressure away from the surface (at $\lambda/4$ and $3\lambda/4$), where intense cavitation clouds form. Moreover, cavitation, in this case, is amplified by the conversion of the strong high pressure peaks arising from nonlinear propagation into great negative pressure by the reflection on the air layer.

The Wetting transition from Cassie to Wenzel state is observed at moderate intensity before cavitation appearance. The transition is shown to be driven by the acoustic radiation pressure, which overcomes the critical capillary pressure required to let the interface slide into the microstructure. The simplified model proposed well captures the transition and the size of Wenzel area at moderate intensities. For higher intensity, strong interaction with the cavitation cloud is expected to modify the local acoustic field, resulting in a deviation with the proposed model.

Underwater superhydrophobic surfaces are mainly studied in the context of drag reduction. 335 In such an application, pressure on the plastron is governed by the hydrodynamic, and the flow 336 can trigger a wetting transition. In this context, our results at low amplitude can contribute to 337 338 developing non-destructive acoustic monitoring of the plastron stability. Moreover, the critical breakdown pressure of a superhydrophobic coating could be characterized and measured 339 340 using acoustic pulses at moderate intensity. For future studies, an accurate measurement of this breakdown pressure would require a better calibration of the acoustic field and more 341 342 extensive studies on the influence of the pulse duration and excitation frequency. 343

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353 Appendix A. Acoustic calibration

The acoustic field is first mapped at low amplitude using a needle hydrophone mounted on a 3-axis stage. This field is then used to find the parameter of the HIFU transducer (focal length and radius) in the nonlinear simulation (Soneson 2009). The axial distribution, *z*, the distance from the transducer, is measured along the axisymmetric axis and shown in Figure 6 (a). The simulated field captures well the pressure field in the focal region and peripheral lobes. Nonetheless, a slight deviation is observed for *z* below 40 mm. The radial distribution shown in Figure 6 (b) is measured in a plane perpendicular to the *z*-axis at the maximum pressure, i.e., at *z* = 62 mm. The simulation results are in good agreement with the experimental measurements.

At higher amplitude, cavitation occurs in the focal region, and no direct measurement can be performed 361 362 using the hydrophone (as the intense cavitation can damage the hydrophone). Therefore, the nonlinear 363 simulation is used to obtain the pressure at high intensity. The unknown in the simulation is the pressure impose at the transducer surface (p_{sim}) . In the experiment, we impose a voltage at the output of the generator 364 (V_{gen}) which is then amplify and converted into pressure by the HIFU. To calibrate our experiment using the 365 simulation we therefore need to fin the relation between p_{sim} and V_{gen} . To find such relation we measured the 366 maximum and minimum pressure ($p_{\text{max}}^{\text{foc}}$ and $p_{\text{min}}^{\text{foc}}$) at low amplitude at z = 62 mm (in the focal region) and over a large range of voltage in the secondary lobes ($p_{\text{max}}^{\text{2nd}}$ and p_{min}^{2nd}) located at z = 81 mm. Simulations were then performed for different amplitudes of p_{sim} , and the functions $p_{\text{max}}^{\text{foc}}(p_{\text{sim}})$, $p_{\text{min}}^{\text{foc}}(p_{\text{sim}})$, $p_{\text{max}}^{2nd}(p_{\text{sim}})$ and 367 368 369 $p_{\min}^{2nd}(p_{sim})$ were extracted. These functions were inverted to convert the experimental measurement into 370 p_{sim}^{min} values. In Figure 6 (c), we show the results of this treatment as a function of V_{gen} . All the experimental 371 372 points collapse into a single line, which is fitted with a 3rd-order polynomial without a constant term. Note

that the curve tends to saturate, which seems to indicate a saturation of the voltage amplifier limited to 200
 W (E&I 1020L).

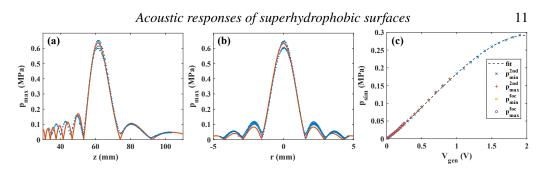


Figure 6: Evolution of the maximum acoustic pressure in the axial (a) and radial (b) direction at low amplitude. The red lines correspond to the prediction of the acoustic simulation using the non-linear code (Soneson 2009), while the blue dots represent the experimental points. (c) Evolution of the input pressure of the simulation (p_{sim}) fitting the different maximum and minimum measurements as a function of the generator voltage (V_{oen}).

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