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# Wetting of soap bubbles on hydrophilic, hydrophobic, and superhydrophobic surfaces

Steve Arscott<sup>a)</sup>

*Institut d'Electronique, de Microelectronique et de Nanotechnologie (IEMN), CNRS UMR8520, The University of Lille, Cité Scientifique, Avenue Poincaré, 59652 Villeneuve d'Ascq, France*

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Wetting of sessile bubbles on various wetting surfaces (solid and liquid) has been studied. A model is presented for the apparent contact angle of a sessile bubble based on a modified Young's equation—the experimental results agree with the model. Wetting a hydrophilic surface results in a bubble contact angle of  $90^\circ$  whereas using a superhydrophobic surface one observes  $134^\circ$ . For hydrophilic surfaces, the bubble angle diminishes with bubble radius whereas on a superhydrophobic surface, the bubble angle increases. The size of the plateau borders governs the bubble contact angle, depending on the wetting of the surface. © 2013 AIP Publishing LLC.

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An understanding of the behavior of bubbles, liquid films, foams, and froths is vital for several fields including mining, manufacturing, materials, security, and food production,<sup>1</sup> and the use of soap bubbles and films has been recently demonstrated in micro<sup>2,3</sup> and nanotechnologies.<sup>4,5</sup> Soap bubbles and films have been studied for some time now;<sup>6–9</sup> more recent investigations include *inter alia* their composition,<sup>10</sup> organization,<sup>11,12</sup> electrification,<sup>13–15</sup> magnetization,<sup>16</sup> wetting,<sup>17–19</sup> stability,<sup>20</sup> and mechanical<sup>21</sup> and optical properties.<sup>22</sup> Here, the wetting of sessile soap bubbles on solid surfaces (hydrophilic to superhydrophobic) and on a liquid surface is studied. The results have potential implications in the aforementioned applications.

Fig. 1 shows ideal cases for a sessile droplet wetting a solid surface [Fig. 1(a)], a sessile bubble wetting a solid surface [Fig. 1(b)], and a sessile bubble resting on the surface of a liquid [Fig. 1(c)]. For the droplet, the balance between the surface tension of the liquid  $\gamma_l$ , the solid-liquid  $\gamma_{sl}$ , and the solid-vapor  $\gamma_{sv}$  surface energies is given by Young's equation<sup>23,24</sup> and leads to a liquid contact angle  $\theta_l$

$$\gamma_{sv} = \gamma_l \cos \theta_l + \gamma_{sl}. \quad (1)$$

For a sessile bubble wetting a solid surface [Fig. 1(b)], Young's equation needs to be modified to take into account the internal surface of the bubble. As the bubble has two surfaces, its effective surface tension  $\gamma_b$  is twice the surface tension of the liquid, i.e.,  $\gamma_b = 2\gamma_l$ . In addition to this, an extra  $\gamma_l$  term is required, acting in the same direction as  $\gamma_{sl}$ , in order to take into account creation of bubble surface inside the bubble. Thus, the modified Young's equation for an ideal sessile bubble forming a contact angle  $\theta_{b0}$  with a solid surface can be written as

$$\gamma_{sv} = 2\gamma_l \cos \theta_{b0} + \gamma_l + \gamma_{sl}. \quad (2)$$

Equations (1) and (2) allow us to write the contact angle of an ideal sessile bubble  $\theta_{b0}$  resting on a surface in terms of the contact angle of a droplet of bubble solution  $\theta_l$  resting on the same surface

$$\cos \theta_{b0} = \frac{1}{2} (\cos \theta_l - 1). \quad (3)$$

Equation (3) assumes that the bubble film thickness at the bubble-solid interface is of the order of the liquid film forming the bubble and that the droplet and bubble are considered to be large enough so that the diminishing contact angle with droplet radius effect,<sup>25</sup> controversially attributed to the line tension,<sup>24</sup> is negligible. For a sessile bubble resting on the surface of a liquid of the same solution [Fig. 1(c)], if the Plateau borders are symmetrical<sup>17</sup> inside and outside the bubble and are much smaller than the bubble base radius, then the apparent contact angle  $\theta_{b0} = 90^\circ$  assuming the surface of the liquid to be perfectly hydrophilic, i.e.,  $\theta_l = 0$ .

A commercially available soap solution (Pustefix, Germany) was used to generate bubbles for the experiments—a soap solution is a mixture of pure water, a second liquid to increase the viscosity of the solution and reduce drainage (e.g., glycerol), and a surfactant (e.g., an organosulphate). The surface tension of the solution was measured to be  $28.2 \text{ mJ m}^{-2}$  (standard deviation =  $0.3 \text{ mJ m}^{-2}$ ) using the pendant drop method<sup>26</sup> and applying the appropriate correction factor (Ref. 27)—a value comparable with other experiments concerning soap bubbles and films.<sup>13,17,18</sup> As a calibration measurement, deionized water was measured, the result was a surface tension of  $72.7 \text{ mJ m}^{-2}$  (1.2). The density of the bubble solution was measured to be  $997.8 \text{ kg m}^{-3}$ .

The different wetting surfaces were fabricated using polished silicon wafers (Siltronix, France). In order of decreasing wetting, “Surface A” is a 200 nm thick layer of silicon dioxide grown on a silicon wafer using wet thermal oxidation. “Surface B” is a  $\sim 100 \mu\text{m}$  thick Polydimethylsiloxane (PDMS) layer—Sylgard<sup>®</sup> 184 (Dow Corning, USA) spin-coated onto a silicon wafer. “Surface C” is a  $\sim 250 \text{ nm}$  Teflon<sup>®</sup> layer obtained using spin-coating of Teflon<sup>®</sup> AF 1600 (Dupont, USA) diluted with Fluorinert FC-75 (3M, USA).<sup>28</sup> “Surface D” is composed of “black silicon”<sup>29</sup> produced using dry etching. This surface was subsequently deposited with a  $\sim 20 \text{ nm}$  thick fluorocarbon layer using a  $\text{C}_4\text{F}_8$  plasma (STS, UK).

The measured contact angles  $\theta_l$  of the soap solution on the surfaces A-D are given in the Table I. As the values of  $\gamma_{sv}$  are

<sup>a)</sup>Electronic mail: [steve.arscott@iemn.univ-lille1.fr](mailto:steve.arscott@iemn.univ-lille1.fr). Tel.: +33 320197979.

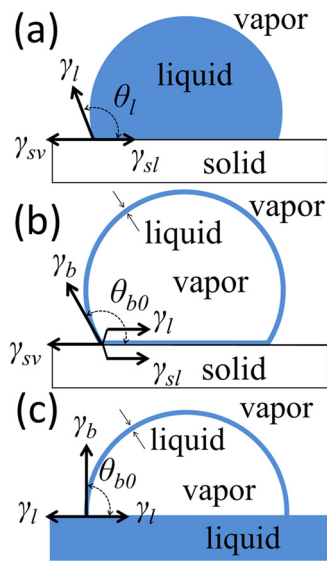


FIG. 1. Schematic diagrams showing the apparent contact angles for (a) a sessile droplet wetting a solid surface, (b) a sessile bubble wetting a solid surface, and (c) a sessile bubble resting on the surface of a liquid.

very well known for Teflon<sup>24</sup> ( $15 \pm 2 \text{ mJ m}^{-2}$ ) and PDMS<sup>30</sup> ( $23.5 \pm 1.5 \text{ mJ m}^{-2}$ ) we can determine the values of  $\gamma_{sl}$  to be ( $1.8 \pm 2.5 \text{ mJ m}^{-2}$ ) and ( $6.5 \pm 2 \text{ mJ m}^{-2}$ ). For a superhydrophobic surface,  $\gamma_{sv}$  is of the order of  $5\text{--}10 \text{ mJ m}^{-2}$  which indicates that  $\gamma_{sl}$  is in the range  $15.5\text{--}20.5 \text{ mJ m}^{-2}$ . Bubbles having radii in the  $0.5\text{--}10 \text{ mm}$  range were generated for the experiments using a pipette (Bio-Rad, France) having a tip diameter of  $\sim 0.5 \text{ mm}$ . All surface preparation and experiments were performed in a class ISO 5/7 clean room ( $T = 20^\circ\text{C} \pm 0.5^\circ\text{C}$ ;  $RH = 45\% \pm 2\%$ ). The data were gathered using a commercial Contact Angle Meter (GBX Scientific Instruments, France).

Bubbles were deposited onto surfaces A-D in order to form sessile bubbles [Fig. 2]. Millimeter-sized soap bubbles are quasi-spherical with radius of curvature  $R$  [Fig. 2(a)] and a contact angle  $\theta_b$  [Fig. 2(b)] as the Bond number<sup>24</sup> is small. Drainage can cause the formation of liquid layer  $h$  (the Plateau border<sup>9</sup>) [Fig. 2(c)] at the bubble-solid interface of radius  $r$  [Fig. 2(d)].

Fig. 3(a) shows plots  $\theta_b$  versus  $r$  in the range  $1.5$  to  $3.5 \text{ mm}$  for the four surfaces tested. In the case of the hydrophilic and hydrophobic surfaces, the value of  $\theta_b$  reduces with reducing  $r$  whereas for the superhydrophobic surface the value of  $\theta_b$  increases with reducing  $r$ . This latter observation appears to call into question a “line tension” explanation<sup>17</sup> for the effect, as the line tension should always act to reduce the contact angle for diminishing  $r$ .<sup>24</sup>

TABLE I. Experimental values of the liquid contact angle  $\theta_l$ , the extrapolated bubble contact angle  $\theta_{bi}$  at  $h/R = 0$ , and the measured slope  $\alpha$ —the modeling values correspond to the bubble contact angle  $\theta_{b0}$  (mod.) and the slope  $\alpha$  (mod.).

	$\theta_l$ (exp.)	$\theta_{bi}$ (exp.)	$\alpha$ (exp.)	$\theta_{b0}$ (mod.)	$\alpha$ (mod.)
Black Si	111.9 (1.2)	132.3	-66.4	133.4	-66.8
Teflon	62.1 (2)	106.6	-50.7	108.2	-58.2
PDMS	52.8 (3)	102.4	-56.7	101.4	-57.7
SiO <sub>2</sub>	9.6 (0.6)	90.2	-64.1	90.4	-58.9
Liquid	0	91.9	-170.4	90	-69.6

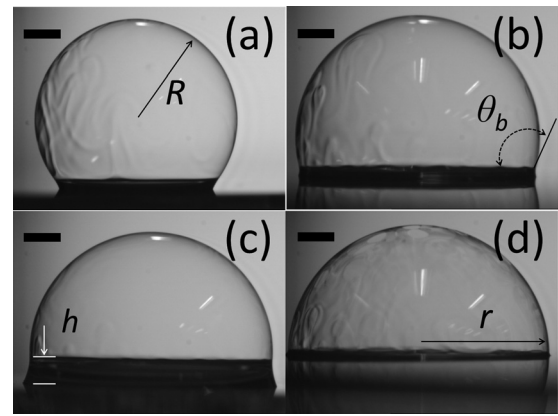


FIG. 2. Sessile bubbles wetting the solid surfaces used in the experiments. (a) Fluorocarbon coated “black silicon,” (b) Teflon<sup>®</sup> AF coated silicon wafer, (c) PDMS, and (d) silicon dioxide. All scale bars = 1 mm. The parameters  $R$  (the curvature radius of the bubble),  $\theta_b$  (the apparent contact angle of the bubble),  $h$  (the liquid film height), and  $r$  (the bubble base radius) are indicated.

Fig. 3(b) plots  $\theta_b$  versus a dimensionless ratio of two lengths  $h/R$  associated with the bubble.<sup>24</sup> The standard deviation of the data points was determined to be  $1.8^\circ$ . For a given surface, the measured value of  $\theta_b$ , corresponding to small values of  $h/R$ , increases from surfaces A to D. Also, as  $h/R$  increases the value of  $\theta_b$  decreases, the data suggesting a near-linear relationship<sup>19</sup> for the four surfaces tested in the

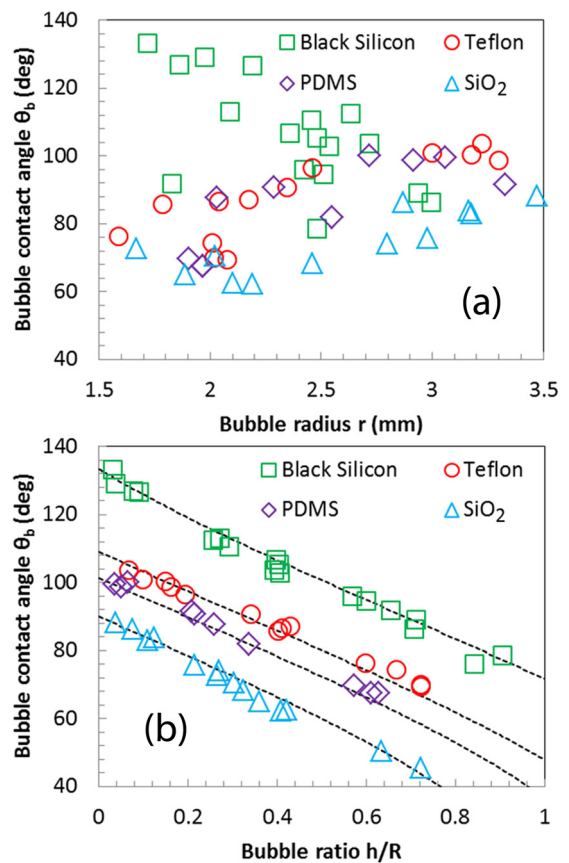


FIG. 3. A plot of the bubble contact angle  $\theta_b$  versus the  $h/R$  ratio for the different surfaces tested. Fluorocarbon coated “black silicon” (open squares), Teflon<sup>®</sup> AF (open circles), PDMS (open diamonds), and silicon dioxide (open triangles). The dashed lines correspond to analytical solutions using Eq. (1) and Ref. 19.



range  $h/R = 0$  to 0.7. A linear fit allows us to calculate the intercept bubble angle  $\theta_{bi}$  using extrapolation. The value of  $\theta_{bi}$  can be compared with the theoretical value of  $\theta_{b0}$  predicted by Eq. (3) as  $h/R \rightarrow 0$  (see Table I) within measurement error; the values compare well for all surfaces tested. A large variation of the bubble contact angle is observed here for the soap bubbles on superhydrophobic surfaces ( $134^\circ \rightarrow 76^\circ$  (Fig. 3(b) open squares)) and on hydrophilic surfaces ( $88.3^\circ \rightarrow 45.4^\circ$  (Fig. 3(b) open triangles)); the observations can be compared to those of Rodrigues *et al.*<sup>17</sup> who observed relatively small variations of the contact angle of a sessile soap bubble resting on a wet ( $\sim 4^\circ$ ) and dry ( $< 10^\circ$ ) surfaces and who invoked the controversial<sup>24</sup> line tension effect to explain their observations.

Equation (3) can explain the measured value of  $\theta_{b0}$  as  $h/R \rightarrow 0$ , i.e., the wetting contact angle of an ideal bubble on a solid surface. However, in order to understand why  $\theta_b$  decreases when  $h/R$  is increased we can implement a model<sup>19</sup> found in the literature for sessile bubbles wetting a solid surface. By using Eq. (3), when  $h/R \rightarrow 0$ ,  $\theta_b \rightarrow \theta_{b0}$ , together with a first-order analytical solution which can be found in Ref. 19, we can compute the apparent contact angle of the bubble  $\theta_b$  as a function of  $h/R$  for the various surfaces by using Eq. (4)

$$\cos \theta_b = \frac{1}{2}(\cos \theta_l - 1) + \frac{h}{R}. \quad (4)$$

The solutions [shown as dashed lines in Fig. 3(b)] correspond well with the experimentally obtained values of  $\theta_b$ . The measured slopes,  $\alpha = d\theta_b/d(h/R)$ , correspond well to those predicted by the model (the near-linear range was taken to be  $h/R = 0 \rightarrow 0.4$ ).

In terms of the bubble film thickness, no measurement of this was performed, but colours<sup>6-8</sup> are visible directly after bubble deposition indicating an initial film thickness in the sub-micrometer range;<sup>24</sup> drainage and evaporation produces black regions<sup>6</sup> (thickness  $< 10$  nm)<sup>24</sup> where presumably the bubble first bursts<sup>20</sup> after a lifetime of seconds to tens of seconds. For an ideal bubble wetting a solid surface, the schematic diagram in Fig. 1(b) indicates an ideal bubble and that the film at the bubble-surface interface is continuous. However, Eqs. (2) and (3) simply imply that a single bubble surface needs to be created inside the bubble, i.e., the extra  $\gamma_l$  term on the *r.h.s.* of Eq. (3). Experimentally, as the sessile bubbles have a finite lifetime ( $\sim$ seconds to tens of seconds), bubble bursting<sup>20</sup> and its outcome can be observed. It was observed that one of two outcomes can be the result from bubble bursting: (i) The bubble bursts leaving a flat film of liquid (of radius  $r$ ) which contracts into a well-defined spherical droplet having a contact angle  $\theta_l$  and (ii) the bubble bursts resulting in a liquid ring of radius  $r$ .<sup>20</sup> This ring is observed to be unstable and either becomes a single droplet, suggesting a continuous film, or breaks-up into smaller droplets having a contact angle  $\theta_l$ , presumably due to a Rayleigh-Plateau instability. Fig. 4 shows these outcomes for small values of  $h/R$ . In general for a large  $h/R$  ratio case (i) is observed. However as  $h/R$  reduces then bursting on a superhydrophobic and hydrophobic surface [Figs. 4(a) and 4(b)] results in a single droplet (indicating the presence of a continuous film at the bubble-solid interface

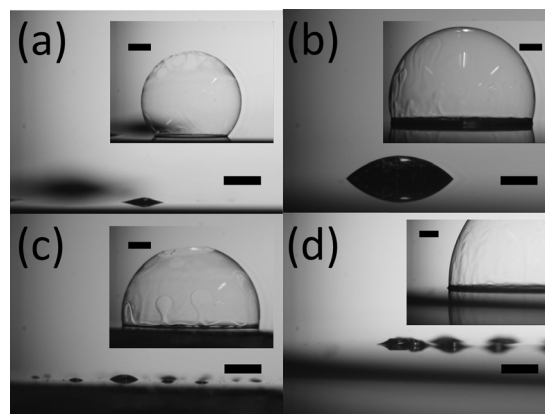


FIG. 4. Sessile bubbles bursting on solid surfaces. (a) Fluorocarbon coated “black silicon,” (b) Teflon<sup>®</sup> AF coated silicon wafer, (c) PDMS, and (d) silicon dioxide. Insets show outcome of bursting. All scale bars = 1 mm.

even for small values of  $h/R$ ) whilst bursting on a more hydrophilic film (PDMS and silicon dioxide) [Figs. 4(c) and 4(d)] results in an unstable liquid ring which stabilizes into a ring of droplets. The value of  $h/R$  for which the transition between forming a droplet after bursting and forming a ring of droplets after bursting was determined to be 0.245 (silicon dioxide) and 0.241 (PDMS). However, prior to bursting, it is important to note that the extrapolated values of  $\theta_{bi}$  correspond very well to those values of  $\theta_{b0}$  computed using Eq. (3).

Finally, let us now consider bubbles resting on the surface of a liquid as shown in Fig. 1(c). Fig. 5 shows sessile bubbles of differing sizes resting on the surface of a liquid composed of the same bubble solution. In general, the Plateau borders are larger—for a given bubble radius—than bubbles of the same size wetting a solid surface. The bubble contact angle is seen to diminish with bubble radius  $r$ . Fig. 6 shows a plot of  $\theta_b$  versus bubble radius  $r$ . As  $r$  is varied from 10 mm to 645  $\mu$ m,  $\theta_b$  changes from 68.4 $^\circ$  to 33.6 $^\circ$ . A plot of  $\theta_b$  versus  $h/R$  reveals itself to be linear [inset to Fig. 6], as is case with bubbles wetting the solid surfaces; the intercept  $\theta_{bi}$  is 91.9 $^\circ$  and the slope  $\alpha$  is  $-170.4$ . The extrapolated intercept value of  $\theta_{bi}$  is close to the 90 $^\circ$  predicted by Eq. (3) assuming  $\theta_l = 0$ . In terms of sessile bubbles wetting a liquid film of the same liquid [Fig. 5], the value of  $\alpha$  predicted by the model,<sup>19</sup> assuming a perfectly hydrophilic surface, does not correspond

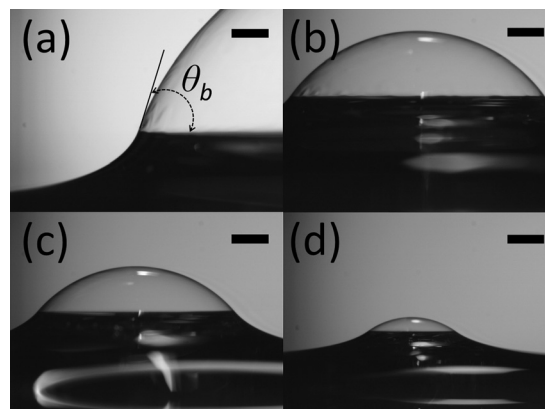


FIG. 5. Sessile bubbles of different base radii wetting a liquid film composed of the bubble solution. The contact angle of the bubble  $\theta_b$  diminishes as the bubble base radius reduces (a)  $\rightarrow$  (d). All scale bars = 1 mm.

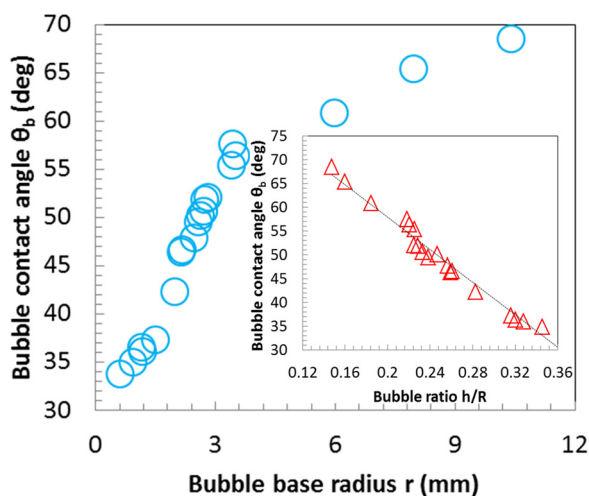


FIG. 6. A plot of the experimental bubble contact angle  $\theta_b$  versus the bubble base radius  $r$ . Inset shows the straight line obtained when plotting the bubble contact angle  $\theta_b$  versus the  $h/R$  ratio.

well with the experimental data for sessile bubbles wetting a liquid surface. Note that the experimental data for a hydrophilic surface given in Ref. 19 gives a slope  $\alpha$  of  $\sim 90$ . Although the data here suggest a near-linear relationship as is the case with bubbles wetting a solid surface [Fig. 3(b)], the measured slope corresponds is 2.4 times the value of  $\alpha$  predicted for wetting on a perfectly hydrophilic surface. In other words, the data indicate that wetting behavior of a bubble on a perfectly hydrophilic solid surface is not the same as wetting of a bubble on a liquid film of the same liquid.

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