

## Modeling the evolution of surface nanobubbles

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Surface nanobubbles are gaseous formations on solid-liquid interfaces. They are interesting in that their lifetime can reach several days, although initially it was assumed only a few nanoseconds. We built a mathematical and computer model of the nanobubble and performed a series of simulations to see how the nanobubble behaves when various external factors change.

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### 1. Introduction

Gaseous formations – bubbles often appear as a result of various physical processes in a liquid. About 25 years ago, it was found that in addition to visible bubbles, nanobubbles invisible human eyes also appear in the liquid. In addition to spherical nanobubbles in bulk, surface nanobubbles also arise, which are located at the liquid-solid interface. It is surprising that these bubbles have long lifetime [1–13]. They can last several hours, in some cases several days. The initially predicted approximate lifetimes of nanobubbles described in [4] are orders of magnitude shorter than those practically observed [5].

Surface nanobubbles in most cases form a hemispherical shape. Some experiments [8, 14–18] have also shown that the shape of a nanobubble can be spherical with a small flat area at the top. It can also be seen from [3, 6, 7] that the shape of a nanobubble is often not always ideally spherical. Due to their size, nanobubbles have unique properties that may improve many physical, chemical and biological processes [5, 19, 20]. The presence of bubbles on a hydrophobic solid contributes to a change in the behavior of the liquid near the liquid-solid interface.

As for modern modeling methods, the most useful one is the molecular dynamics [3], since it allows one to obtain the most accurate description of processes occurring in objects that do not exceed hundreds of nanometers in size, as well as the simulated nanobubbles. But this method also has its limitation; for example, significantly complex calculations, which, on a regular computer, can take a huge amount of time. Therefore, for this method, various highly efficient algorithms are used which make it possible to reduce the number of computer calculations by an order of magnitude [1–3]. Analogously to [21], we construct two-dimensional molecular dynamics model which is an approximation only but, nevertheless, allows one to determine the properties of real physical system.

### 2. Description of the model

The simulated nanobubble consists of many gas molecules. In this model, we will assume that the gas is ideal. All collisions of a gas molecule with the upper boundary of the nanobubble are absolutely elastic, without energy loss.

The model is a two-dimensional section of the bubble. The lower boundary is the boundary of the rigid body, specified by the function  $l(x) = 0$ , where  $x$  is the horizontal coordinate. The upper boundary – the “roof” of the nanobubble, is introduced as a set of points, i.e. pairs of numbers  $(x, y)$ , where  $x$  is the point horizontal coordinate,  $y$  is the vertical coordinate. The initial position of the upper boundary is given by the function:

$$h(x) = \frac{4h_s x(w_s - x)}{w_s^2},$$

where  $h_s$  is the initial value of the nanobubble height in nanometers,  $w_s$  is the initial nanobubble diameter in nanometers,  $x$  is the horizontal coordinate, and  $h(x)$  is the nanobubble height at point  $x$ .

There are  $N$  gas molecules inside the bubble. Each molecule has its own normalized direction vector, the coordinate of its location in the system, velocity modulus and mass. The modulus of the velocity  $v$  of each molecule is chosen on the basis of the Maxwell distribution function for the absolute values of the velocities:

$$F(v) = 4\pi \left( \frac{m_0}{2\pi kT} \right)^{3/2} \cdot v^2 \cdot \exp \left( -\frac{m_0 v^2}{2kT} \right),$$

where  $T$  is the temperature,  $k = 1.380649 \cdot 10^{-23} \text{ JK}^{-1}$  is the Boltzmann constant,  $m_0$  is the mass of a gas molecule.

The number of gas molecules in a nanobubble:  $N = nV$ , where  $n$  is the concentration of molecules, and

$$V = \frac{\pi h_s(3w_s^2 + 4h_s^2)}{24}$$

is the volume of a nanobubble (based on work [3]).

In turn, the concentration can be calculated using the following formula

$$n = \frac{\rho}{m},$$

where  $\rho$  is the density of the gas, and  $m$  is the mass of the gas molecule.

As a result, we come to the following formula:

$$N = \frac{\rho}{m} \frac{\pi h_s(3w_s^2 + 4h_s^2)}{24}$$

As for the dynamics for each molecule, we assume that colliding with the bubble boundary, the molecule does not lose velocity and the velocity vector is mirrored:

$$\vec{b} = \vec{a} - 2\vec{n} \frac{(\vec{n}, \vec{a})}{(\vec{n}, \vec{n})},$$

where  $\vec{a}$  is the direction vector of the gas molecule,  $\vec{n}$  is the normal vector at the point of collision of the molecule and the upper boundary of the bubble,  $\vec{b}$  is the reflected vector of the molecule.

At each moment of time, three main forces act on each point of the upper boundary of the bubble:  $F_p$  is the pressure force which is directed vertically downward;  $F_m$  is caused by elastic collision of gas molecules;  $F_a$  is the force of attraction of molecules. The pressure force is as follows:

$$F_p = \rho g H S,$$

where  $\rho$  is the density of the liquid,  $H$  is the height of the layer of the simulated liquid above the liquid solid interface with the nanobubbles,  $S$  is the unit of area,  $g = 9.81 \frac{\text{m}}{\text{s}^2}$  is the acceleration due to gravity. The force caused by elastic collision of gas molecules with the border:

$$\vec{F}_m = \sum_{j=1}^{n_c} \vec{F}_{m_j},$$

where  $n_c$  is the number of molecules which collide with the current upper boundary point, and:

$$F_{m_j} = \frac{d\vec{p}_j}{dt},$$

The third force is as follows:

$$F_a = \alpha l,$$

where  $\alpha$  is the liquid surface tension coefficient,  $l$  – distance between molecules.

### 3. Results

#### 3.1. Behavior of a nanobubble over flat substrate

Firstly, let us simulate the bubble under normal conditions:

$$T = 300 \text{ K}, \quad H = 0.2 \text{ m},$$

$$\rho_w = 1000 \text{ kg} \cdot \text{m}^{-3}, \quad \alpha = 7.2 \cdot 10^{-2} \text{ H} \cdot \text{m}^{-1}.$$

Let us choose the initial width and height of the simulated nanobubble in the following way:

$$h_s = 20 \cdot 10^{-9} \text{ m},$$

$$w_s = 200 \cdot 10^{-9} \text{ m}.$$

As for the gas molecules, we consider two cases: nitrogen molecules and carbon dioxide molecules. The mass of one nitrogen molecule and the density is as follows:

$$m = 4.67 \cdot 10^{-26} \text{ kg}, \quad \rho = 1.1649 \text{ kg} \cdot \text{m}^{-3}.$$

We simulate the state of a nanobubble in 20 microseconds and collect the results in Table 1, where  $h$  is the height of the nanobubble and  $s$  is the volume.

The same simulation results for the carbon dioxide instead of the nitrogen are collected in Table 2.

TABLE 1. Parameters of nanobubble with nitrogen in water in the equilibrium state

	$T = 300 \text{ K}$	$T = 330 \text{ K}$	$T = 380 \text{ K}$
$H = 0.2 \text{ m}$	$h = 12 \text{ nm}, s = 8000 \text{ nm}^3$	$h = 13 \text{ nm}, s = 8500 \text{ nm}^3$	$h = 15 \text{ nm}, s = 10000 \text{ nm}^3$
$H = 0.4 \text{ m}$	$h = 8 \text{ nm}, s = 4200 \text{ nm}^3$	$h = 8.5 \text{ nm}, s = 5000 \text{ nm}^3$	$h = 9 \text{ nm}, s = 6000 \text{ nm}^3$

TABLE 2. Parameters of a nanobubble with carbon dioxide in water in the equilibrium state

	$T = 300 \text{ K}$	$T = 330 \text{ K}$	$T = 380 \text{ K}$
$H = 0.2 \text{ m}$	$h = 11 \text{ nm}, s = 6800 \text{ nm}^3$	$h = 12 \text{ nm}, s = 7200 \text{ nm}^3$	$h = 14 \text{ nm}, s = 8800 \text{ nm}^3$
$H = 0.4 \text{ m}$	$h = 8 \text{ nm}, s = 4500 \text{ nm}^3$	$h = 8.5 \text{ nm}, s = 4650 \text{ nm}^3$	$h = 9 \text{ nm}, s = 5200 \text{ nm}^3$

The same simulations were performed for ethanol instead of water. Parameters of the nanobubble are shown in Table 3 and Table 4.

TABLE 3. Parameters of a nanobubble with nitrogen in ethanol in the equilibrium state

	$T = 300 \text{ K}$	$T = 330 \text{ K}$	$T = 380 \text{ K}$
$H = 0.2 \text{ m}$	$h = 11 \text{ nm}, s = 6800 \text{ nm}^3$	$h = 12 \text{ nm}, s = 7200 \text{ nm}^3$	$h = 14 \text{ nm}, s = 8800 \text{ nm}^3$
$H = 0.4 \text{ m}$	$h = 8 \text{ nm}, s = 4500 \text{ nm}^3$	$h = 8.5 \text{ nm}, s = 4650 \text{ nm}^3$	$h = 9 \text{ nm}, s = 5200 \text{ nm}^3$

TABLE 4. Parameters of a nanobubble with the carbon dioxide in ethanol in the equilibrium state

	$T = 300 \text{ K}$	$T = 330 \text{ K}$	$T = 380 \text{ K}$
$H = 0.2 \text{ m}$	$h = 14.9 \text{ nm}, s = 9200 \text{ nm}^3$	$h = 15.5 \text{ nm}, s = 9400 \text{ nm}^3$	$h = 16.3 \text{ nm}, s = 9640 \text{ nm}^3$
$H = 0.4 \text{ m}$	$h = 8.6 \text{ nm}, s = 8100 \text{ nm}^3$	$h = 8.8 \text{ nm}, s = 8250 \text{ nm}^3$	$h = 9.1 \text{ nm}, s = 8400 \text{ nm}^3$

The tables show obvious dependencies: as the temperature in both simulated liquids increases, the nanobubble, as expected, increases its volume due to the increase in the internal energy of the gas molecules. And with a relatively small increase in depth in both simulated fluids, the nanobubble volume significantly decreases due to an increase in external pressure.

### 3.2. Behavior of nanobubble over rough substrate

In practice, a solid border is almost never perfectly flat. To see what will happen to the nanobubble with “rough” solid boundary, we change the function  $l(x) = 0$ , which defines the lower bound, to a piecewise linear function  $g(x)$ , which is presented below:

$$g(x) = \begin{cases} 0, & x \geq 150; \\ -5, & 50 < x < 150; \\ 0, & x \leq 50. \end{cases}$$

The parameters of the nanobubble are the same as in the simulation with water above: the temperature  $T = 300 \text{ K}$ , depth  $H = 0.2 \text{ m}$ . The modeled gas molecules are nitrogen and carbon dioxide. The form of nanobubble is shown in the Figs. 1,2.

The simulation results turned out to be quite interesting. The height of the bubble has indeed decreased, but the diameter increased significantly. This can be explained by the fact that the surface tension tries to reduce the contact angle, despite the fact that there is no interaction between the substrate and the liquid. But in this model, we assumed that the substrate is hydrophobic, which means that the external contact angle between the liquid and the substrate is always greater than  $\pi/2$ ; therefore, the only way to reduce this angle is to increase the nanobubble diameter. The figures also show that the bubble has become less streamlined. Probably, such transformations occurred precisely because the molecular motion changed sufficiently and after collision with the lower boundary the molecule cannot get into the left or the right part of the bubble because it is blocked by the wall of the lower boundary. This assumption is confirmed by atomic force microscope measurements carried out in [3, 6]. An image of a nanobubble from [3] is shown in Fig. 3.

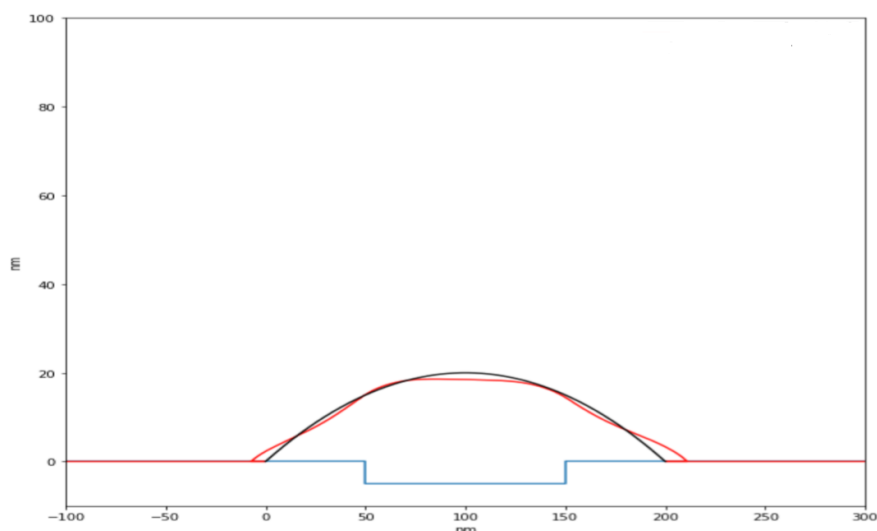


FIG. 1. Changing a nanobubble with nitrogen over the rough substrate. Black line is the initial form of the nanobubble, red line corresponds to the nanobubble in the equilibrium state

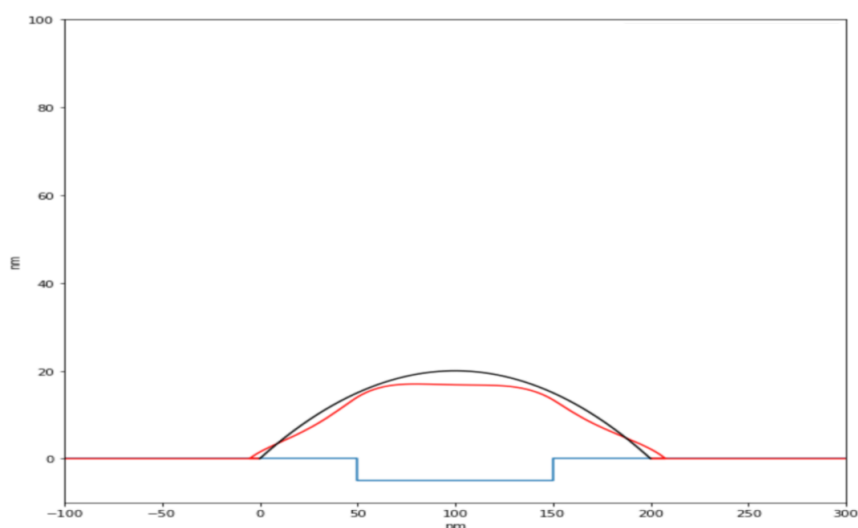


FIG. 2. Changing a nanobubble with carbon dioxide over the rough substrate. Black line is the initial form of the nanobubble, red line corresponds to the nanobubble in the equilibrium state

Figure 3 shows that the shape of the nanobubble is not just spherical, but the same as in Fig. 1,2. The base of the nanobubble is flat, and closer to the point with the maximum height, a small, almost flat area appears at the bubble. Moreover, proceeding from Fig. 3, it cannot be said that this local maximum occurs over the entire cross section of the nanobubble. In our model, the local maximum are almost symmetric, since the notch in the substrate was symmetrical and exactly in the center of the nanobubble. This suggests that the substrate is often not perfectly flat, and the shape of the nanobubble does not always represent an ideal hemisphere and depends very much on the shape of the substrate. Fig. 4 shows a possible reason for the appearance of two maximum at the liquid-gas interface.

### 3.3. Heating and cooling the substrate and liquid

We add some parameters to our model:

1.  $T(x)$  – function of substrate temperature at point  $x$ ;
2.  $T_w$  – water temperature.

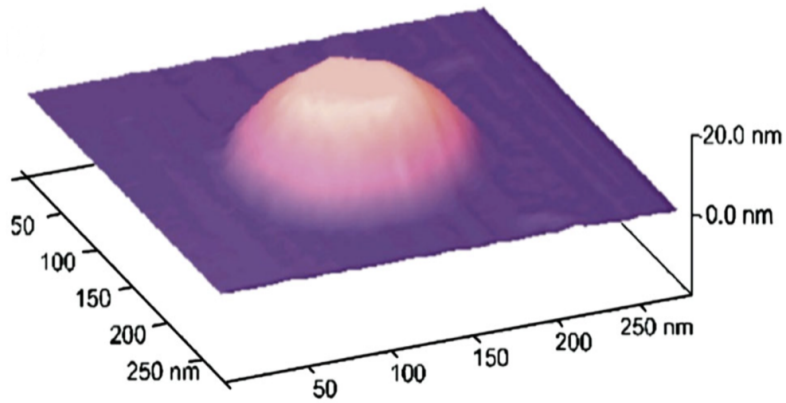


FIG. 3. Atomic force microscopy image of a nanobubble in work [3]

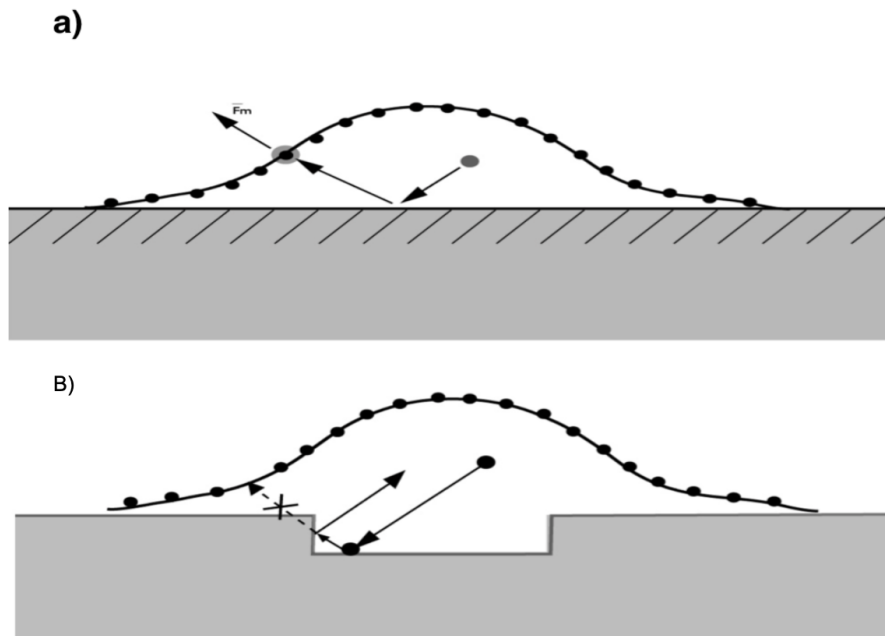


FIG. 4. Demonstration of the movement of a molecule in the case of a) an flat substrate, b) a rough substrate

We will assume that a gas molecule, when struck with the boundary of a liquid, receives a velocity:

$$v = \sqrt{\frac{3kT_w}{m_0}},$$

and upon collision with the boundary of a rigid body, the velocity is obtained:

$$v = \sqrt{\frac{3kT(x)}{m_0}}.$$

Let  $T_w = 300$  K and set the  $T(x)$  function like:

$$T(x) = \begin{cases} 300, & x \leq 150; \\ 350, & 150 < x < 300; \\ 300, & x \geq 300. \end{cases}$$

Figure 5 clearly shows a tendency to move to the right and an increase in the height of the nanobubble to the right, although the left part is in no hurry to move to the right, this is because the nanobubble is in a near-equilibrium position and when gas molecules collide with a heated region, the internal energy increases and this energy enough to maintain the current volume of the nanobubble until the bubble expands to the right even more.

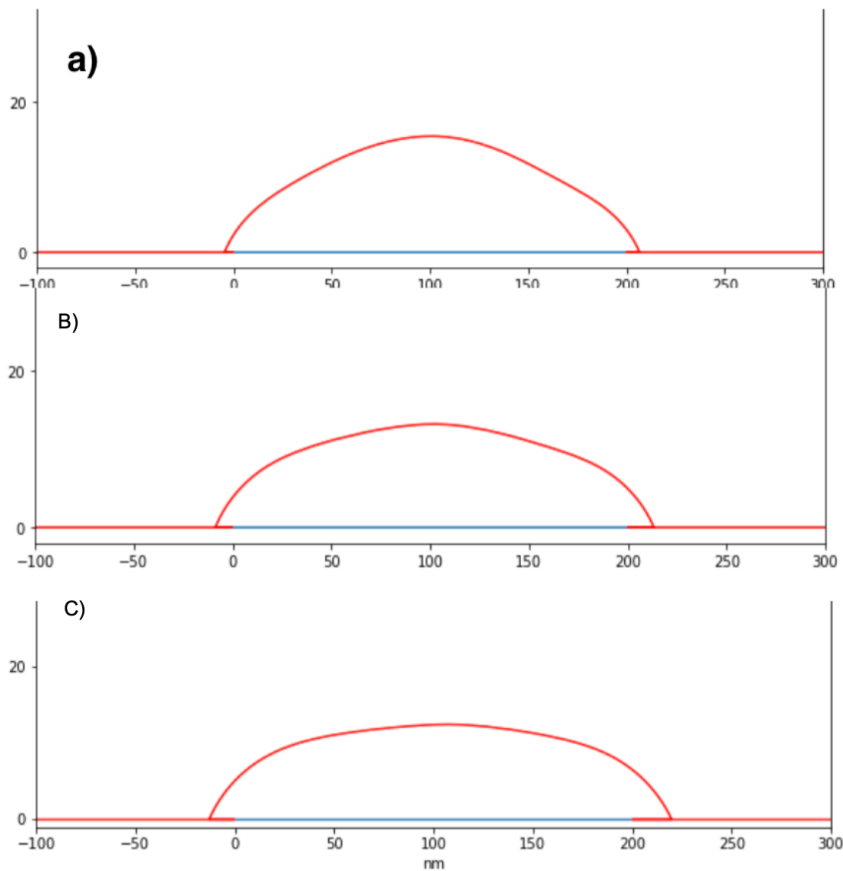


FIG. 5. Changes in the shape and position of a nanobubble over time a)  $t = 5 \mu s$ , b)  $t = 10 \mu s$ , c)  $t = 15 \mu s$

Now construct the temperature gradient, which at the point  $x = 100$  takes the highest temperature, and decreases in both directions according to the following formula:

$$T(x) = T_{\max} - \gamma|100 - x|,$$

where  $\gamma$  temperature gradient  $K \cdot nm^{-1}$ .

Let  $T_{\max} = 350$  K and  $\gamma = 0.3 K \cdot nm^{-1}$ . Fig. 6 shows the expected formation of the elongated tip of the nanobubble, as well as the absence of lateral movement. The diameter only increased by about 15 nm due to the fact that the nanobubble tends to the equilibrium position.

The data in Table 5 show the height of the nanobubble at various parameters, provided that  $T_{\max} = 350$  K.

Instead of  $T_{\max}$ , put  $T_{\min}$ . The coldest point will be at  $x = 100$ . In this case, the bubble at the point  $x = 100$  will have a lower height compared to other points and a tendency for the bubble to burst by 2 will be seen. Let's try to find

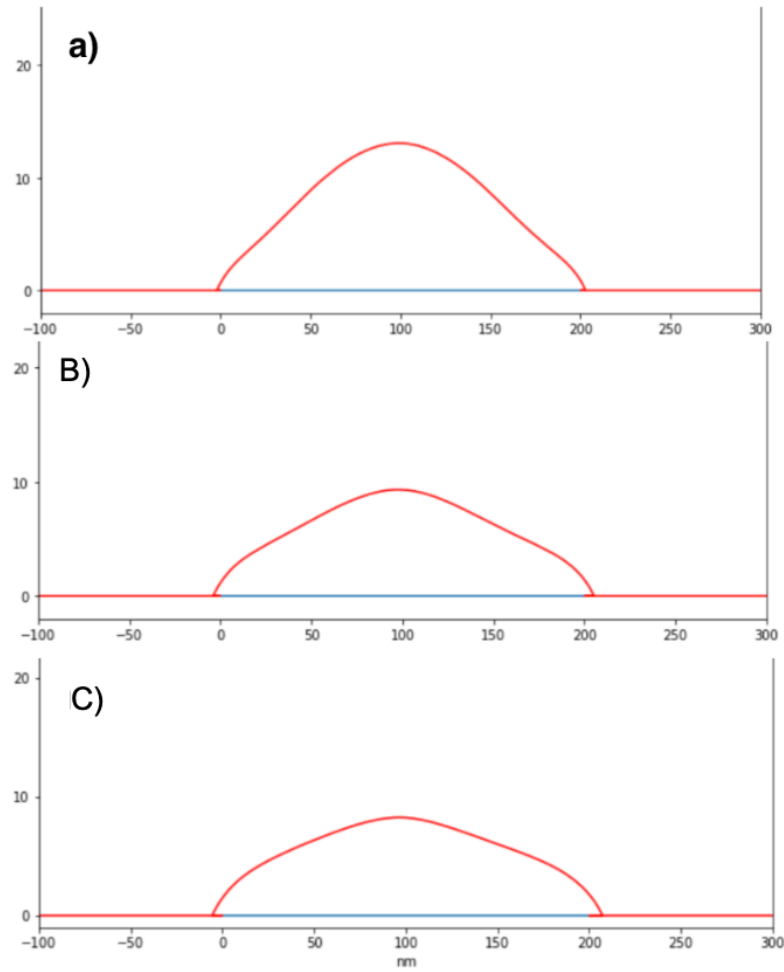


FIG. 6. Change in the shape and position of a nanobubble with a heated substrate over time a)  $t = 5 \mu\text{s}$ , b)  $t = 10 \mu\text{s}$ , c)  $t = 15 \mu\text{s}$

TABLE 5. Maximum height of a nanobubble in water at different temperature gradients and water temperature  $T_w$

	$\gamma = 0.3 \text{ K}\cdot\text{nm}^{-1}$	$\gamma = 0.1 \text{ K}\cdot\text{nm}^{-1}$	$\gamma = 0.05 \text{ K}\cdot\text{nm}^{-1}$
$T_w = 300 \text{ K}$	$h = 9.2 \text{ nm}$	$h = 11.3 \text{ nm}$	$h = 12.4 \text{ nm}$
$T_w = 330 \text{ K}$	$h = 11.5 \text{ nm}$	$h = 12.4 \text{ nm}$	$h = 13.1 \text{ nm}$
$T_w = 350 \text{ K}$	$h = 12.7 \text{ nm}$	$h = 13.4 \text{ nm}$	$h = 13.8 \text{ nm}$

the critical parameters. Let us increase the depth of the liquid column to  $H = 0.4 \text{ m}$  and change  $T(x)$  function in the following way:

$$T(x) = T_{\min} + \gamma|100 - x|,$$

$$T_{\min} = 260 \text{ K}, \gamma = 0.5 \text{ K} \cdot \text{nm}^{-1}$$

Simulation data shown on Table 6

What happened to the nanobubble in the case of  $H = 1.5$  is shown in Fig. 7.

the nanobubble on the verge of rupture after  $13 \mu\text{s}$  does not split in half, but moves in the direction where there are more gas molecules present. From the figure above, it follows that surface tension plays an important role in the formation of a nanobubble. In general, this behavior of a nanobubble can be justified by the fact that when a molecule enters the largest of the forming nanobubbles, it is more difficult for it to get out of it and enter another. Therefore, with sufficient surface tension, it is only a matter of time before one of the forming nanobubbles overflows into another.

TABLE 6. Change in the maximum height at the assumed point of rupture  $x = 100$  over time at different depths of the liquid. Temperature gradient  $\gamma = 0.5 \text{ K}\cdot\text{nm}^{-1}$ ,  $T_{\min} = 260 \text{ K}$ ,  $T_w = 300 \text{ K}$

	$t = 5 \mu\text{s}$	$t = 10 \mu\text{s}$	$t = 15 \mu\text{s}$
$H = 0.4 \text{ m}$	$h = 5.2 \text{ nm}$	$h = 3.6 \text{ nm}$	$h = 3.4 \text{ nm}$
$H = 0.6 \text{ m}$	$h = 4.5 \text{ nm}$	$h = 3.1 \text{ nm}$	$h = 2.88 \text{ nm}$
$H = 1 \text{ m}$	$h = 2.19 \text{ nm}$	$h = 1.5 \text{ nm}$	$h = 1.44 \text{ nm}$
$H = 1.25 \text{ m}$	$h = 1.3 \text{ nm}$	$h = 1.22 \text{ nm}$	$h = 1.14 \text{ nm}$
$H = 1.35 \text{ m}$	$h = 0.9 \text{ nm}$	$h = 0.95 \text{ nm}$	$h = 1.11 \text{ nm}$
$H = 1.45 \text{ m}$	$h = 0.92 \text{ nm}$	$h = 0.97 \text{ nm}$	$h = 0.6 \text{ nm}$
$H = 1.5 \text{ m}$	$h = 0.8 \text{ nm}$	$h = 0.88 \text{ nm}$	$h = 0 \text{ nm}$

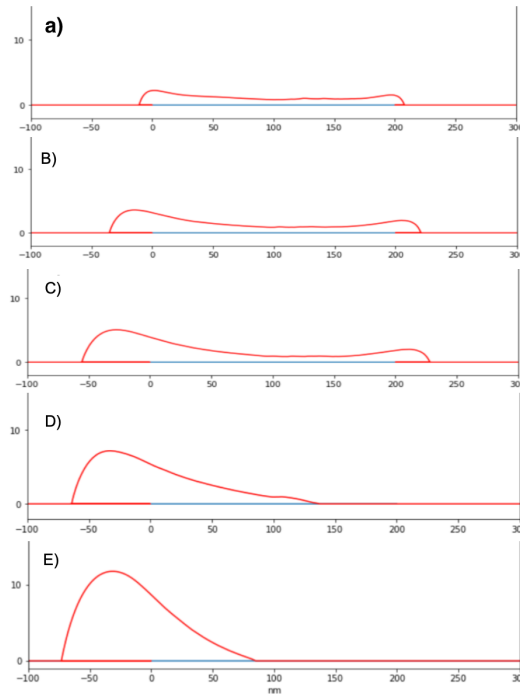


FIG. 7. change in a nanobubble at a depth of  $H = 1.5 \text{ m}$  with a temperature gradient. a)  $5 \mu\text{s}$ , b)  $10 \mu\text{s}$ , c)  $13 \mu\text{s}$ , d)  $14 \mu\text{s}$ , e)  $15 \mu\text{s}$

At  $H = 1.35 \text{ m}$ , the height at the supposed rupture point does not decrease with time, but, on the contrary, increases, which indicates that the bubble began to completely shift to the left.

The table shows that up to a depth of  $1 \text{ m}$ , the dependence is almost linear. After that, the altitude remains approximately at the same level and a sharp jump occurs at approximately  $H = 1.47$ . At this depth, the bubble overflows completely to the left side, forming a new one. There was no break in 2 nanobubbles.

Let  $H = 0.4 \text{ m}$ ,  $T_{\min} = 260 \text{ K}$ ,  $T_w = 300 \text{ K}$ . We will change the parameter of the temperature gradient. The simulation results are in Table 7.

It was not possible to rupture the bubble again. It can be seen from Table 7 that the closest gradient to the separation of the bubble into two is the value:  $\gamma 0.5 \text{ K}\cdot\text{nm}^{-1}$ . With a gradient greater than  $0.5$ , the temperature rises rapidly and the center of the bubble sags less and less in height due to the rapid growth of internal energy inside the bubble. With a gradient of less than  $0.5$ , the temperature, on the contrary, grows more slowly and, accordingly, there is almost no movement to the sides, and therefore more energy is spent on maintaining the current volume of the nanobubble.



TABLE 7. Change in the maximum height at the assumed break point  $x = 100$  over time for various parameters of the temperature gradient.  $H = 0.4$  m,  $T_{\min} = 260$  K,  $T_w = 300$  K

	$t = 5 \mu s$	$t = 10 \mu s$	$t = 15 \mu s$
$\gamma = 1 \text{ K}\cdot\text{nm}^{-1}$	$h = 8.7 \text{ nm}$	$h = 4.0 \text{ nm}$	$h = 3.9 \text{ nm}$
$\gamma = 0.8 \text{ K}\cdot\text{nm}^{-1}$	$h = 9.4 \text{ nm}$	$h = 4.1 \text{ nm}$	$h = 3.8 \text{ nm}$
$\gamma = 0.5 \text{ K}\cdot\text{nm}^{-1}$	$h = 5.2 \text{ nm}$	$h = 3.6 \text{ nm}$	$h = 3.4 \text{ nm}$
$\gamma = 0.3 \text{ K}\cdot\text{nm}^{-1}$	$h = 8.0 \text{ nm}$	$h = 3.8 \text{ nm}$	$h = 5.65 \text{ nm}$
$\gamma = 0.1 \text{ K}\cdot\text{nm}^{-1}$	$h = 7.9 \text{ nm}$	$h = 6.1 \text{ nm}$	$h = 4.3 \text{ nm}$
$\gamma = 0.05 \text{ K}\cdot\text{nm}^{-1}$	$h = 7.95 \text{ nm}$	$h = 6.15 \text{ nm}$	$h = 5.2 \text{ nm}$

It makes no sense to consider the situation at  $T_{\min} < 260$  K, since this temperature already is a bit lower than freezing point of water and, therefore, it can be concluded that a nanobubble with current parameters at any temperature gradient in water cannot be divided into two parts.

#### 4. Conclusion

In this work, using the molecular dynamics method in combination with highly efficient algorithms a model of a surface nanobubbles was constructed. The model shows how the shape and behavior of the bubble changes over time. The models include various parameters such as liquid and gas temperatures, initial nanobubble shape, solid boundary shape, temperature gradient, liquid surface tension, and other important factors influencing nanobubble evolution. The model demonstrates the stable shape of the nanobubble and establishes the relationship between liquid, gas and the size of the nanobubble in a stable shape. The behavior of a nanobubble with a more real, rough surface of the substrate is shown, which was confirmed by studies carried out years earlier. When heating and cooling different parts of the substrate and liquid, the motion of the nanobubble was substantiated. It is shown that, with the simulated parameters, it is impossible to divide the bubble into two parts with such small sizes and temperature gradient in water: the tension force plays a significant role.

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