Fluid instabilities in the birth and death of antibubbles

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Abstract. Antibubbles have been produced and studied with the help of a high-speed camera. An antibubble is defined as a fluid object constituted by a thin air shell surrounding a liquid and surrounded by the same liquid. Images reveal some key physical processes and fluid instabilities which take place when an antibubble forms and dies. The collapsing speed of the air film has been measured. Culik’s theory does not apply. A new mechanism has been introduced.

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1. Introduction

Soap bubbles are familiar objects. Everybody has spent a large number of hours making soap bubbles and watching the amazing colours of these spheres carried away by air turbulence. From a physical point of view, a bubble is a spherical film of liquid surrounding an air pocket. The Laplace law, which relates the radius of curvature $R$ and the pressure difference $\Delta p$ across a thin film, reads

$$\Delta p = \frac{4\gamma}{R},$$

where $\gamma$ is the surface tension [1].
A less common physical entity is an antibubble. An antibubble is a spherical air shell surrounding a liquid. Hughes and Hughes [2] reported the existence of such an unusual object. They found that a thin film of air can enclose a liquid globule in a surrounding liquid. The liquids forming the globule and the surrounding liquid are the same (figure 1). Connett [3] coined the term antibubble for this liquid–gas entity.

Although the physics of bubbles is well established [5], very few studies have been conducted on antibubbles. Recently, antibubbles attracted our attention and we succeeded in producing them. We have shown that they are stable objects with a lifetime of about 2 min [4].

Both entities, bubbles and antibubbles, are composed of two surfactant layers separated by a distance \( \varepsilon \). The inner radius \( R \) is about \( 10^4 \) times larger than the thickness \( \varepsilon \) of the liquid (bubble) or the air shell (antibubble). Despite their apparently similar structures, bubbles and antibubbles have very different properties. This is mainly due to the compressibility of the air shell. Consequently, antibubbles are more sensitive to external variations of pressure.

Antibubbles seem to be a new way of creating an interface between two miscible fluids since the air expulsion is very quick. Indeed, antibubbles can be seen as giant vesicles or as giant capsules. In this paper, we describe the birth and death of an antibubble. In other words, the collapse dynamics of an air shell is studied. Fluid instabilities between the inner and outer fluids have been identified.

2. Birth of antibubbles

Antibubbles are created by using a mixture of water and dishwashing soap (volume fraction is 0.1%). Surfactant molecules provide some elasticity to the fluid interfaces. Moreover, the air–liquid interface is composed of oriented surfactant molecules. This particular structure ensures air-shell stability [4].

The experimental set-up that we used consists of a \( 20 \times 20 \times 200 \text{ cm}^3 \) parallelepiped cell. The cell is open on the top and is completely filled with the liquid mixture. The cell is placed in a larger tank to prevent leaks. Moreover, a continuous flow is maintained with a pump to keep the liquid surface clean, namely free from any remaining bubbles. With the help of a bécher, a small amount of the same liquid is gently poured over the surface. A globule of liquid is first formed at the liquid surface. Such a globule does not immediately coalesce with
Figure 2. (a) Liquid jet breaking up below the liquid surface. A number of forming and newly born antibubbles can be seen. (b) Sketch of the antibubble formation. A bécher gently drops the liquid at the surface. The formed globule is advected down by the flow imposed by the bécher. The electric connection has not been represented but it can be seen on the right in (a).

The antibubble formation is simplified when a copper wire is used between the flowing liquid and the vessel. Indeed, the antibubble forms a kind of electrical capacitor with concentric electrodes. If a difference of potential exists, the air film becomes unstable. The electrical connection prevents any electrical potential difference due to triboelectric effects.

Figure 2 shows the formation of several antibubbles. The bécher is out of the picture but the electrical connection can be seen on the right. The liquid flow from the bécher passes through the air–liquid interface and forms a liquid jet beneath the free surface. This streamer breaks up due to the Rayleigh–Plateau instability. This can be observed just beneath the surface: the jet forms a rosary of droplets. This instability is responsible for the antibubble creation.

In pictures (e.g. figure 2), the antibubbles seem to have a thick black border. This is an optical effect due to total reflection at the liquid–air interface. The air-shell thickness has been estimated from the value of the small air bubble created from the air shell when it collapses. This thickness is approximately 3 μm. Another way of determining the air thickness is to measure the speed limit when the antibubbles rise towards the surface. This speed depends on the density difference. The air fraction can then be calculated and ε estimated. These measurements corroborate the first estimations.

Circular interference fringes can be observed in some cases. Those fringes seem to be similar to Newton rings [7]. The centre of those concentric fringes is located a little above the equatorial plane of the antibubble. Actually, a slight deformation can be seen at the top of the antibubble: the air shell is thinner at the bottom. This is due to buoyancy. The symmetry of the antibubble is cylindrical, exactly like soap bubbles. Note that in the case of a bubble, the liquid shell is largest at the bottom due to gravity and to the subsequent free drainage.

It is also possible to create antibubbles with an inner fluid different from the surrounding liquid. For example, by dissolving some salt in the bécher, heavy antibubbles can be created. They sink and pop up at the bottom of the vessel (if the depth is not so large, see below).
By adjusting the quantity of salt, one could also have floating antibubbles when gravity and buoyancy are equal. The lifetime of the floating antibubbles is much longer than that of antibubbles sticking at the surface. They could last several minutes before popping.

3. Death of antibubbles

Buoyancy will also bring antibubbles towards the surface where they stick before naturally pop up or they may pop up when sinking. The physics involved in both cases is quite different. Such pop-up events provide an additional view of the phenomenon.

3.1. Collapse of the air shell

In order to study the popping of antibubbles, we artificially pierced the air shell with a pin. A high-speed video camera (Redlake MotionPro 2000) has been used for recording the dying antibubbles. A 1000 W incandescent lamp has been placed behind the vessel at the same height as the camera. Movies have been recorded at a frequency of 1000 frames per second.

The collapse of the air shell is quite spectacular. Figure 3 presents six successive stages of a popping antibubble. The air shell is first pierced and a circular propagating front appears. The fluid contained in the antibubble is suddenly put into contact with the surrounding liquid. Afterwards, the air film shrinks near the initial popping but in the opposite direction. In the fifth picture, we can see that the air film undulates and is rippled. Finally, the air film disintegrates into air pockets. The total collapse time is shorter than 50 ms. To sum up, the air shell breaks at a certain point and the air film quickly collapses in a direction opposite to the initial popping. This is quite similar to what is observed with popping bubbles.

A sketch of this process is also proposed in figure 3. The arrows indicate the direction of the propagating front velocity. The apparent aperture $h$ (see figure 3) has been measured as a function of time. Due to total reflection at the edge of the antibubble, some data are missing. The angle $\alpha$ is defined as the angle between the zenith point with respect to the popping point and the propagating front. The speed $v$ of the front is then given by

$$v = R \frac{d\alpha}{dt}.$$  

(2)

In figure 4, both the angle $\alpha$ and the time derivative $d\alpha/dt$ are reported with respect to time. The speed is maximum at the beginning; the slope $\alpha(t)$ is the largest there. The speed decreases until the total collapse of the air shell. This decrease is attributed to some energy dissipation in the formation of air pocket.

The film opening reminds the dewetting of a surface. Culick [8, 9] considers the growth of a hole produced in the middle of a soap film. The model is based on the inertial movement of the liquid edge surrounding the hole. Indeed, the edge can be represented by a cylinder that becomes heavier and heavier with time since the edge accumulates the liquid. This theory cannot be applied here since the film is made of air. The mass of the edge can be neglected. Moreover, the speed does not remain constant during film collapse (Culik’s first hypothesis). By analogy to the dewetting, the air film collapses along a front of wetting. Two forces act during the collapse: (i) the superficial tension pulls the air in a direction opposite to the piercing point and (ii) the drag force due to fluid friction. The data obtained for the speed $R d\alpha/dt$ suggests that the movement of the air film uniformly slows down by the drag force.

Figure 3. Six successive stages of the air shell collapse of an antibubble (from top left to top right). The bottom of the figure depicts a sketch of the dewetting of the inner globule of an antibubble. The arrows indicate the direction of propagation of the dewetting front. The relevant parameters are \( h \) and \( \alpha \).

The inner part of the antibubbles can be coloured by adding methylene blue in the b´echer. This is quite useful because the fluid motion can be recorded, leaving the density nearly unchanged. Figure 5 illustrates different successive steps of the popping of an antibubble at the surface of the liquid. Once the air shell is pierced on the left-hand side, both liquids (inside and outside of the antibubble) are suddenly in contact without mixing. Indeed, the blue globule seems to keep its shape until the air bubble is expelled from the globule from the right-hand side. This motion creates a brief shock on the globule. Subsequently, an instability is produced in the globule. A vortex is created at the initial point of breaking. This vortex always appears in a direction opposite to the air-bubble ejection. This well-known process simply follows momentum conservation.

The first animation of figure 7 shows the popping of an antibubble below the surface when its speed is very low. A small amount of salt has been added to counterbalance the
Figure 4. Evolution of the aperture angle $\alpha$ with respect to time. The time derivative of $\alpha$ is also represented (right scale). The solid curves are a guide to the eye.

Figure 5. Successive pictures of the popping of an antibubble ‘glued’ below the free surface (from top left to bottom right). The popping occurs in the left-hand side panel.

buoyancy, i.e. to slow down the rising motion. As in figure 5, only one ring is observed. It appears at the breaking point and moves in a direction opposite to the direction of expelled air. Note also that a small amount of the inner globule is dragged by the ejected air bubble.

3.2. Sinking antibubbles

An equation exists to describe the relation between the thickness of the air shell $\varepsilon$ and the pressure difference $\Delta p$ between the air shell and the surrounding liquid. This equation is similar to the Laplace law found for the bubbles. The Laplace law for an air bubble of radius $R + \varepsilon$ reads

$$\Delta p = \frac{2\gamma}{R + \varepsilon}.$$  \hspace{1cm} (3)

This equation gives a means of popping an antibubble. Indeed, any increase in pressure decreases the air-shell thickness since the air is compressible. This decrease can be such that the interaction between the surfactant layers becomes attractive. The air film then collapses as a critical pressure is reached [4]. As the air shell is thinner at the bottom than at the top of the antibubble, the antibubble pops from the bottom.

The second animation of figure 7 shows falling antibubbles in a plexiglas vessel of 200 cm height. This scene occurs at about 150 cm below the surface. The vertical range is about 20 cm. All the antibubbles pop up in this range irrespective of their radius and speed [4]. The pressure is such that the gas in the shell is so compressed that the air film collapses. The air shell breaks at its weakest point, namely at the bottom of the vessel since this area is thinner. As soon as the antibubble pops up, a large vortex ring appears. Its diameter is larger than the size of the antibubble. Meanwhile, the air pocket comes back to the surface and a second vortex ring appears. The latter is smaller than the first. The obtained configuration with two rings is known as leap-frogging [11].

When antibubbles are located 1.5 m below the surface, the pressure increased to more than 10%. This is enough to provoke film rupture at the bottom of the antibubbles since the film is thinnest there. The interaction between two surfactant layers is still a delicate problem since surfactant tails are in opposition and the medium between the layers is air (contrary to bubbles).

A single popping has been isolated in figure 6 where the pictures are separated by 0.1 s. The first ring is due to the sudden contact between a moving spherical liquid globule with
the liquid at rest. These shapes recall the Rychtmeyer–Meshkov instability found between miscible fluids because of the relative velocity [11, 12]. It takes more than 0.2 s for the second ring to be formed. It is probable that the origin of this vortex is the ejected air bubble. Popping antibubbles are represented in figure 7. The vertices are more clearly seen there.

4. Conclusions

Both formation and popping of antibubbles are linked to fluid instabilities. Rayleigh–Plateau instabilities are responsible for antibubble formation. The droplets of the incoming liquid are covered by an air film to form antibubbles. When a resting antibubble pops up, a vortex appears due to the expelled air pocket formed by the collapse of the air shell. The same phenomenon is observed when a moving antibubble pops up. Moreover, because of the relative speed of the liquid contained in the globule and the surrounding liquid, a quicker phenomenon occurs and Rychtmeyer–Meshkov instabilities are observed. Two vortices appear and turn in the same direction.

Because of the speed of the popping process and the air-shell thickness, these experiments open a new means for creating the interface between miscible fluids.

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