Investigation of a mutual interaction force at different pressure amplitudes in sulfuric acid*

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This paper investigates the secondary Bjerknes force for two oscillating bubbles in various pressure amplitudes in a concentration of 95% sulfuric acid. The equilibrium radii of the bubbles are assumed to be smaller than 10 μm at a frequency of 37 kHz in various strong driving acoustical fields around 2.0 bars (1 bar=10^5 Pa). The secondary Bjerknes force is investigated in uncoupled and coupled states between the bubbles, with regard to the quasi-adiabatic model for the bubble interior. It finds that the value of the secondary Bjerknes force depends on the driven pressure of sulfuric acid and its amount would be increased by liquid pressure amplitude enhancement. The results show that the repulsion area of the interaction force would be increased by increasing the driven pressure because of nonlinear oscillation of bubbles.

Keywords: multi-bubble sonoluminescence, secondary Bjerknes force, coupled and uncoupled state, nonlinear oscillation

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1. Introduction

Sonoluminescence (SL) is the phenomenon of light emission from a collapsing bubble, which is trapped in a liquid by a high acoustic field.[1] When SL is emitted from a cavitation bubble, it is called single bubble SL (SBSL),[2] and when the light is emitted from many cavitation bubbles, it is called multibubble SL (MBSL).[3−5] Different characteristics in the SBSL model have been studied, such as intensity and spectrum of the emitted light,[6−9] dissolved gas in liquids,[10] compression viscosity of the liquids,[11,12] and thermal effects in the forced radial oscillations of gas bubbles in liquid.[13] Also the effect of thermal conductivities of various noble gases on the SL temperature has recently been investigated by Moshaii *et al.*[14], and in 2009 they investigated the radiation produced from a stable SBSL in different concentrations of sulfuric acid.[15] The light intensity radiated from the collapsing bubble in sulfuric acid is 2700 times more than the light emitted from the cavitation bubble in water at room temperature.[16]

In the MBSL model, many parameters affect the observed SL intensity, e.g. the secondary Bjerknes force between two bubbles is known as the interaction force,[17−19] temperature in MBSL,[20] acoustic power in various liquids,[21] and ultrasonic frequency.[22] Ida *et al.* investigated the cavitation bubbles in liquid mercury.[23] Also, Eddingsaas *et al.* studied the spectrum of Ar emission from the MBSL in 95% sulfuric acid in 2007.[24] Although in these papers very useful information has been demonstrated about the MBSL, the influence of the pressure amplitude on the secondary Bjerknes force between the bubbles in 95% sulfuric acid has never been investigated, except for the initial work carried out in water.[17]

Generally, there is a time delay $t = d/c$ between an oscillation of the first bubble and the effect of the pressure on the second bubble at a distance d. For $d = 0.2$ mm, as a coupled distance of bubbles, the delay time $t = 13/8$ ms is less than 1% of the driving period ($f = 37$ kHz, $T = 0.27$ ms), which is negligible, we have found that there is not obvious influence on figures and data. This is a similar result as Mettin *et al.* mentioned before that for a typical distance $d = 1$ mm, the delay time is only about 1% of the driving period $f = 20$ kHz, which is ignorable.[17] In the uncoupled distance $d = 1.0$ cm, although the de-

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lay is much higher, however, we have chosen this large analytical amount to show the big gap between two assumed cases. To the best of our knowledge, the delay effect is not considered in multi-bubble SL in earlier works. This needs a completely different approach, which can be done in separate work.

In the present work, for the first time, the mutual interaction force between two bubbles for large (uncoupled) and small (coupled) distances in various pressure amplitudes of ultrasound field is studied, by considering the nonlinear radial oscillations for cavitation bubbles. We chose 95% wt sulfuric acid as a host liquid, based on the experimental report of Eddingsaas *et al.* on the MBSL in 95% sulfuric acid, and observing sonoluminescing light at this concentration.[24]

2. The model

The secondary Bjerknes force was discovered by Bjerknes,[17,25−27] as a mutual interaction force,[28] which is studied in different pressure amplitudes in sulfuric acid by considering the acoustic pressure gradient as the main resource of this force. The model used in the present work is almost similar to the previous methods used on the MBSL systems[17,29] with the quasi-adiabatic approach.[2,30,31] In this model, the wavelength of the driven ultrasonic field is assumed to be large in comparison with the distance between the bubbles' centres and the radius of the bubbles. The Rayleigh–Plesset equation is used to explain the radial dynamics of the bubbles corresponding to the secondary Bjerknes force.[32] The dynamics of the bubbles based on the Keller–Miksis equation[33−35] explains the radial motion of the spherical bubble, which has a nonlinear model. Therefore the equation for the second bubble is

$$\begin{aligned}&\left(1-\frac{R_i}{c}\right)R_j\ddot{R}_j+\left(\frac{3}{2}-\frac{R_j}{2c}\right)\dot{R}_j^2\\&=\frac{1}{\rho}\left(1-\frac{R_i}{c}\right)P_{s,j}+\frac{R_j}{\rho c\mathrm{d}t}P_{s,j}\\&\quad-\sum_{i=1,i\neq j}^{N}\frac{1}{D_{i,j}}\frac{\mathrm{d}(R_i^2,R_j)}{\mathrm{d}t},\end{aligned}\tag{1}$$

where i, j, and D_{ij} represent the first bubble, the second bubble, and the distance between the two bubbles, respectively. Since the first bubble's oscillations affect the second bubble's oscillations, for calculating the secondary Bjerknes force for the second one, the first bubble's oscillations should also be considered. The last term on the right-hand side of Eq. (1) denotes the acoustical mutual force between the bubbles, which couples the equations of the bubbles $1-N$. Also, $P_{s,j}$ is obtained by considering an adiabatic compression of an ideal gas within the bubble,

$$\begin{aligned}P_{s,j}=&\left(P_{\mathrm{stat}}+\frac{2\sigma}{R_{j0}}\right)\left(\frac{R_{j0}^3-h_j^3}{R_j^3-h_j^3}\right)^{\gamma}-\frac{2\sigma}{R_j}\\&-\frac{4\mu}{R_j}\dot{R}_j-P_{\mathrm{ex}}(t)-P_{\mathrm{stat}},\end{aligned}\tag{2}$$

where R_{j0}, P_{stat}, γ, σ, and μ are the equilibrium radius of the second bubble, static pressure, polytropic component, surface tension, and the viscosity of the liquid, respectively, and $P_{\mathrm{ex}}=P_a\sin(2\pi f(t)$ is the exerted driving pressure. It is clear that in order to calculate the effect of the pressure field of the second bubble on the first one, indices $i\longleftrightarrow j$ would be exchanged in Eqs. (1) and (2).

The $u_i(r,\,t)$ is the velocity field, which is

$$u_i=\frac{R_i^2R_j}{r^2},\tag{3}$$

where r, $R_i(t)$, and $R_j(t)$ are the distance from a point in flask to the centre of the first oscillating bubble, the first and the second bubbles' radii, respectively.

The pressure gradient applied on the bubble results from the driving pressure (primary Bjerknes force) and the effect of the neighbouring bubbles (secondary Bjerknes force). It is mentioned that the pressure gradient field is the source of the secondary Bjerknes force. If the bubbles' distance is small enough, the driving pressure of the bubbles would not be equal to the external driving pressure, due to the amplitude of the neighbouring bubbles' oscillations. As a result, the pressure field represents an additional driving pressure for the second bubble.[17,29] In this work, the bubbles are supposed to be far enough from each other, so the additional pressure would not be disturbed severely by the spherical shape of the other bubble.

By the Euler equation (the equation of liquid flow), the exerted pressure on the second bubble by the first one is calculated from

$$\rho\frac{\partial u_i}{\partial t}+\frac{\partial P_i}{\partial r}=0,\tag{4}$$

where ρ and P_i are the density of the liquid and the pressure field radiated from the first bubble. So, the complete form of the Euler equation is

$$\frac{\partial u_i}{\partial t}+u_i\frac{\partial u_i}{\partial r}=-\frac{1}{\rho}\frac{\partial P_i}{\partial r}.\tag{5}$$

In Eq. (5), $u_i\partial u_i/\partial r$ is of the order of $1/r^5$ and it is much smaller than the first term of the Euler equation, therefore it is negligible.

Substituting Eq. (3) into Eq. (4) for the pressure affected from the first bubble to the second one, we have

$$P_i = \frac{\rho}{r}\frac{\mathrm{d}}{\mathrm{d}t}(R_i^2\dot{R}_i). \tag{6}$$

The secondary Bjerknes force exerted on the second bubble under a pressure gradient ∇P_i with volume v_j is

$$F_{ij} = -v_j \nabla P_i, \tag{7}$$

$$F_{ij} = -v_j \frac{\partial P_i}{\partial r}\bigg|_{r=d} e_r = v_j \frac{\rho}{d^2}\frac{\mathrm{d}}{\mathrm{d}t}(R_i^2\dot{R}_i)e_r = \frac{\rho}{4\pi d^2} v_j \frac{\mathrm{d}^2 v_i}{\mathrm{d}t^2} e_r, \tag{8}$$

where v_1 and e_r are the volume of the first bubble and the radial unit vector directed from the first bubble toward the second one. The secondary Bjerknes force, F_{B} is obtained by integration of Eq. (6) over a period of the volume oscillations

$$F_{\mathrm{B}} = \langle F_{ij}\rangle = -\frac{\rho}{4\pi d^2}\langle \dot{v}_i \dot{v}_k\rangle e_r = -\frac{f_{\mathrm{b}}}{d^2} e_r, \tag{9}$$

where $\langle \dot{v}_i \dot{v}_j\rangle$ and f_{b} denote the time average of the bubbles' volume variations and secondary Bjerknes coefficient respectively. Therefore, the secondary Bjerknes force is proportional to radii oscillations $\dot{R}_i\dot{R}_j$, furthermore, with regard to Eqs. (1) and (2) the radial oscillations are different for various liquids, such as Ref. [17]. The secondary Bjerknes force will be obtained by substituting the solution of Eq. (8) into Eq. (7) after the averaging procedure. The secondary Bjerknes force of both bubbles is symmetric, where $\langle F_{ji}\rangle = -\langle F_{ij}\rangle$.

3. Numerical results

In this paper, the influence of the pressure amplitude on the secondary Bjerknes force is studied in 95% sulfuric acid. Here, the micro bubbles are smaller than 10 μm and the driven acoustical frequency is 37 kHz. Other parameters that are necessary in calculations for sulfuric acid are $\rho = -1833.7$ kg/m^3, $\mu = 26.59$ cp, $\sigma = 53.24$ mN/m, $c = 1439.69$ m/s, where, c and ρ are sound velocity in liquid and density of 95% sulfuric acid, respectively. Because in the instant of bubble collapse, the bubble wall velocity is extremely high, the transfer of mass and heat is negligible. Furthermore, during the whole cycle of the bubble's oscillation, except for the collapse time, the interior bubble revolution follows the isothermal model. Therefore, the quasi-adiabatic model is used.[2,30,31]

Because the pressure gradient is the source of the interaction force, the host liquid of this work is investigated under different driven pressures of $\sim$ 2.0 bar. Furthermore, the variation in secondary Bjerknes force has the main role in the intensity emitted by MBSL. As it was studied before, when this force increases the compact cluster of micro bubbles is formed, causing less active bubbles and reduction in the emitted intensity from MBSL. By decreasing the secondary Bjerknes force, the open cluster of bubbles is formed, which includes more active bubbles and is consequently more intense SL.[21] Another important parameter, which affects the secondary Bjerknes force, is the distance between the bubbles. Here, the distances between two bubbles in uncoupled and coupled situations are assumed to be 1 cm and 0.2 mm, respectively.

The secondary Bjerknes force can dedicate a positive or negative sign, when $F_{\mathrm{B}} < 0$ the interaction force between two bubbles is attraction, and when $F_{\mathrm{B}} > 0$, it is repulsion. In Figs. 1 and 2, the white areas represent the repulsion forces and the gray regions denote the attraction forces. Furthermore, the light and dark gray regions refer to the weak and strong attraction of secondary Bjerknes force, respectively. The darkest part of the attraction regions refer to the highest value of the secondary Bjerknes force or the strong attraction force between the bubbles.

In Fig. 1, the secondary Bjerkens forces are compared for two bubbles, in three different pressure amplitudes for the 95% sulfuric acid in the uncoupled state. R_{10} and R_{20} (the ambient radii of the first and the second bubbles) vary between 0.2 μm and 10 μm. Figure 1 shows that by decreasing the driven pressure, the interaction force would be increased, and by increasing the pressure amplitude, the repulsion region occupies a smaller area, which is shown by the white colour in the figures. For two bubbles with a big difference in their sizes, the probability of the repulsion force (resulting from nonlinear oscillations) increases at the pressure amplitude 2.2 bar than the other pressures. In general, when the driven pressure increases, the repulsion (white) region penetrates to the attraction (gray) area. On the other hand, by enhancement of the pressure amplitude, the influence of the nonlinear oscillation increases for two bubbles with a big difference in their sizes.

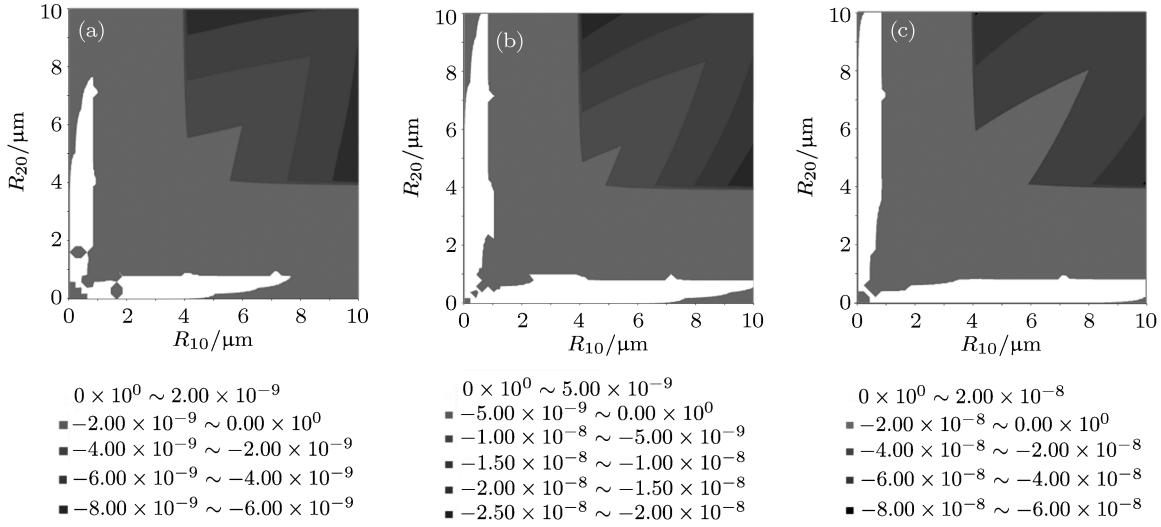


Fig. 1. The secondary Bjerknes force for the ambient radii of the first and second bubble R_{10}, R_{20} in a range of sizes smaller than 10 μm at different pressure amplitudes for the uncoupled distance (d = 1 cm) in 95% concentrations of sulfuric acid: (a) 1.8 bar, (b) 2.0 bar, (c) 2.2 bar. The secondary Bjerknes force increases as the driven pressure increases, as shown here.

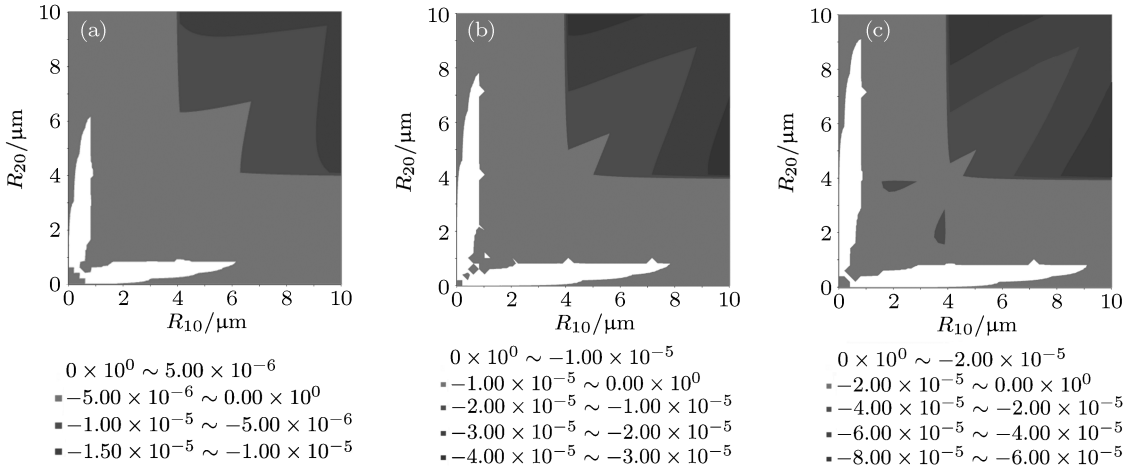


Fig. 2. The secondary Bjerknes force for the ambient radii of the first and the second bubble R_{10}, R_{20} in a range of sizes smaller than 10 μm at different pressure amplitudes for the coupled distance (d = 0.2 mm) in 95% concentrations of sulfuric acid: (a) 1.8 bar, (b) 2.0 bar, (c) 2.2 bar. The secondary Bjerknes force value increases when bubbles are in small distances, as shown here.

Figure 2 shows the secondary Bjerknes force for the bubbles at different acoustical driven pressures in sulfuric acid in the coupled state. It is found that in Fig. 2, the repulsion parts become thin and they occupy the regions where the bubbles have smaller sizes. Where the bubbles get closer, they would attract each other with much higher interaction force. Furthermore, variation of the interaction force value increases from the uncoupled state to the coupled state in the gray part, for the whole region of the attraction force. By comparing Figs. 1 and 2, we found that the secondary Bjerknes force has the highest value at a pressure amplitude of 2.2 bar in the coupled state.

In Fig 3, the oscillation radius is calculated for two bubbles, with an equal radius of 5 μm in 95% sulfuric acid at a pressure amplitude of 1.8, 2.0, 2.2 bar. Because two bubbles have the same size, they represent the same behaviours. Therefore, radius variation in a cycle under the interaction force of the first bubble in both states uncoupled (a), and coupled (b), is shown in Fig. 4. As shown in Fig. 4, the maximum radius of the bubble is proportional to the pressure amplitude. This enhancement in half cycle is clear according to the increase in the negative pressure amplitude. The

comparison between the coupled and uncoupled states indicates that the influence of the bubbles' coupled oscillations can be seen as a limitation on the behaviour of the bubbles. On the other hand, when the bubbles move closer, the oscillation ability of each bubble decreases, and consequently the maximum radius of the bubbles in the coupled state is reduced.

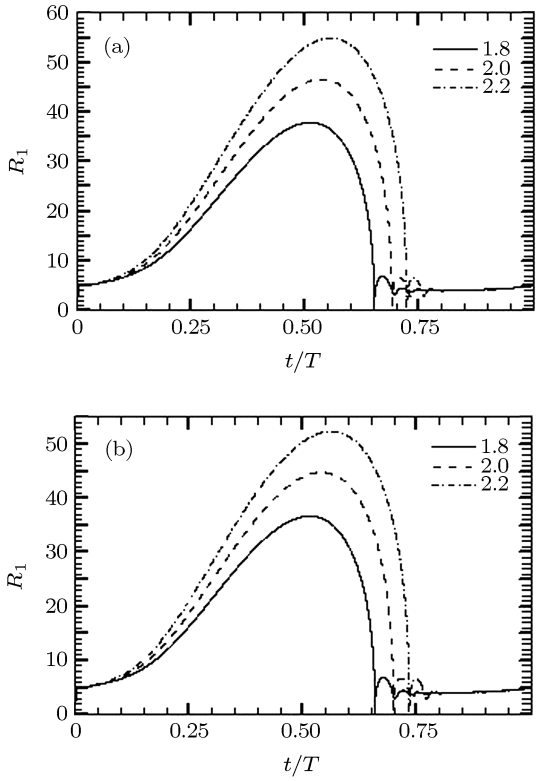


Fig. 3. Bubble radius R_1 versus normalized t/T during one driving period in different pressure amplitudes (1.8, 2.0, and 2.2 bar) for $R_{20} = R_{10} = 5$ μm for (a) uncoupled distance = 1 cm and (b) coupled distance = 0.2 mm.

Figure 4 shows the wall velocity variation of the first bubble during one cycle for different liquid driven pressures in both states: (a) uncoupled and (b) coupled states. At the collapse moment, the bubble wall velocity is increased by pressure enhancement and the maximum of the bubble wall velocity decreases in the coupled state. Because the secondary Bjerknes force is proportional to the bubble volume, the enhancement in its value is expected by decreasing the driven pressure. The variation in the secondary Bjerknes force is shown in Fig. 5. This figure represents two peaks during one acoustical cycle of radial oscillation. The small peak illustrates the fast expansion of the bubble in 0.25 to 0.5 of a cycle and the big peak refers to the instant bubble collapse. The latter relates to the secondary Bjerknes force, which is the maximum value of the interaction force.

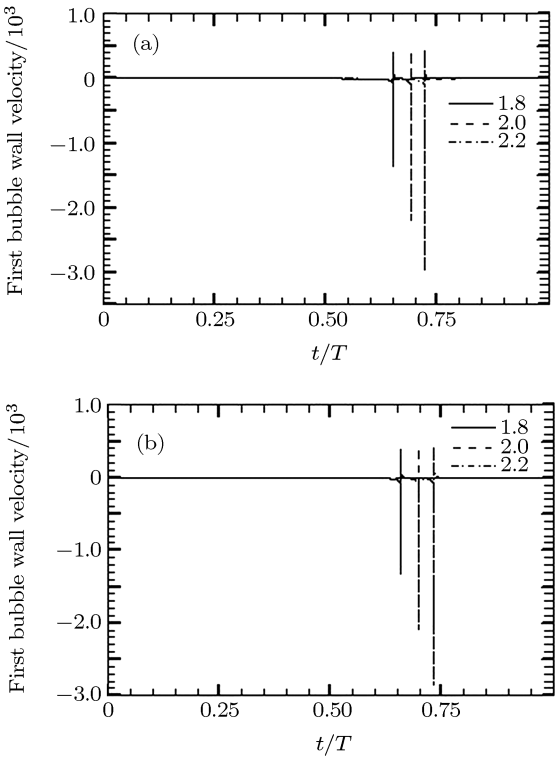


Fig. 4. Bubble wall velocity versus normalized t/T during one driving period in different pressure amplitudes (1.8, 2.0, and 2.2 bar) for $R_{20} = R_{10} = 5$ μm for (a) uncoupled distance = 1 cm and (b) coupled distance = 0.2 mm.

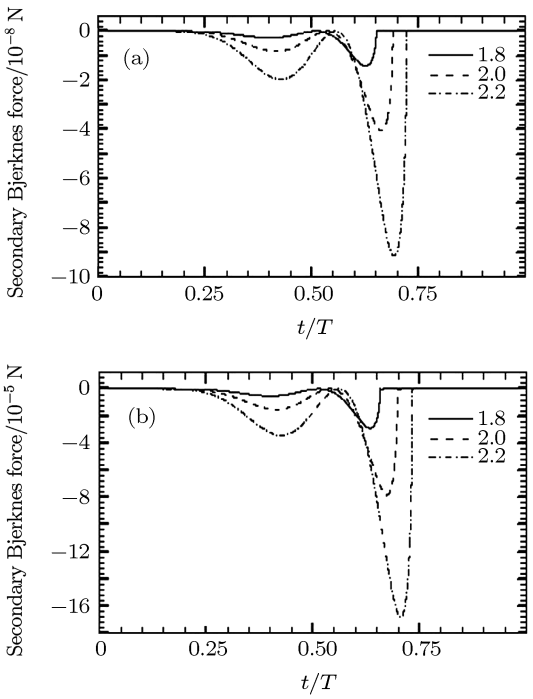


Fig. 5. Secondary Bjerknes force of the first bubble versus normalized t/T during one driving period in different pressure amplitudes (1.8, 2.0, and 2.2 bar) for $R_{20} = R_{10} = 5$ μm for (a) uncoupled distance = 1 cm and (b) coupled distance = 0.2 mm.

4. Conclusion

The variation in the secondary Bjerknes force for a wide range of bubble sizes (0.2 μm to 10 μm) was calculated for 95% sulfuric acid in different driving acoustical fields at pressures of $\sim$ 2 bar. The calculations showed that the secondary Bjerknes force was enhanced by pressure enhancement, and the repulsion interaction force between the bubbles was increased by increasing the driven pressure. The repulsion interaction force included the small domain of the bubble sizes, which was a result of the nonlinear bubble oscillations.

Another important factor was the influence of the coupled state, which was considered in the behaviour of a 5-μm bubble, e.g., the first bubble wall velocity and bubble radius oscillation in Figs. 4 and 5. It is found that the radial oscillation and the wall velocity of bubbles decrease with closer bubble distance. This causes the secondary Bjerknes force decrement in the coupled state.

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