

Effects of the second harmonic on the secondary Bjerknes force

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The time-averaged interaction force exerted by an acoustic field between two gas bubbles, known as the secondary Bjerknes force, is derived with an accuracy up to a component induced by the second harmonic of bubble oscillations. The surrounding medium is assumed to be an incompressible viscous liquid and the distance between the bubbles much larger than their radii. It is shown that the second-harmonic component of the interaction force in many cases prevents the bubbles from coalescing, causing them either to repel each other or to form a bound pair with some stable separation. This can occur providing the imposed field is strong enough so that the second-harmonic force component is comparable to the “linear” (produced by the linear oscillations of the bubbles) interaction force. The obtained results are of interest in understanding collective bubble phenomena in strong acoustic fields, such as cavitation streamer formation. [S1063-651X(99)11003-1]

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I. INTRODUCTION

The secondary Bjerknes force is a well-known effect in nonlinear acoustics. It is a time-averaged mutual interaction force of two gas bubbles in an acoustically driven liquid. This force constitutes an important component of many acoustic phenomena and applications such as acoustic cavitation, ultrasonic degassing, multibubble sonoluminescence, and medical ultrasonics [1–5]. It is named after C.A. Bjerknes and his son V.F.K. Bjerknes [6], who were the first to investigate experimentally and theoretically this effect. They derived the following expression for the force:

$$F_B = \frac{2\pi|A|^2\omega^2 R_{10}R_{20}}{\rho L^2(\omega_1^2 - \omega^2)(\omega_2^2 - \omega^2)}, \quad (1)$$

where A is the complex pressure amplitude of the incident acoustic field, ω is the angular driving frequency, R_{10} and R_{20} are the equilibrium radii of the bubbles, ρ is the equilibrium density of the liquid, L is the distance between the equilibrium centers of the bubbles, ω_1 and ω_2 are the monopole resonance frequencies of the bubbles, $F_B > 0$ corresponds to the mutual attraction of the bubbles, and $F_B < 0$ to the mutual repulsion. It is seen from Eq. (1) that the repulsion occurs when ω lies between ω_1 and ω_2 . Otherwise the bubbles are attracted to each other. The Bjerknes theory is based on the following assumptions: (i) The surrounding medium is an ideal incompressible fluid; (ii) the gas within the bubbles obeys the adiabatic law; (iii) $R_{10}, R_{20} \ll L$ so that the shape deviations of the bubbles from sphericity and the scattered waves of higher order than the primary ones can be neglected; (iv) $|A|$ is small enough so that the bubbles oscillate linearly with the driving frequency alone. When these conditions are met, the Bjerknes theory is in agreement with experiments [7,8]. If, however, this is not the case, then certain effects are observed that cannot be explained by using Eq. (1). One such effect is the formation of stable bubble clusters that were first reported by Kobelev *et al.* [9] and observed more recently by Marston *et al.* [10]. The clusters consisted of several bubbles noticeably larger than resonance

size. They neither coagulated nor broke down into individual bubbles as long as the sound field was on. A very important point is that the sound field used in [9] was rather weak: The ratio of the driving pressure amplitude to the hydrostatic pressure did not exceed 0.035. (In [10], stronger fields were applied.) This fact suggests that the observed clusters are not associated with nonlinear bubble oscillations. The key to this problem was given by Zabolotskaya [11]. She has shown that Eq. (1) fails because of ignoring radiation coupling of the two bubbles, i.e., the influence of the bubbles' scattered fields on each other's pulsations. Allowing for this coupling yields a refined formula for the interaction force that provides an insight into the nature of the bubble clusters. According to that formula, two bubbles, when approaching each other, behave as if their resonance frequencies ω_1 and ω_2 were increased. Therefore, if both bubbles are driven above resonance and one (or both) of their resonance frequencies is close enough to ω , the interaction force may change from attraction to repulsion as the bubbles are coming closer to each other. This result was confirmed (and extended) later by more accurate calculations allowing for multiple scattering of sound between the bubbles and their shape oscillations [12]. An extensive numerical investigation of the relative motion of two bubbles in stronger sound fields (with relative driving pressure amplitudes of 0.2–0.3) has been conducted in [13]. It revealed that the mechanism proposed by Zabolotskaya works in such fields, too, which is confirmed by the experiments reported in [10].

When the wavelength of sound is comparable to separation distances between bubbles, the compressibility of the surrounding liquid is no longer negligible. Its effect on the secondary Bjerknes force has been examined in [14], keeping up all the other limitations of the Bjerknes theory. It has been found that the compressibility of the liquid gives rise to long-range terms in the interaction force that are inversely proportional to L instead of L^2 . Those terms can make the two bubbles form a stable bound pair with a spacing of the order of the wavelength of sound. This prediction has not been borne out by experiment yet.

Bubbles are able to form stable structures not only in the cases considered above. It is well known from experiments

that in strong acoustic fields they group themselves into branched filamentary structures named ‘‘acoustic streamers’’ [15]. These are formed mainly from bubbles smaller than resonance size, although according to the Bjerknæs and Zabolotskaya theories bubbles of this sort should always coalesce. Oguz and Prosperetti [16] have investigated numerically the interaction of two bubbles, maintaining all the restrictions of the Bjerknæs theory but assuming that the bubbles oscillate slightly nonlinearly. They have found that nonlinear effects can change the sign of the interaction force with respect to the predictions of Eq. (1). In particular, repulsion may also appear even if both of the bubbles are driven below their fundamental resonance frequencies. It was observed that the repulsive force emerged if 2ω lay between ω_1 and ω_2 and the forcing was high enough, on the order of 0.5 bar at a static pressure of 1 bar. For lower driving pressures and other relations between the frequencies, the effect disappeared. Oguz and Prosperetti conjectured that it was caused by a strong component at twice the driving frequency developing in bubble pulsations due to the strong forcing. Mettin *et al.* [17] have examined even stronger sound fields, with driving pressure amplitudes exceeding 1 bar. They allowed for radiation coupling of two bubbles but assumed that the bubbles were far enough apart and therefore remained spherical at all times. The resonance frequencies of both bubbles were chosen to be well above the driving frequency. It was found that for some bubble pairs a mutual approach changed the interaction force from attraction to repulsion, resulting in a stable separation distance. Obviously, as ω_1 and ω_2 were far above 2ω , a physical mechanism of this process is other than that proposed in [16]. It has been shown that the changeover of the force in this case is likely to be associated with a nonlinear resonancelike response of bubbles, occurring in very strong sound fields if the equilibrium bubble radius is larger (but not much) than a certain value (dynamical Blake threshold [18]) which is typically equal to a few microns.

From the above overview it can be seen that the physical mechanisms responsible for the bubble clusters in weak acoustic fields are clear enough while the nature of the bubble structures occurring in medium- and high-intensity fields strongly calls for further investigation. This paper seeks to gain some insight into how the radiation interbubble forces are affected by nonlinear oscillations. To this end, in Sec. I an expression is derived for the Bjerknæs force between two bubbles, including the component produced by the second harmonic of the bubble oscillations. The obtained equations are then discussed in Sec. II.

II. THEORY

Let two gas bubbles be in a liquid driven by a sound wave field. Suppose that the wavelength of sound is much larger than the distance L between the bubbles and that L is large compared with the equilibrium radii R_{10} and R_{20} of the bubbles. Then both the compressibility of the liquid and the shape deviations of the bubbles from sphericity can be neglected. The viscosity of the liquid is taken into account but acoustic streaming around the bubbles is assumed to be negligible. Under these conditions, equations of the radial pulsations of two interacting bubbles are given by [17]

$$R_1 \ddot{R}_1 + \frac{3}{2} \dot{R}_1^2 - \frac{p_{10}}{\rho} \left(\frac{R_{10}}{R_1} \right)^{3\gamma} + \frac{2\sigma}{\rho R_1} + \frac{4\nu \dot{R}_1}{R_1} + \frac{1}{L} \frac{d}{dt} (\dot{R}_2 R_2^2) = \frac{p_{v1} - p_0 - p_I}{\rho}, \quad (2)$$

$$R_2 \ddot{R}_2 + \frac{3}{2} \dot{R}_2^2 - \frac{p_{20}}{\rho} \left(\frac{R_{20}}{R_2} \right)^{3\gamma} + \frac{2\sigma}{\rho R_2} + \frac{4\nu \dot{R}_2}{R_2} + \frac{1}{L} \frac{d}{dt} (\dot{R}_1 R_1^2) = \frac{p_{v2} - p_0 - p_I}{\rho}, \quad (3)$$

where $R_j(t)$ is the instantaneous radius of the j th bubble ($j = 1, 2$), the dot denotes the time derivative, p_{j0} is the equilibrium gas pressure inside the j th bubble, γ is the polytropic exponent of the gas, σ is the surface tension, ν is the kinematic viscosity of the liquid, p_{vj} is the vapor pressure inside the j th bubble, p_0 is the static pressure in the liquid, and p_I is the driving acoustic pressure.

To calculate the interaction force up to the second-harmonic component, Eqs. (2) and (3) should be solved with accuracy up to the second harmonic. For this purpose, $R_j(t)$ is represented as

$$R_j(t) = R_{j0} + x_j^{(1)}(\omega t) + x_j^{(2)}(2\omega t), \quad (4)$$

where $x_j^{(1)}(\omega t)$ is the linear change of the j th bubble’s radius, proportional to the driving pressure amplitude A , and $x_j^{(2)}(2\omega t)$ is the second-order change proportional to $|A|^2$ and involving a time-varying (with twice the driving frequency) term and a time-constant term:

$$x_j^{(2)}(2\omega t) = \tilde{x}_j^{(2)}(2\omega t) + \bar{x}_j^{(2)}. \quad (5)$$

A. Linear equations

To find $x_j^{(1)}(\omega t)$, we substitute Eq. (4) into Eqs. (2) and (3) and retain only the linear terms:

$$\ddot{x}_1^{(1)} + \omega \delta_1 \dot{x}_1^{(1)} + \omega_1^2 x_1^{(1)} + \frac{R_{20}^2}{R_{10} L} \ddot{x}_2^{(1)} = -\frac{p_I}{\rho R_{10}}, \quad (6)$$

$$\ddot{x}_2^{(1)} + \omega \delta_2 \dot{x}_2^{(1)} + \omega_2^2 x_2^{(1)} + \frac{R_{10}^2}{R_{20} L} \ddot{x}_1^{(1)} = -\frac{p_I}{\rho R_{20}}. \quad (7)$$

Here ω_j and δ_j are, respectively, the monopole resonance frequency and the dimensionless viscous damping constant of the j th bubble, given by

$$\omega_j = \frac{1}{R_{j0}} \left(\frac{3\gamma p_{j0}}{\rho} - \frac{2\sigma}{\rho R_{j0}} \right)^{1/2}, \quad (8)$$

$$\delta_j = \frac{4\nu}{\omega R_{j0}^2}. \quad (9)$$

Solutions to Eqs. (6) and (7) are more conveniently sought in the complex form. The real quantities p_I and $x_j^{(1)}(\omega t)$ can be represented as

$$p_I = \text{Im}(P_I) \quad \text{and} \quad x_j^{(1)}(\omega t) = \text{Im}(X_j^{(1)}), \quad (10)$$

where Im denotes ‘‘the imaginary part of’’ and the complex quantities P_I and $X_j^{(1)}$ are given by

$$P_I = A \exp(i\omega t), \quad (11)$$

$$X_j^{(1)} = A_j^{(1)} \exp(i\omega t). \quad (12)$$

Substitution of Eqs. (11) and (12) into Eqs. (6) and (7) yields

$$(\omega_1^2 - \omega^2 + i\omega^2 \delta_1) A_1^{(1)} - \frac{R_{20}}{R_{10}} \omega^2 \xi_2 A_2^{(1)} = -\frac{A}{\rho R_{10}}, \quad (13)$$

$$(\omega_2^2 - \omega^2 + i\omega^2 \delta_2) A_2^{(1)} - \frac{R_{10}}{R_{20}} \omega^2 \xi_1 A_1^{(1)} = -\frac{A}{\rho R_{20}}, \quad (14)$$

where $\xi_j = R_{j0}/L$. Solving these equations for the unknowns $A_1^{(1)}$ and $A_2^{(1)}$, one obtains

$$A_1^{(1)} = -\frac{A}{\rho R_{10} D_1} (\xi_2 \omega^2 + \omega_2^2 - \omega^2 + i\omega^2 \delta_2), \quad (15)$$

$$A_2^{(1)} = -\frac{A}{\rho R_{20} D_1} (\xi_1 \omega^2 + \omega_1^2 - \omega^2 + i\omega^2 \delta_1), \quad (16)$$

where

$$D_1 = (\omega_1^2 - \omega^2 + i\omega^2 \delta_1)(\omega_2^2 - \omega^2 + i\omega^2 \delta_2) - \xi_1 \xi_2 \omega^4. \quad (17)$$

B. Equations for $x_j^{(2)}(2\omega t)$

Substituting Eq. (4) into Eqs. (2) and (3) and retaining only the second-order terms, one has

$$\begin{aligned} \ddot{x}_1^{(2)} + \omega \delta_1 \dot{x}_1^{(2)} + \omega_1^2 x_1^{(2)} + \frac{R_{20}}{R_{10}} \xi_2 \ddot{x}_2^{(2)} \\ = \frac{1}{R_{10}} \left[\Omega_1^2 (x_1^{(1)})^2 + \omega \delta_1 x_1^{(1)} \dot{x}_1^{(1)} - x_1^{(1)} \ddot{x}_1^{(1)} - \frac{3}{2} (\dot{x}_1^{(1)})^2 \right. \\ \left. - 2\xi_2 \frac{d}{dt} (x_2^{(1)} \dot{x}_2^{(1)}), \right] \end{aligned} \quad (18)$$

$$\begin{aligned} \ddot{x}_2^{(2)} + \omega \delta_2 \dot{x}_2^{(2)} + \omega_2^2 x_2^{(2)} + \frac{R_{10}}{R_{20}} \xi_1 \ddot{x}_1^{(2)} \\ = \frac{1}{R_{20}} \left[\Omega_2^2 (x_2^{(1)})^2 + \omega \delta_2 x_2^{(1)} \dot{x}_2^{(1)} - x_2^{(1)} \ddot{x}_2^{(1)} - \frac{3}{2} (\dot{x}_2^{(1)})^2 \right. \\ \left. - 2\xi_1 \frac{d}{dt} (x_1^{(1)} \dot{x}_1^{(1)}), \right] \end{aligned} \quad (19)$$

where

$$\Omega_j^2 = \frac{1}{\rho R_{j0}^2} \left[\frac{3}{2} \gamma (3\gamma + 1) p_{j0} - \frac{2\sigma}{R_{j0}} \right]. \quad (20)$$

Let us again take advantage of the complex representation. Writing the real quantities $x_j^{(1)}(\omega t)$ and $\tilde{x}_j^{(2)}(2\omega t)$ as

$$x_j^{(1)}(\omega t) = \frac{i}{2} (X_j^{(1)*} - X_j^{(1)}) \quad (21)$$

[see Eqs. (10); the asterisk denotes the complex conjugate] and

$$\tilde{x}_j^{(2)}(2\omega t) = \text{Im}(X_j^{(2)}), \quad (22)$$

one finds from Eqs. (18) and (19)

$$\bar{x}_j^{(2)} = \frac{2\Omega_j^2 - \omega^2}{4R_{j0}\omega_j^2} |A_j^{(1)}|^2, \quad (23)$$

$$\begin{aligned} \ddot{X}_1^{(2)} + \omega \delta_1 \dot{X}_1^{(2)} + \omega_1^2 X_1^{(2)} + \frac{R_{20}}{R_{10}} \xi_2 \ddot{X}_2^{(2)} \\ = \frac{i}{2R_{10}} \left[X_1^{(1)} \ddot{X}_1^{(1)} + \frac{3}{2} (\dot{X}_1^{(1)})^2 - \Omega_1^2 (X_1^{(1)})^2 \right. \\ \left. - \omega \delta_1 X_1^{(1)} \dot{X}_1^{(1)} + 2\xi_2 \frac{d}{dt} (X_2^{(1)} \dot{X}_2^{(1)}) \right], \end{aligned} \quad (24)$$

$$\begin{aligned} \ddot{X}_2^{(2)} + \omega \delta_2 \dot{X}_2^{(2)} + \omega_2^2 X_2^{(2)} + \frac{R_{10}}{R_{20}} \xi_1 \ddot{X}_1^{(2)} \\ = \frac{i}{2R_{20}} \left[X_2^{(1)} \ddot{X}_2^{(1)} + \frac{3}{2} (\dot{X}_2^{(1)})^2 - \Omega_2^2 (X_2^{(1)})^2 \right. \\ \left. - \omega \delta_2 X_2^{(1)} \dot{X}_2^{(1)} + 2\xi_1 \frac{d}{dt} (X_1^{(1)} \dot{X}_1^{(1)}) \right]. \end{aligned} \quad (25)$$

Equation (23) gives the constant term of $x_j^{(2)}(2\omega t)$. It is not required for calculating the interaction force and presented here only for completeness of exposition. The other two equations are complex equations for the time-varying term of $x_j^{(2)}(2\omega t)$. Upon substitution of

$$X_j^{(2)} = A_j^{(2)} \exp(2i\omega t), \quad (26)$$

they yield

$$\begin{aligned} (\omega_1^2 - 4\omega^2 + 2i\omega^2 \delta_1) A_1^{(2)} - \frac{4R_{20}\omega^2}{R_{10}} \xi_2 A_2^{(2)} \\ = -\frac{i}{2R_{10}} \left[\left(\Omega_1^2 + \frac{5}{2} \omega^2 + i\omega^2 \delta_1 \right) (A_1^{(1)})^2 \right. \\ \left. + 4\omega^2 \xi_2 (A_2^{(1)})^2 \right], \end{aligned} \quad (27)$$

$$\begin{aligned} (\omega_2^2 - 4\omega^2 + 2i\omega^2 \delta_2) A_2^{(2)} - \frac{4R_{10}\omega^2}{R_{20}} \xi_1 A_1^{(2)} \\ = -\frac{i}{2R_{20}} \left[\left(\Omega_2^2 + \frac{5}{2} \omega^2 + i\omega^2 \delta_2 \right) (A_2^{(1)})^2 \right. \\ \left. + 4\omega^2 \xi_1 (A_1^{(1)})^2 \right]. \end{aligned} \quad (28)$$

Solving Eqs. (27) and (28) for the unknowns $A_1^{(2)}$ and $A_2^{(2)}$, one obtains

$$A_1^{(2)} = -\frac{i}{2R_{10}D_2} \left[\left(\Omega_1^2 + \frac{5}{2}\omega^2 + i\omega^2\delta_1 \right) (\omega_2^2 - 4\omega^2 + 2i\omega^2\delta_2)(A_1^{(1)})^2 + 4\omega^2\xi_2 \left(\omega_2^2 + \Omega_2^2 - \frac{3}{2}\omega^2 + 3i\omega^2\delta_2 \right) (A_2^{(1)})^2 \right], \quad (29)$$

$$A_2^{(2)} = -\frac{i}{2R_{20}D_2} \left[\left(\Omega_2^2 + \frac{5}{2}\omega^2 + i\omega^2\delta_2 \right) (\omega_1^2 - 4\omega^2 + 2i\omega^2\delta_1)(A_2^{(1)})^2 + 4\omega^2\xi_1 \times \left(\omega_1^2 + \Omega_1^2 - \frac{3}{2}\omega^2 + 3i\omega^2\delta_1 \right) (A_1^{(1)})^2 \right], \quad (30)$$

where

$$D_2 = (\omega_1^2 - 4\omega^2 + 2i\omega^2\delta_1)(\omega_2^2 - 4\omega^2 + 2i\omega^2\delta_2) - 16\omega^4\xi_1\xi_2. \quad (31)$$

So, Eqs. (4), (5), (10), (12), (15)–(17), (22), (23), (26), and (29)–(31) give us the instantaneous radii of the two interacting bubbles up to the second harmonic. We can now proceed to calculate the interaction force itself.

C. Interaction force

Since the interaction force on one bubble is equal and opposite to that on the other bubble, it is sufficient to calculate one of them, say, the force on bubble 1. This is given by [17]

$$\mathbf{F}_1 = -\langle v_1 \nabla p_2 \rangle, \quad (32)$$

where $\langle \rangle$ denotes the time average, $v_1 = 4\pi R_1^3/3$ is the instantaneous volume of bubble 1, and ∇p_2 is the pressure gradient generated by bubble 2 at the equilibrium center of bubble 1, given by

$$\nabla p_2 = \frac{\rho}{L^2} \frac{d}{dt} (R_2^2 \dot{R}_2) \mathbf{e}_{12}, \quad (33)$$

in which \mathbf{e}_{12} is the unit vector directed from the equilibrium center of bubble 1 to that of bubble 2. Setting $v_1 = \text{Im}(V_1)$ and $p_2 = \text{Im}(P_2)$, where V_1 and P_2 are the respective complex volume and pressure, Eq. (32) can be rewritten as

$$\mathbf{F}_1 = -\frac{1}{2} \text{Re}(\tilde{V}_1^* \nabla P_2), \quad (34)$$

where Re denotes ‘‘the real part of’’ and \tilde{V}_1 is the time-varying part of V_1 . With accuracy up to the second-order terms, \tilde{V}_1 and ∇P_2 are given by

$$\tilde{V}_1 = 2\pi R_{10} [2R_{10}X_1^{(1)} + 2R_{10}X_1^{(2)} - i(X_1^{(1)})^2], \quad (35)$$

$$\nabla P_2 = \frac{\rho R_{20}}{L^2} \frac{d}{dt} (R_{20}\dot{X}_2^{(1)} + R_{20}\dot{X}_2^{(2)} - iX_2^{(1)}\dot{X}_2^{(1)}) \mathbf{e}_{12}. \quad (36)$$

Substituting these equations into Eq. (34) and using Eqs. (12) and (26), one finally obtains

$$\mathbf{F}_1 = (F_1^{(1)} + F_1^{(2)}) \mathbf{e}_{12}, \quad (37)$$

where

$$F_1^{(1)} = 2\pi\rho\omega^2 R_{10}R_{20}\xi_1\xi_2 \text{Re}(A_1^{(1)*}A_2^{(1)}), \quad (38)$$

$$F_1^{(2)} = 2\pi\rho\omega^2\xi_1\xi_2 \text{Re}[4R_{10}R_{20}A_1^{(2)*}A_2^{(2)} + (A_1^{(1)*})^2(A_2^{(1)})^2 + 2iR_{10}A_1^{(2)}(A_2^{(1)*})^2 + 2iR_{20}A_2^{(2)}(A_1^{(1)*})^2]. \quad (39)$$

Equations (38) and (39) are, respectively, the ‘‘linear’’ (produced by the linear oscillations) and the second-harmonic components of the radiation interaction force acting on bubble 1.

III. DISCUSSION

Substituting Eqs. (15) and (16) for $A_1^{(1)}$ and $A_2^{(1)}$, one obtains the ‘‘linear’’ component, Eq. (38), in an explicit form:

$$F_1^{(1)} = \frac{2\pi|A|^2\omega^2 R_{10}R_{20}}{\rho L^2 |D_1|^2} \times [(\xi_1\omega^2 + \omega_1^2 - \omega^2)(\xi_2\omega^2 + \omega_2^2 - \omega^2) + \omega^4\delta_1\delta_2]. \quad (40)$$

The similar equation was first obtained by Zabolotskaya [11]. It improves the Bjerknes formula, Eq. (1), by allowing for the dissipation of energy and radiation coupling of the bubbles. It is seen from Eq. (40) that due to the radiation coupling between the bubbles the ‘‘linear’’ force can change its sign as the bubbles approach each other and the change comes about in such a manner as if their resonance frequencies were increased with reducing the separation distance. As has already been mentioned in the Introduction, this explains the way bubbles driven above resonance form stable clusters in weak acoustic fields [9,10,12].

However, our prime interest here is the second-harmonic component of the interaction force. Substitution of Eqs. (15), (16), (29), and (30) into Eq. (39) yields

$$F_1^{(2)} = \frac{2\pi|A|^4\omega^2}{\rho^3 R_{10}R_{20}L^2 |D_1|^4 |D_2|^2} \text{Re}\{T_1 T_2^* G_1 G_2^* H_1 H_2^*\}, \quad (41)$$

where

$$T_j = (\omega_j^2 - \omega^2 + \xi_j\omega^2 + i\omega^2\delta_j)^2, \quad (42)$$

$$G_j = \omega_j^2 + \Omega_j^2 - \frac{3}{2}\omega^2 - 3i\omega^2\delta_j, \quad (43)$$

$$H_j = \omega_j^2 - 4\omega^2 + 4\omega^2 B_j \xi_j + 2i\omega^2\delta_j, \quad (44)$$

$$B_j = \left(\frac{R_{3-j0}}{R_{j0}} \right)^2 \frac{T_{3-j} G_j^*}{T_j G_{3-j}^*}. \quad (45)$$

Allowing for dissipation makes the analysis of Eq. (41) too difficult. To avoid numerical computations and yet to reveal the main features of the second-harmonic force component, we will set $\delta_1, \delta_2 = 0$. As a result, Eq. (41) is simplified to

$$F_1^{(2)} = \frac{2\pi|A|^2\omega^2(\omega_1^2 - \omega^2 + \xi_1\omega^2)^2(\omega_2^2 - \omega^2 + \xi_2\omega^2)^2 M_1 M_2 N_1 N_2}{\rho^3 L^2 R_{10} R_{20} (\omega_1^2 - \omega^2)^4 (\omega_2^2 - \omega^2)^4 (\omega_1^2 - 4\omega^2)^2 (\omega_2^2 - 4\omega^2)^2}, \quad (46)$$

where

$$M_j = \omega_j^2 - 4\omega^2 + 4\omega^2 b_j \xi_j, \quad (47)$$

$$N_j = \omega_j^2 + \Omega_j^2 - \frac{3}{2}\omega^2, \quad (48)$$

$$b_1 = \frac{R_{20}^2(\omega_2^2 - \omega^2)^2 N_1}{R_{10}^2(\omega_1^2 - \omega^2)^2 N_2}, \quad b_2 = 1/b_1. \quad (49)$$

It is seen that the sign of $F_1^{(2)}$, which is of special interest to us, is determined by the factors $M_1, M_2, N_1,$ and N_2 .

Let us first consider the case where both bubbles are driven below the main resonance ($\omega < \omega_1, \omega_2$), the case of ‘‘small bubbles.’’ For such bubbles $N_1, N_2 > 0$ as $\Omega_j > \omega_j$ [cf. Eqs. (8) and (20)] and hence $\omega_j^2 + \Omega_j^2 > 1.5\omega^2$. The sign of $M_1 M_2$ depends on the relation between $\omega_1, \omega_2,$ and 2ω . If 2ω lies between ω_1 and ω_2 , then $F_1^{(2)}$ is a repulsive force which counteracts the attractive ‘‘linear’’ force. As a result, for high enough driving pressures the total force can become repulsive as well and thus prevent the bubbles from coalescing. Equation (46) also shows that the sign of $F_1^{(2)}$ depends on the distance L between the bubbles. In the case considered, it varies in such a way as if the monopole resonance frequencies of both bubbles increased as L decreases. It follows that, even if both resonance frequencies are initially below 2ω , the force $F_1^{(2)}$ can change from attraction to repulsion as the bubbles approach each other provided one (or

both) of their resonance frequencies is close enough to 2ω . This effect suggests one of the possible physical mechanisms that give rise to stable structures (such as acoustic streamers) formed by ‘‘small’’ bubbles in strong fields.

Consider the next example. Assume that one of the bubbles (say, bubble 1) is big so that $\omega_1^2 + \Omega_1^2 < 1.5\omega^2$, while the second bubble is small so that $\omega_2 > \omega$. For this case the ‘‘linear’’ force $F_1^{(1)}$ is repulsive. $M_1, N_1 < 0, N_2 > 0$, and the sign of M_2 is determined by the ratio between ω_2 and 2ω . If $\omega_2 < 2\omega$, then $M_2 < 0$ and hence $F_1^{(2)}$ hinders coalescence as well. For $\omega_2 > 2\omega$, $F_1^{(2)}$ is an attractive force counteracting $F_1^{(1)}$. Note also that in the case considered $b_1, b_2 < 0$. This means that for ω_2 close to 2ω , the sign of $F_1^{(2)}$ varies in such a way as if the resonance frequency of bubble 2 were lowered with decreasing L . In particular, if ω_2 is slightly below 2ω , then the sign of $F_1^{(2)}$ may change from repulsion to attraction as the bubbles are moving apart. As a result, $F_1^{(2)}$ will begin to counteract the repulsion caused by $F_1^{(1)}$. This may lead to a stable separation between the bubbles if, of course, the imposed field is strong enough.

Consider one more example. Let both bubbles be driven above the main resonance ($\omega_1, \omega_2 < \omega$) provided that $N_1 < 0$ and $N_2 > 0$. It is easy to see that in this case $F_1^{(1)}$ and $F_1^{(2)}$ again counteract each other ($F_1^{(1)} > 0, F_1^{(2)} < 0$). Hence the sign of the total force depends on their relative magnitudes.

So far we have neglected the dissipation terms, assuming them to be small. Even so, however, they can play a role if the bubbles are near resonances. To estimate it, consider the most interesting case where ω_1 and ω_2 are close to 2ω so that $\omega_j^2 - 4\omega^2 = \omega^2 \epsilon_j$ with $\epsilon_j \ll 1$. For this case, Eq. (41) gives

$$F_1^{(2)} \approx \frac{\pi|A|^4 (5 + 2\Omega_1^2/\omega^2)(5 + 2\Omega_2^2/\omega^2) [(\epsilon_1 + 4B_1\xi_1)(\epsilon_2 + 4B_2\xi_2) + 4\delta_1\delta_2]}{162\rho^3\omega^6 R_{10}R_{20}L^2(\epsilon_1^2 + 4\delta_1^2)(\epsilon_2^2 + 4\delta_2^2)}. \quad (50)$$

As is seen from Eq. (50), dissipation increases the mismatch between the natural frequencies of the bubbles needed for $F_1^{(2)}$ to be a repulsive force.

We can also use Eq. (50) in order to estimate the threshold pressure at which $F_1^{(2)}$ becomes comparable to $F_1^{(1)}$ since it is reasonable to assume that for ω_1 and ω_2 close to 2ω this threshold is minimum. By comparing Eq. (50) with Eq. (40), one finds

$$|A|_{th} = 6\rho\omega^2 R_{10}R_{20} \left[\frac{(\epsilon_1^2 + 4\delta_1^2)(\epsilon_2^2 + 4\delta_2^2)}{(5 + 2\Omega_1^2/\omega^2)(5 + 2\Omega_2^2/\omega^2) |(\epsilon_1 + 4B_1\xi_1)(\epsilon_2 + 4B_2\xi_2) + 4\delta_1\delta_2|} \right]^{1/2}. \quad (51)$$

As an example, let us consider two air bubbles in water, borrowing data from [16]: $R_{10}=0.1$ mm, $R_{20}=0.09$ mm, $L=5$ mm, $\omega=0.51\omega_1$ ($f=\omega/2\pi\approx 16.8$ kHz), and $p_0=1$ bar. For such bubbles, the main contribution to damping comes from heat and radiation losses. Therefore, it is reasonable to substitute for the viscous damping constants δ_1 and δ_2 in Eq. (51) the total damping constants allowing for all three kinds of losses. In this case, Eq. (51) gives $|A|_{\text{th}}\approx 0.31$ bar. This value is quite close to 0.5 bar obtained in [16] on the basis of a different (numerical) approach for the same parameters. However, it should not be expected that Eq. (51) will be in good agreement with experimental values, as the present theory is based upon the assumption of weak field such that $|x_j^{(2)}|\ll|x_j^{(1)}|\ll R_{j0}$. For this reason, the most that can be anticipated from Eq. (51) is apparently the order of magnitude of the threshold pressure.

The above discussion shows that in strong acoustic fields the radiation interaction between bubbles gets much more

intricate. As a consequence, bubble coalescence is by no means the most probable event, which is just confirmed by experiments [15]. Of course, the results of the present study are of qualitative rather than quantitative concern since they are based on the assumption of weak field. Nevertheless, they give some (initial) insight as to how nonlinear oscillations influence the radiation interbubble forces. From this standpoint, the present results are of immediate interest in understanding collective bubble phenomena in strong acoustic fields such as cavitation streamers, ultrasonic degassing, multibubble sonoluminescence, etc. They can also be helpful in the interpretation of more quantitatively correct numerical results.

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