Power-law dependence describing subharmonic generation from a non-spherically oscillating bubble

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Abstract: In this paper, we propose a technique to detect the surface oscillation of an attached bubble with radius ranging from 20 to 200 μm in an acoustic standing wave using a laser Doppler vibrometer (LDV) and a high-speed camera. The threshold condition, where the surface oscillation mode of the bubble was excited, was investigated for three different driving frequencies of 28, 39, and 81 kHz. Frequency spectrum analyses of bubble oscillation measured by the LDV and the images of the bubble simultaneously obtained by the high-speed camera experimentally demonstrated that the surface oscillation was excited when the power-law dependence \( R_{th}^{1/2} \) was satisfied. Here, \( R_0 \) is the initial bubble radius and \( R_{th} \) is the oscillation displacement of the bubble for the fundamental frequency of the incident ultrasound under the threshold condition where the displacement for the subharmonic component abruptly increased. Interestingly, the coefficient \( B \) was independent of the driving frequency. This result suggests that the proposed system can be used to check the validity of current models of surface instability on an oscillating bubble.

Keywords: Microbubble, Surface oscillation, Subharmonic, Nonlinearity, Threshold pressure

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1. INTRODUCTION

When microbubbles are exposed to ultrasound, they exhibit the spherically symmetric oscillation with a zeroth order of spherical harmonic function (i.e., the breathing oscillation mode). If the amplitude of the incident sound exceeds a threshold value, the bubble starts to show non-spherical oscillation (i.e., the surface oscillation mode) generated by parametric resonance with the breathing mode. In the case of an attached bubble, it is well known that the surface mode leads to the generation of interesting behaviors, such as microstreaming and a microjet, in terms of physics and engineering [1–6]. Many researchers have studied the dynamics of these fluid dynamical phenomena such as the threshold condition to excite the surface mode in an acoustic field [7–10]. In many applications, such as ultrasound cleaning and surface processing, acoustically driven bubbles are not only found in the bulk but are also attached to rigid surfaces where they grow from gaseous stabilized nuclei [11,12]. In the medical field, microbubbles as contrast agent for ultrasonic imaging often become attached to or near blood vessel walls. Thus, the dynamics and characteristics of phenomena related to bubbles attached to or near a wall have been intensively investigated [4,13–18]. In the sonochemical field, the shape instability of bubbles has been a classical topic in bubble dynamics. Its effect should be related to various phenomena such as the splitting of a bubble leading to dancing motion [7], the stability of luminescence in single-bubble sonoluminescence (SBSL) [19], and mass transport through a gas-liquid interface [20]. The analysis of shape instability, which was pioneered by Plesset [21,22], Birkhoff [23], Strube [24] and Prosperetti [25], has been applied to the SBSL.

In this paper, we propose an experimental method to detect the surface mode exhibited by an attached bubble. A simple and convenient method to evaluate bubble behavior is an acoustical method, such as the use of a hydrophone. We can roughly estimate bubble behaviors by observing the secondary sound radiated from oscillating bubbles.
However, it is difficult to acoustically detect the sound resulting from the surface mode because it decays proportionally to $r^{-n}$, where $r$ and $n$ denote the distance from the bubble and the order of the spherical harmonic function, respectively [26]. In contrast, high-speed imaging, which enables us to visualize the bubble, is an effective technique to analyze the behavior of extremely deformed bubbles. Trinh et al. investigated the nonlinear shape oscillations of drops and bubbles with an effective radius of 0.2–0.8 cm, which were levitated or trapped in a liquid [27]. They optically observed their contours, and fitted them with the usual expansion in terms of the time-dependent surface spherical harmonics to obtain the amplitude of each surface mode. Versluis et al. observed the oscillation of a micron-size single air bubble at a driving frequency of 130 kHz and discussed the shape mode in the parameter space of the sound pressure and initial bubble size [28]. Prabowo and Ohl summarized the characteristics of the onset of the surface oscillation and jetting of a bubble with a radius of 100 μm order on the basis of optical observation and image analysis. [8] In our previous study, a laser Doppler vibrometer (LDV) was incorporated into the observation system with a high-speed camera [29,30]. Connecting the LDV to an objective lens has a specific advantage of enabling the quantitatively measurement of the displacement of a local bubble surface with a very small amplitude of 10 nm order. On the other hand, the bubble shape cannot be observed using a LDV. The frequency spectrum of the observed displacement signal, however, can provide information to evaluate whether or not the surface mode appears. This is because the subharmonic generation is a possible factor indicating the generation of the surface mode due to parametric resonance with the breathing mode [9]. This implies that a surface oscillation with a very small amplitude, which is unable to be visualized by a camera system, can be detected using an LDV.

The focus of this work is to demonstrate the utility of an LDV system for investigating the criterion for the shape instability of an oscillating bubble. In previous works, the threshold condition to excite the surface mode was predicted by numerically solving the dynamical equation for the bubble distortion, and it was compared with the observed condition for the dancing motion and the condition for generating the surface mode [21–25,28]. Many previous works discussed the threshold condition in the parameter plane of the sound pressure amplitude and initial bubble size ($\Delta P-R_0$ plane). This is because the amplitude of bubble oscillation ($\Delta R$) could not be experimentally evaluated in detail. In this work, we attempted to evaluate the threshold condition in the $\Delta R-R_0$ plane using a simultaneous observation system with a camera and an LDV. The combined use of a camera and an LDV enabled the quantitative evaluation of the displacement of bubble oscillation. The evaluation in the $\Delta R-R_0$ plane provided the power-law dependence required to express the threshold condition.

2. EXPERIMENT SYSTEM

Figure 1 shows our experimental system, which is composed of an LDV (Polytec, NLV-2500), a high-speed video camera (Shimadzu, HPV-1), and an experimental bubble observation cell. The experimental cell was filled with normal saline solution to a depth of 45 mm. An electrical signal with a frequency of 28, 39 or 81 kHz was input to a Langevin transducer fixed to the bottom of the cell. An acoustic standing wave was formed in the cell during sound irradiation. An oxygen bubble was generated by electrolysis of the saline solution, and was situated on the glass. The contact angle between the bubble and the glass was determined by the equation $	heta = \cos^{-1}\left(\frac{H}{D}\right)$, where $H$ is the distance from the bubble center to the glass surface and $D$ is the bubble diameter in the horizontal direction. The contact angle was $32.9 \pm 12.5^\circ$. The thickness of the
glass (150μm) was sufficiently small compared with the wavelength of the incident sound so that the disturbance of the standing wave in the cell was negligible. The bubble was confined to a 5 mm² region. Because the initial bubble shape was not spherical owing to the wettability, the initial bubble radius was defined as half the bubble length in the horizontal direction \(R_0 = D_0/2\), where the subscript of zero means the initial value. The radius ranged from 20 to 200 μm.

An LDV can measure only the AC components of an oscillation signal using the Doppler effect. An objective lens (Mitutoyo, M Plan Apo 20×) was connected to the LDV head in order to focus a laser beam on the object being observed. The magnification, the working distance, and the numerical aperture of the object lens were 20, 20 mm, and 0.42, respectively. A helium-neon laser with a wavelength of 633 nm was used in the LDV. The focal spot size of the LDV beam and the focal depth were calculated to be approximately 1.5 and 3.8 μm, respectively. A helium-neon laser with a wavelength of the incident sound so that the disturbance of the standing wave in the cell was negligible. The bubble oscillation in the cell could be visualized with bright-field illumination. The onset time of the recording was synchronized with the LDV measurement by inputting a trigger signal from a delay generator to the camera and the LDV. The bubble oscillation was recorded for 0.4 ms. The recording rate in the camera system was 250,000 frames/s and the exposure time was 1 μs. The number of pixels of each image was 312 × 260 and the length per pixel was 4.2 μm/pixel. The lateral optical resolution of the image system could be varied in the range of 1.1–2 μm.

The measurement was carried out for various values of the incident sound pressure amplitude \(P_i\). For all incident ultrasound frequencies, the sound pressure of 0.2–1.4 kPa was measured using a commercial hydrophone (8103, Brüel & Kjær). The upper cutoff frequency of the hydrophone (−6 dB) was 180 kHz. When the sound pressure was very low, we were unable to measure its value, owing to the small output signal from the hydrophone. In such cases, we calculated the pressure by extrapolation under the assumption of a linear relationship between the intensities of the output signal from the hydrophone and the input electrical signal to the transducer.

### 3. BUBBLE BEHAVIOR VERSUS SOUND PRESSURE

The advantage of the high-speed camera system is that it allows the bubble shape to be clearly visualized, even though it only provides two-dimensional images. Previous researchers have emphasized the effectiveness of this characteristic for analyzing non-spherical bubble behavior, especially the extreme deformations that lead to liquid jet formation. On the other hand, several recent reports have suggested that an LDV offers a distinct advantage not provided by the high-speed camera. In a previous work using an LDV, Argo et al. observed the oscillation of bubbles attached to nylon lines and measured the resonant frequency via frequency analysis of the bubble oscillation [31]. In addition, we have previously demonstrated that an LDV enables the detection of displacement amplitudes of as small as 10 nm order [29,30]. These measurements suggest that bubble oscillations with very small amplitudes can be measured. By using both a high-speed camera and an LDV, we assert that sub-harmonic generation from a non-spherical oscillation can be investigated in detail.
The high-speed camera imaging confirmed that the bubble behavior changed with increasing incident sound pressure. At low pressures, the bubble appeared to undergo a breathing oscillation with a small displacement amplitude, although this could not be clearly observed owing to the poor spatial resolution of the imaging system. As the sound pressure increased, the bubble exhibited a surface oscillation or was torn apart by the extreme deformation.

We performed a frequency analysis of the displacement oscillation measured using the LDV. Figure 2(a) shows typical examples of velocity-time curves at three different pressures. Stable sinusoidal oscillation with the driving frequency was confirmed at the lowest pressure of 0.33 kPa. In contrast, half the frequency of the driving sound was observed at the largest pressure of 0.67 kPa. From the high-speed imaging, we confirmed that the bubble non-spherically oscillated at this pressure. Near the threshold between the above cases, we found a temporal change in the period of the velocity–time curve. The velocity started to oscillate with one period of the driving frequency. After the amplitude rapidly increased at 2.5 ms, it was found that the sub-harmonic of the driving frequency was generated. This was clearly confirmed by Fig. 2(b), which shows the spectra of the time–velocity curves. Figure 2(c) shows the normalized amplitude of each frequency component ($\Delta R_f/R_0$, $\Delta R_{f/2}/R_0$, and $\Delta R_{2f}/R_0$) as a function of the incident sound pressure $P_i$. The displayed bubble images were observed under each condition using the high-speed camera. From Fig. 2(c), it was found that the fundamental component (solid line) increased linearly up to about 0.6 kPa. The normalized displacement amplitude $\Delta R_f/R_0$ as a function of $P_i$. $\Delta R_f$ is the displacement amplitude. The circles, squares and triangles represent the respective displacement amplitudes of the fundamental, second-harmonic, and sub-harmonic components ($f$, $2f$, and $f/2$). The vertical solid line denotes the condition $P_i = P_{th}$. Bubble images observed during the ultrasound irradiation in the cases of (i) $P_i < P_{th}$, (ii) $P_i = P_{th}$, and (iii) $P_i > P_{th}$ are displayed above the graph.

When the incident sound pressure reached a threshold value, we found that $\Delta R_{f/2}$ increased abruptly [see Fig. 2(b)]. In contrast, the linearity between $\Delta R_f$ and $P_i$ was often lost above the threshold. In this paper, the threshold is defined as the sound pressure threshold $P_{th}$ where $\Delta R_{f/2}$ exceeds 0.01$\Delta R_f$. There are two possible causes for the sub-harmonic generation. One cause is the strong nonlinearity of the breathing oscillation [32]. The other is the parametric resonance of the shape oscillation with the breathing oscillation. The former may have little relation with the observed sub-harmonic generation because
\( \Delta R_{3f} / \Delta R_f \) was typically smaller than 0.1 at the threshold. On the other hand, the high-speed imaging revealed that the surface oscillation mode appeared at \( P_1 > P_{th} \). This indicated that the sub-harmonic was generated as a result of the instability of the bubble surface (gas-liquid interface).

With increasing \( P_1 \), almost all the measurement results for \( P_1 > P_{th} \) confirmed that \( \Delta R_f / R_0 \) and \( \Delta R_{3f} / R_0 \) changed randomly. Note that in this pressure range, the measured displacement amplitude may have been incorrect because 1) the laser beam was not perpendicular to the bubble surface owing to its extreme deformation and 2) the oscillation amplitude of the bubble surface was much larger than the focal depth in our LDV measurement system [see the case of 0.67 kPa in Fig. 2(c)].

4. LINEAR RESONANT CHARACTERISTIC AT LOW SOUND PRESSURES

Unlike high-speed imaging, LDV measurement has not been a mainstream observation method for bubble oscillation. In order to demonstrate that the system is a powerful tool for investigating bubble oscillation with a small amplitude, we summarize the resonant characteristic of bubble oscillation below the pressure threshold. On the basis of the results in the previous section, we consider that the oscillation was dominated by the breathing mode in this pressure range. Therefore, we also compared the experimental results with a calculation based on the theoretical model of the breathing mode in the work of Prosperetti [33]. Assuming that the ambient pressure oscillates with a small amplitude (\( P = P_0 + P_1 \sin \omega t, \ P_1 \ll P_0 \)) and the radial oscillation of a bubble for the fundamental frequency component is also small (\( R_f = R_0 + \Delta R_f = (1 + \varepsilon_f)R_0, \ \varepsilon_f \ll 1 \)), the dynamical equation for \( \varepsilon \) is expressed as the following differential equation:

\[
\ddot{\varepsilon}_f + 2\beta \dot{\varepsilon}_f + \omega_0^2 \varepsilon_f = -\frac{P_1}{\rho R_0^2} \sin \omega t,
\]

\[
\omega_0^2 = -\frac{3}{\rho R_0^2} \left\{ \frac{3k}{\rho} \left( \frac{P_0 + \frac{2\sigma}{R_0}}{R_0} - \frac{2\sigma}{R_0^2} \right) \right\},
\]

\[
\beta = \beta_\Lambda + \beta_T + \beta_v,
\]

where

\[
\kappa = \frac{1}{3} \text{Re}(F),
\]

\[
\beta_\Lambda = \frac{\omega^2 R_0}{2c},
\]

\[
\beta_T = \frac{1}{2 \rho c R_0^2} \left( \frac{P_0 + \frac{2\sigma}{R_0}}{R_0} \right) \text{Im}(F),
\]

\[
\beta_v = \frac{\mu}{\rho R_0^2},
\]

and \( \omega \) and \( \omega_0 \) are the angular frequency of the incident sound and the specific (natural) angular frequency, respectively. \( \beta \) is the damping constant and \( \beta_\Lambda, \beta_T, \) and \( \beta_v \) are classified into the effects of compressibility, heat transport, and viscosity, respectively. \( \rho, \sigma, \) and \( \mu \) are the density, surface tension coefficient, and viscosity coefficient of water, respectively. \( \kappa \) is the polytropic index. \( F \) is a complex function expressing heat diffusion in the bubble and is given by

\[
F(\theta) = \frac{3\gamma \theta^2}{\theta(\theta + 3(\gamma - 1)A_{-} - 3(\gamma - 1)(\theta A_{+} - 2))},
\]

\[
\theta = R_0 \sqrt{\frac{2\omega}{(\lambda g_0/\rho_0 c_{pg})}},
\]

\[
A_\pm = \frac{\sinh \theta \pm \sin \theta}{\cosh \theta - \cos \theta},
\]

where \( \lambda g_0, \rho_0, c_{pg}, \) and \( \gamma \) are heat conductivity, density, constant-pressure specific heat, and specific heat ratio of the gas in the bubble under the equilibrium condition.

The displacement amplitude of the fundamental component \( \Delta R_f / R_0 \) linearly increased with increasing \( P_1 \). The amplitude ratio between the fundamental component and the second harmonic \( \Delta R_{2f} / R_f \) was typically smaller than 0.1 in many cases with an exceptional driving condition (second-harmonic generation was confirmed near \( R_0 = 50 \mu m \) even at 28 kHz, below the pressure threshold, where \( \Delta R_{2f} / R_f \) was larger than 0.1). These findings ensure the linearity of bubble oscillation with the incident ultrasound. The slope of the \( \Delta R_f / R_0 - P_1 \) characteristic shows the sensitivity of bubble oscillation to the incident ultrasound and should depend on the driving frequency and initial bubble size. Under the assumption of linear oscillation, we can estimate the value of \( \Delta R_f / R_0 \) at a given sound pressure by extrapolating or interpolating the curve of \( \Delta R_f / R_0 - P_1 \) as shown in Fig. 2(c). Figure 3 shows the \( \Delta R_f / R_0 - R_0 \) characteristic at \( P_1 = 1 \) kPa for the three different driving frequencies. The resonant peak appeared near 90 nm for the driving frequency of 28 kHz and near 75 nm for the driving frequency of 39 kHz. For the driving frequency of 81 kHz, the right side of the resonant curve was observed although the peak could not be confirmed. Compared with the theoretical calculation, it was found that the experimentally obtained resonant radius was smaller, i.e., the experimental resonant curve moved to the smaller side. Focusing on the curve for the driving frequency of 28 kHz, the \( Q \) value of the experimental resonant curve seems to be smaller than that of the theoretical curve. These mismatches imply that the theoretical calculation was carried out with no consideration of 1) the vaporization and condensation of water vapor and 2) the effect of the wall where the bubble was attached. Matsumoto theoretically discussed how the former factor influences on bubble oscillation [34]. In Matsumoto’s work, it was demonstrated that the temperature in the bubble changed isothermally owing to the effect of the vaporization and condensation of water vapor.
As a result, the natural frequency of the bubble was less than that in the case of an adiabatic change, which means that the resonant radius was also smaller. The wall effect should induce the interaction between the bubble and the “mirror (image)” bubble [26]. Van der Meer et al. asserted that this effect possibly decreases the resonant frequency on the basis of their experimental results and theoretical calculation [35]. Although further discussion on the disagreement between the experiment and theory is necessary, the tendency that the resonant radius decreases with increasing the driving frequency qualitatively coincides in both cases. These data support our suggestion that LDV measurement is useful for investigating the characteristics of bubble oscillation with a small amplitude in the nanometer range.

5. POWER-LOW DEPENDANCE OF THE THRESHOLD

The generation condition to excite the surface mode, i.e., the condition for shape instability, was investigated. As shown in Fig. 2, it was observed that the surface oscillation abruptly grew along with subharmonic generation near the threshold condition. This result suggested that the cause of this growth was the parametric interaction with the breathing mode. The threshold condition, therefore, should depend on the oscillation of the breathing mode. In the pioneering works, the formulation of the threshold was theoretically discussed in relation to the displacement amplitude of the breathing mode [7,9]. We attempt to summarize the threshold condition on the basis of our the experimental results.

In the case of $P_1 \leq P_{th}$, the oscillation amplitude of the breathing mode may have corresponded to the displacement amplitude of the fundamental component $\Delta R_f$. $\Delta R_f$ at the threshold pressure was defined as the threshold amplitude $\Delta R_{f,th}$. The experimental results clearly showed that $\Delta R_{f,th}$ decreased with increasing $R_0$. In addition, we noticed that the relationship between them could be expressed as a power-law dependence. Figure 4 shows a log–log chart of the relationship between $\Delta R_{f,th}$ and $R_0$ for the three different driving frequencies. Red, blue, and green plots represent the results for the frequencies of 28, 39, and 81 kHz, respectively. Although the variability of $\Delta R_{f,th}$ was large near $R_0 = 50 \mu m$ for the driving frequency of 28 kHz, the cause was possibly associated with the harmonic resonance of the breathing mode. Our previous study experimentally demonstrated that the resonant radius for a driving frequency of 28 kHz was 90 \mu m [30]. It was considered that half of this value corresponded to the radius for the harmonic resonance. In the case of near $R_0 \approx 45 \mu m$ ($P_1 < P_{th}$), it was found that $\Delta R_{2f}$ was not negligible compared with $\Delta R_f$. Figure 5 shows the characteristics of $\Delta R_{2f}/\Delta R_f$ and $\Delta R_{1/2}/\Delta R_f$ in the cases of (a) $R_0 = 174 \mu m$ and (b) $R_0 = 65 \mu m$. We clearly found that $\Delta R_{2f}/\Delta R_f$ reached 0.1 below the threshold pressure in the case of $R_0 = 65 \mu m$, although this trend could not be confirmed in the case of $R_0 = 174 \mu m$. This result proved that the nonlinearity of bubble oscillation was more apparent in the case of $R_0 = 65 \mu m$. Figure 6 shows log–
log charts of (a) the $\Delta R_{f,th} - R_0$ and $\Delta R_{2f,th} - R_0$ characteristics in the case of $\Delta R_{2f,th}/\Delta R_{f,th} > 0.1$ and (b) the $\Delta R_{2f,th}/\Delta R_{f,th}$ characteristic, where $\Delta R_{2f,th}$ is defined as the displacement amplitude of the second-harmonic component at the threshold pressure. In Fig. 6(a), it appears that the relationship between $\Delta R_{2f,th}$ and $R_0$ is dominated by a power-law dependence in contrast to the relationship between $\Delta R_{f,th}$ and $R_0$. From Fig. 6(b), we found that $\Delta R_{2f,th}/\Delta R_{f,th}$ increased with decreasing $R_0$. Although the harmonic resonance peak was not clearly found in the curve, we considered that the increase was due to the harmonic resonance. Our interpretation of these results is that $\Delta R_{2f,th}$ (i.e., the nonlinearity of bubble oscillation) has a strong influence on the shape instability near $R_0 = 50 \mu m$. Thus, the shapes of the plots in Fig. 4 were classified by the criterion for the nonlinearity of bubble oscillation, i.e., the value of $\Delta R_{2f,th}/\Delta R_{f,th}$. The circular and triangular plots represent the linear case ($\Delta R_{2f,th}/\Delta R_{f,th} < 0.1$) and the nonlinear case ($\Delta R_{2f,th}/\Delta R_{f,th} > 0.1$), respectively.

We obtained fitting curves of the experimental data using the expression

$$\Delta R_{th} = AR_0^B. \quad (13)$$

The subscript ‘th’ should be changed to ‘f_th’ (‘2f_th’) in the case of the fundamental (harmonic) component. For the driving frequency of 28 kHz, the characteristics in the linear and nonlinear cases were fitted separately. For the driving frequency of 39 and 81 kHz, there were no nonlinear cases.

The values of $A$ and $B$ and the correlation coefficients of the fitting curves are summarized in Table 1. The large determination coefficients demonstrate that the threshold condition can be described with this power-law dependence. In the case of the $\Delta R_{f,th} - R_0$ characteristic for $f = 28$ kHz and $\Delta R_{2f,th}/\Delta R_{f,th} > 0.1$. $\Delta R_{f,th}$ and $\Delta R_{2f,th}$ are defined as the displacement amplitudes of the fundamental and second harmonic frequency components at the threshold pressure, respectively. (b) $\Delta R_{2f,th}/\Delta R_{f,th}$ characteristic for $f = 28$ kHz.
firmed that the shape mode changed with the initial bubble size. Figure 7 shows examples of observed surface oscillations for the driving frequency of 28 kHz. In the displayed bubble images, the white area in the black area (bubble shadow) corresponds to the region where the back light penetrates through the bubble. Judging from the shape of this area and the contour of the bubble, the shape modes were different under four radial conditions shown in Fig. 7. In addition, it is well known that the threshold for shape instability should depend on the order of the shape mode. Considering these facts, therefore, Eq. (13) should express the value obtained by superimposing the threshold of each shape mode as shown in Fig. 4 of [28]. In conclusion, it is interesting that the superimposed threshold follows the power-law dependence. This might provide a clue to solve the underlying mechanism of surface oscillation.

6. SOUND PRESSURE THRESHOLD TO EXCITE THE SURFACE MODE

By transforming Eq. (1), we can obtain an expression for the sound pressure threshold. Figures 2 and 3 experimentally demonstrated that the fundamental component \((\Delta R_f - R_0)\) of the displacement oscillation in the case of a low sound pressure was dominated by a linear resonance

| Frequency of incident sound | A        | B        | Determination coefficient |
|----------------------------|----------|----------|--------------------------|
| Fundamental (\(\Delta R_f - R_0\)) | 28 kHz (Linear case: \(\Delta R_{2f, th}/\Delta R_{f, th} > 0.1\)) | 4.30e-10 | -0.83 | 0.10 |
| 28 kHz (Nonlinear case: \(\Delta R_{2f, th}/\Delta R_{f, th} < 0.1\)) | 2.12e-9 | -0.66 | 0.61 |
| 38 kHz | 1.29e-9 | -0.68 | 0.80 |
| 81 kHz | 7.09e-10 | -0.65 | 0.89 |
| Harmonic (\(\Delta R_{2f, th} - R_0\)) | 28 kHz (Nonlinear case: \(\Delta R_{2f, th}/\Delta R_{f, th} > 0.1\)) | 2.79e-19 | -2.83 | 0.70 |

Fig. 7 Examples of surface oscillations observed above the threshold pressure for the driving frequency of 28 kHz: (a) \(R_0 = 76\) μm, \(P_i = 1.3\) kPa, and \(P_0 = 1.14\) kPa, (b) \(R_0 = 100\) μm, \(P_i = 0.54\) kPa, and \(P_0 = 0.48\) kPa, (c) \(R_0 = 120\) μm, \(P_i = 0.82\) kPa, and \(P_0 = 0.72\) kPa, (d) \(R_0 = 151\) μm, \(P_i = 1.72\) kPa, and \(P_0 = 1.6\) kPa. The observed duration is equal to twice the period of the driving ultrasound.
system. In this case, therefore, we assume that the following second-order differential equation can be applied as the dynamical equation for $\Delta R_f$:

$$\ddot{\epsilon}_f + 2\beta \dot{\epsilon}_f + \omega_0^2 \epsilon_f = -\frac{p_1}{\rho R_0^2}, \quad (14)$$

where $\omega_0$ and $\beta$ are the natural angular frequency and damping coefficient, respectively, $p_1 = P_1 \sin \omega t$ is the sound pressure of the incident ultrasound, and $\epsilon_f$ is the normalized displacement amplitude of the breathing oscillation mode ($\Delta R_f/R_0$). Therefore, $\Delta R_f$ can be expressed as

$$\Delta R_f = \frac{P_1}{\rho R_0} \frac{1}{\sqrt{\left(\omega^2 - \omega_0^2\right)^2 + (2\beta \omega)^2}}. \quad (15)$$

By substituting Eq. (3) into Eq. (1), the following expression for the sound pressure threshold can be obtained:

$$P_{th} = \rho \sqrt{\left(\omega^2 - \omega_0^2\right)^2 + (2\beta \omega)^2} R_0^{1/2}. \quad (16)$$

Figure 8 shows the experimental data for the sound pressure threshold and the estimation obtained using Eq. (4). For the estimation, the values of $\omega_0$ and $\beta$ were calculated by the theory of Prosperetti [33], because these values could not be obtained from our experimental data shown in Fig. 3. The experimental threshold curves for the driving frequency of 28 and 39 kHz had the same saddle shape as the characteristic in previous works [28]. The disagreement between the experimental and calculated values resulted from the same reason as that for the linear resonance curve shown in Fig. 3. In order to predict the sound pressure threshold using Eq. (4), we need a method for quantitatively measuring the natural frequency and damping coefficient of a bubble. Such a measurement technique has been proposed in several previous studies. Argo et al. demonstrated the measurement of the resonant frequency in the case of millimeter bubbles using an LDV, although the damping coefficient was not measured [31]. Their measurement values coincided with the theoretical prediction. Van der Meer et al. presented a method for obtaining the eigenfrequency (natural frequency) and damping coefficient of ultrasound contrast agents [35]. They insonified a bubble several times successively while sweeping the driving frequency of the incident burst sound and they recorded movies of the bubble response using a high-speed camera. The eigenfrequency and damping coefficient were quantified by the frequency spectrum analysis of the bubble radial oscillation. If a measurement technique with high reliability can be established, Eq. (4) will be useful for predicting the sound pressure threshold for a parametric shape instability.

7. CONCLUSION

Using a high-speed camera and an LDV, we quantitatively investigated conditions under which an attached bubble oscillates non-spherically. By analysis of the frequency components contained in bubble oscillation, the generation of a subharmonic frequency component from the bubble was used to evaluate the appearance of the surface mode. It was found that the amplitude of the subharmonic component ($\Delta R_{f,h}$) abruptly increased when the amplitude of the fundamental component ($\Delta R_f$) reached a threshold value ($\Delta R_{f,th}$). This result strongly indicated that the surface oscillation was generated by the parametric resonance with the breathing mode. In addition, we found that $\Delta R_{f,th}$ significantly depended on $R_0$. The relation between them could be fitted by the expression $\Delta R_{f,th} = AR_0^B$. This demonstrated that the proposal system using a high-speed camera and an LDV enables the evaluation of the threshold condition at which the bubble starts to exhibit the surface mode.

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**Fig. 8** Sound pressure threshold to excite surface oscillation for driving frequencies of 28, 39, and 81 kHz. Plots and solid lines respectively show the experimental data and the values calculated using Eq. (4). $\omega_0$ and $\beta$ were calculated by the theory of Prosperetti [33].
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