Application of the subharmonic threshold to the measurement of the damping of oscillating gas bubbles*

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It is suggested that the damping affecting the forced radial oscillations of gas bubbles can be determined very accurately through the measurement of the subharmonic threshold for the oscillations. The advantage of the method is that, while the subharmonic oscillations have a large amplitude and can be easily detected, the theory can be accurately based on a perturbation approach. The theoretical basis for the subharmonic generation is also reviewed and its results illustrated for the case of an air bubble in water.

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INTRODUCTION

The main difficulty in the measurement of the damping affecting the radial oscillations of a gas bubble in a liquid stems from the smallness of the effects produced by an individual bubble. For this reason past experiments on single bubbles had to be made either on largeamplitude free oscillations, or on forced oscillations driven at or near resonance. 1-6 Other experimenters have tried to circumvent the difficulty by exploiting the cooperative effects of many bubbles, and have measured the attenuation of sound waves in bubbly liquids. 7-9 Both approaches, however, are subject to serious uncertainties in the interpretation and reduction of the data. For the first type of experiments these difficulties arise because present theories of bubble oscillations are based on linearized or on perturbative approaches which are incapable of dealing with largeamplitude motions. In the case of the second class of experiments, it is unclear how to take into proper account nonuniformities in bubble size and distribution, mutual interactions, and bubble motion. The result of this state of affairs is a considerable scatter in the data points (see, e.g., Fig. 2 of Ref. 4), an unfortunate situation in view of the importance of bubble oscillations in processes such as acoustic cavitation 10 and the flow of bubbly liquids. 11,12

In this study we propose a new experimental approach the theory of which can be based on small-amplitude forced oscillations of single bubbles, but which in practice does not require inherently more accurate measurements than the techniques used in the past. The basis for this method is that when a bubble is driven into oscillation at nearly twice its resonant frequency, the amplitude of the motion is usually quite small, so that a linearized theory is adequate. For certain values of the excitation and of the frequency, which depend critically on the damping, however, this small-amplitude motion is unstable against the growth of a subharmonic component, which produces a rather sharply defined order-of-magnitude increase in the amplitude. Since this event is the result of the instability of a small-amplitude oscillation, a perturbation approach is sufficient to predict very accurately in terms of the damping the values of the excitation and of the frequency at which it takes place. 13,14 To give an idea of the magnitude of the effect involved, we compare in Fig. 1 the

Fourier spectrum of the purely harmonic solution with that of the steady subharmonic oscillations for a case of relatively low excitation. ¹⁵ Since the frequency region below the driving frequency is usually silent, the presence or absence of a large subharmonic signal should be very clearly discernible over any background noise.

The experimental practicality of this approach has been demonstrated by Neppiras in the course of experiments undertaken to elucidate the apparent connection between subharmonic signal and cavitation. ^{16,17} It appears that some slight refinements of his technique could yield excellent data on the damping of oscillating gas bubbles in liquids.

It is the purpose of the present study to develop in some detail the theory of the subharmonic thresholds on the basis of the results derived in two previous studies. ^{13,14} The limits of validity and the accuracy of the theory are illustrated by comparison with some numerical computations, and explicit results are shown for the case of an air bubble in water. Finally, some possible experiments are briefly discussed.

I. SUBHARMONIC THRESHOLDS

We shall make use of several results derived in Refs. 13 and 14, whose notation we also adopt. The Rayleigh-

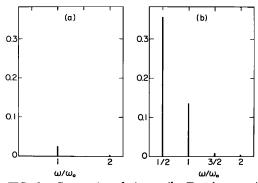


FIG. 1. Comparison between the Fourier spectrum of a bubble in purely harmonic (a) and in subharmonic oscillations (b). The parameters have the value b=0.0187, w=0.1266, $\gamma=1.33$, $\omega/\omega_0=1.955$, and $\eta=0.3$, corresponding to an air bubble of radius $R_0=10^{-3}$ cm in water if only viscous damping is considered. Both oscillatory regimes are possible depending on the initial conditions.

Plesset equation of motion for the radius R of a bubble subject to a sound field of wavelength large compared to R, which produces a pressure variation of the form $p_{\infty}(1-\eta\cos\Omega t)$ in the neighborhood of the bubble, can be written

$$R\frac{d^{2}R}{dt^{2}} + \frac{3}{2} \left(\frac{dR}{dt}\right)^{2} = \frac{1}{\rho} \left[p_{0} \left(\frac{R_{0}}{R}\right)^{3r} - p_{\infty} (1 - \eta \cos\Omega t) - \frac{2\sigma}{R} - \frac{4\mu}{R} \frac{dR}{dt} \right]. \tag{1}$$

In this equation ρ is the density of the liquid, σ the surface tension, and μ an "effective viscosity" that includes contributions from the three damping mechanisms, i.e., viscous, thermal, and acoustic. ¹⁸ The internal pressure of the bubble, which is p_0 at equilibrium, follows a polytropic law of variation, the exponent γ of which is a function among other things, of the sound frequency Ω and of the equilibrium radius R_0 . ¹⁸ The equilibrium internal pressure p_0 and the average ambient pressure p_∞ are connected by $p_0 - p_\infty = 2\sigma/R_0$, and the natural frequency of the bubble Ω_0 has the value

$$\Omega_0^2 = \frac{3\gamma p_0 - 2\sigma/R_0}{\rho R_0^2} .$$
(2)

In this equation acoustic effects have been disregarded, which is permissible provided that ωR_0 is much smaller than the speed of sound in the liquid. ¹⁸

In terms of a dimensionless time au and frequency ω defined by

$$\tau = (p_0/\rho)^{1/2}R_0^{-1}t$$
, $\omega = R_0(\rho/p_0)^{1/2}\Omega$,

to leading order the steady-state solution of (1) in the subharmonic region can be written

$$R/R_0 = 1 + \xi [(\omega^2 - \omega_0^2)^2 + 4b^2\omega^2]^{-1/2}\cos(\omega\tau + \delta) + C\cos(\frac{1}{2}\omega\tau + \varphi), \qquad (3)$$

where $\xi = (1 - w)\eta$ is the effective pressure amplitude, and the pure number w, the dimensionless damping b, and the dimensionless natural frequency of the bubble ω_0 are given by

$$w = 2\sigma/R_0 p_0$$
,
 $b = 2\mu/R_0(\rho p_0)^{1/2}$,
 $\omega_0^2 = 3\gamma - w$;

the phase angle of the purely harmonic component δ satisfies

$$\tan \delta = 2b\omega/(\omega^2 - \omega_0^2) .$$

Higher-order terms in Eq. (3) are given in Ref. 13. In the subharmonic region, where ω is close to $2\omega_0$, the amplitude C can either vanish, in which case we have purely harmonic oscillations, or have the value (correct to second order in ξ)

$$C(\omega, \xi) = \left[\frac{\omega_0^2 - \frac{1}{4}\omega^2 + g_1 \xi^2 + [\beta^2 \xi^2 - \omega^2 b^2]^{1/2}}{g_0}\right]^{1/2}, \quad (4)$$

where β , g_0 , and g_1 are given by

$$\begin{split} \beta &= \frac{1}{2} - \frac{\alpha_1 - \frac{3}{4}\omega^2}{\omega_0^2 - \omega^2} \;, \\ g_0 &= \alpha_1 \bigg(\frac{\alpha_1 - \frac{3}{8}\omega^2}{\omega_0^2} + \frac{1}{2} \; \frac{\alpha_1 + \frac{3}{8}\omega^2}{\omega_0^2 - \omega^2} \bigg) - \frac{3}{4}\alpha_2 \\ &\quad + \frac{3}{8}\omega^2 \bigg(\frac{1}{4} - \frac{\alpha_1 + \frac{3}{8}\omega^2}{\omega_0^2 - \omega^2} \bigg) \;, \\ g_1 &= \frac{\alpha_1/\omega_0^2}{\omega_0^2 - \omega^2} \bigg(1 - \frac{\alpha_1 - \frac{3}{2}\omega^2}{\omega_0^2 - \omega^2} \bigg) + \frac{3}{2} \; \frac{\alpha_2 - \frac{1}{2}\omega^2}{(\omega_0^2 - \omega^2)^2} - \frac{1}{\omega_0^2 - \omega^2} \\ &\quad + (\omega_0^2 - \frac{9}{4}\omega^2) \bigg(\frac{\alpha_1 + \frac{3}{4}\omega^2}{\omega_0^2 - \omega^2} - \frac{1}{2} \bigg) \bigg(\frac{1}{2} - \frac{\alpha_1 - \frac{9}{4}\omega^2}{\omega_0^2 - \omega^2} \bigg) \;, \end{split}$$

with

$$\alpha_1 = \frac{9}{2}\gamma(\gamma+1) - 2w$$
, $\alpha_2 = \frac{1}{2}\gamma(9\gamma^2 + 18\gamma + 11) - 3w$;

 β , g_0 , and g_1 are all positive quantities in the neighborhood of $\omega=2\omega_0$. A necessary condition for a subharmonic oscillation to be possible at a certain frequency is that the inner radicand in Eq. (4) be real. This requirement determines the mathematical threshold for the subharmonic oscillations $\xi_{\rm th}$ as

$$\xi_{\rm th}(\omega) = \omega b/\beta \ . \tag{5}$$

The condition $\xi > \xi_{\rm th}(\omega)$ is not sufficient in general, since the steady-state motion may or may not contain a subharmonic component depending on the initial conditions, as was discussed at length in Ref. 14. The subharmonic oscillation, however, is the only possible motion in that range of frequencies and amplitudes in which the purely harmonic regime is unstable. The condition for this instability is 19

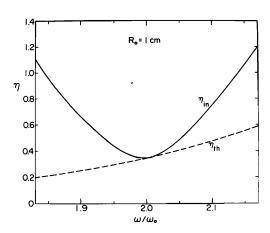
$$\left| \omega_0^2 - \frac{1}{4}\omega^2 + g_1 \xi^2 \right| < (\beta^2 \xi^2 - \omega^2 b^2)^{1/2} . \tag{6}$$

Solving for ξ one finds that, for a fixed ω , the purely harmonic solution becomes unstable only when ξ exceeds the value $\xi_{in}(\omega)$ given by

$$\xi_{in}(\omega) = \left(\frac{\beta^2 - 2g_1(\omega_0^2 - \frac{1}{4}\omega^2) - \left\{\beta^4 - 4g_1[(\omega_0^2 - \frac{1}{4}\omega^2)\beta^2 + g_1\omega^2b^2]\right\}^{1/2}}{2g_1^2}\right)^{1/2}.$$
 (7)

We shall refer to this value as to the *instability threshold*; this is the "subharmonic threshold" commonly mentioned in the literature. Equation (6) has another acceptable root, which sets an upper limit to the excitation amplitude at which the purely harmonic oscillations are unstable at a given frequency. Actually, the corresponding value of ξ is so large that it is doubtful whether the result is meaningful.

In Figs. 2-5 we present graphs of Eqs. (5) and (7) for the case of an air bubble in water. The values of the damping and of the polytropic exponent used in these figures have been taken from Ref. 18. It is seen that $\xi_{\rm in}$ has a minimum near $\omega=2\omega_0$, where it differs very little from the corresponding value of $\xi_{\rm th}$. Setting $\omega=2\omega_0$ in (5) and returning to dimensional variables we find



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FIG. 2. The instability threshold $\eta_{\rm in}$ and the true mathematical threshold $\eta_{\rm th}$ for an air bubble of radius $R_0=1$ cm in water $(p_\infty=1$ atm, $\gamma=1.39$, and $w=1.44\times 10^{-4})$. In the subharmonic region the parameter b is expressible approximately as $b=0.0533~\omega/\omega_0-0.0473$ (Ref. 18).

$$\left(\frac{p_{\text{max}}}{p_{\infty}}\right)_{\text{th}} = \frac{(3\gamma - w)^{3/2}}{(1 - w)(9\gamma^2 - w)} \frac{24\mu}{R_0(\rho p_0)^{1/2}}.$$
 (8)

This expression gives approximately the minimum value of $\xi_{in}(\omega)$.

The physical origin of the subharmonic threshold ξ_{th} can be readily explained in the following terms. 20 In a linear system, any mode other than the one sustained by the external force is eventually damped out by the dissipative mechanisms. In a nonlinear system, however, there are couplings between the different modes, so that energy can be transferred from the one corresponding to the driving frequency to sustain others at different frequencies. The ultraharmonic oscillations are driven essentially by the coupling of the external force with itself. The subharmonic modes are driven instead by the coupling of the external force to the subharmonic mode. The rate at which energy is transferred to this mode increases with its own amplitude, which in turn generally increases with the amplitude of the driving force. Therefore, if this quantity is too small, energy cannot

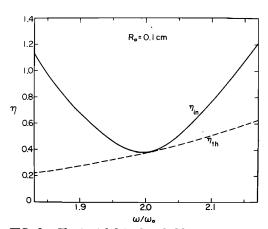


FIG. 3. The instability threshold $\eta_{\rm in}$ and the true mathematical threshold $\eta_{\rm th}$ for an air bubble of radius $R_0=0.1$ cm in water $(p_\infty=1 \text{ atm}, \ \gamma=1.38, \ w=1.44\times 10^{-3})$. In the subharmonic region the parameter b is expressible approximately as b=0.0490 $\omega/\omega_0=0.0339$ (Ref. 18).

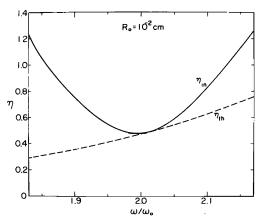


FIG. 4. The instability threshold $\eta_{\rm in}$ and the true mathematical threshold $\eta_{\rm th}$ for an air bubble of radius $R_0=10^{-2}$ cm in water $(p_\infty=1$ atm, $\gamma=1.33$, and w=0.0142). In the subharmonic region the parameter b is expressible approximately as $b=0.0370 \times \omega/\omega_0+4.48\times 10^{-3}$ (Ref. 18).

be transferred to the subharmonic mode at a sufficient rate to compensate for the dissipation, and no subharmonic oscillation can sustain itself. Quite different is the mechanism which gives rise to the instability threshold ξ_{in} . The reader is referred to a paper by Lord Rayleigh²¹ for its description.

An important question is of course that of the accuracy of the analytical expressions given above. The estimate of the error is greatly facilitated by the results of Lauterborn, 22 who has performed a very extensive numerical investigation of nonlinear bubble oscillations. In Fig. 6 we compare his numerical results 23 with $\xi_{\rm in}(\omega)$ given by Eq. (7) for the case $\gamma=1.33,~\mu=0.01$ P, $\sigma=72.5$ dyn/cm, $p_{\infty}=1$ bar, and $R_0=10^{-3}$ cm; notice that for such a relatively large value of the radius essentially $\xi=\eta$. For $\omega<2\omega_0$ the numerical threshold was determined only to be within the intervals indicated in the figure. It is seen that Eq. (7) is quite accurate for $1.9 \lesssim \omega/\omega_0 \lesssim 2.1$. For smaller values of $\omega/2\omega_0$ the agreement is less satisfactory, reflecting the fact that for the accuracy of the perturbation method

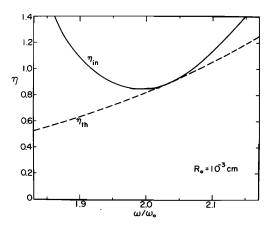


FIG. 5. The instability threshold $\eta_{\rm in}$ and the true mathematical threshold $\eta_{\rm th}$ for an air bubble of radius $R_0=10^{-3}\,{\rm cm}$ in water $(p_\infty=1\ {\rm atm},\ \gamma=1.19,\ {\rm and}\ w=0.126)$. In the subharmonic region the parameter b is expressible approximately as $b=0.0190\times\omega/\omega_0+0.0798$ (Ref. 18).

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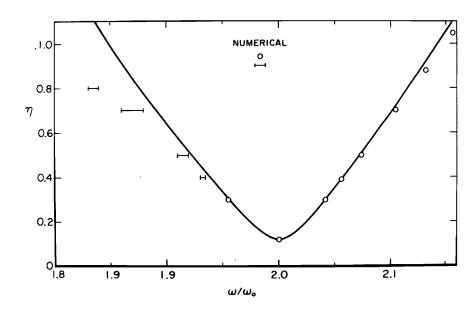


FIG. 6. Comparison between Eq. (7) and Lauterborn's numerical results, indicated by the circles and the horizontal bars (p_{∞} = 1 bar, $R_0 = 10^{-3}$ cm, $\gamma = 1.33$, $\mu = 10^{-2}$ P, and w = 0.126).

used to derive (7) not only the amplitude of the oscillation but also $|\omega-2\omega_0|/2\omega_0$ should be small. That the cause for the disagreement should be imputed to ω being too far from $2\omega_0$ rather than to the large excitation amplitude is supported by the fact that Eq. (8) coincides with Lauterborn's results up to $\xi\simeq 2.5$ (Ref. 22, Fig. 13).

It is seen therefore that the analytical results (5) and (7) are quite satisfactory over a frequency range sufficiently wide to insure their applicability to the analysis and reduction of experimental data. The reason for this great accuracy is that in the subharmonic region the amplitude of the purely harmonic oscillation is quite small, so that the perturbation method used in Ref. 13 converges very rapidly.

On comparing the difference between the numerical and the analytical results for $\omega < 2\omega_0$ and for $\omega > 2\omega_0$ it is seen that the agreement is closer in the second case. Probably this circumstance reflects the fact that for $\omega > 2\omega_0$ the subharmonic amplitude is much smaller than for $\omega < 2\omega_0$.

II. DISCUSSION

It was shown above that for a value of ξ less than $\xi_{\rm th}(\omega)$ no subharmonic oscillation is possible for a driving frequency ω . When $\xi_{th}(\omega) < \xi < \xi_{in}(\omega)$, the purely harmonic solution is still stable, but a subharmonic oscillation is also possible provided that it is suitably excited. 14 The situation is that corresponding to curve (a) in Fig. 7, in which the steady-state amplitude $C(\omega, \xi)$ does not extend down to the value C=0. Increasing ξ , the lowest point of the curve moves downwards and towards $\omega = 2\omega_0$ [curve (b) in Fig. 7], until it touches the frequency axis [curve (c), Fig. 7]. For this value of ξ , which we denote by ξ_0 , there is a frequency interval in which Eq. (6) is first satisfied. The right-hand extremum of this interval is the point where C=0, and the left-hand one ξ_{in} has been indicated by a vertical broken line which extends to intersect the curve $C(\omega, \xi_0)$. This broken line indicates that at that point the purely harmonic oscillations become unstable and that a subharmonic component of amplitude C develops. Increasing the value of ξ beyond ξ_0 , the instability interval widens [curve (d), Fig. 7]. We may now describe some possible experiments on single bubbles.

To measure the instability threshold $\xi_{\rm in}$ at a fixed frequency ω , the excitation amplitude is gradually increased. When $\xi=\xi_{\rm in}(\omega)$ a large subharmonic component develops in a few cycles. It is clear from (7) that in order for $\xi_{\rm in}$ to be very sensitive to the damping, the sound frequency must be close enough to $2\omega_0$ that the quantity $|\omega_0^2-\frac{1}{4}\omega^2|\beta^2$ is smaller than $g_1\omega^2b^2$. The subharmonic oscillations that set in when the threshold value is attained are large-amplitude ones, and may lead to a fragmentation of the bubble through an instability of its spherical shape.

Another type of experiment appears suitable for the

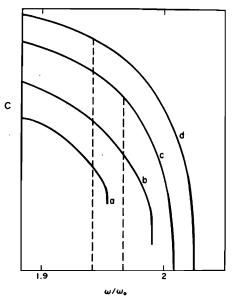


FIG. 7. Schematic illustration of the dependence of the sub-harmonic steady-state amplitude C on ω/ω_0 for different values of the driving amplitude η , with $\eta_a < \eta_b < \eta_c < \eta_{d^*}$.

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FIG. 8. Schematic illustration of the hysteresis effect of sub-harmonic bubble oscillations.

ω

measurement of the true mathematical threshold $\xi_{\rm th}(\omega)$. Here the subharmonic oscillations would have to be excited first, for instance increasing ξ above $\xi_{\rm in}(\omega)$, or temporarily adjusting the frequency closer to $2\omega_0$, or with a strong pressure pulse. Then, at fixed frequency, the driving amplitude is gradually decreased until the subharmonic signal disappears when ξ falls just below $\xi_{\rm th}$. It should be noted that the disappearance of the signal is determined by the rate of energy dissipation. Hence, if the damping is small, the process may be relatively slow, unlike that of the growth of the subharmonic which is caused by a dynamical effect and indeed impeded by the damping.

A third type of experiment of conceptual, as well as practical interest, may be described with reference to Fig. 8. Although the discussion of Sec. I is inadequate to provide a complete theoretical basis for it, still it is sufficient to give a semiquantitative framework. The sound amplitude is kept fixed at a level at which there is an interval of instability. If the frequency is gradually increased from some relatively low value below ω = $2\omega_0$, the amplitude of the subharmonic signal jumps to a large level at the value ω^* at which the applied ξ coincides with $\xi_{in}(\omega^*)$. Further increase of the frequency causes a decline of the subharmonic output as the amplitude C decreases, until at the point where the value C=0 is reached the subharmonic disappears. If now the frequency is decreased, the subharmonic amplitude again follows the curve $C(\omega, \xi)$, but this time beyond ω^* . The purely harmonic oscillation is recovered through an instability of the subharmonic oscillation, which however cannot be predicted on the basis of the perturbative approach used to derive the above results. This behavior illustrates a hysteresis effect which is typical of nonlinear oscillations. 25

Finally, a remark should be made on the possible effects of transients on the measurements proposed here. On the basis of the results of Ref. 14 it is possible to estimate that the time needed to reach the steady regime is of the order of 100 cycles. Thus, at the frequencies of common interest (1 kHz or higher) the steady state would be reached in a fraction of one second. Therefore, for all practical purposes, the dis-

cussion can be safely based on the steady oscillations, as has been done above.

In principle, measurements of the subharmonic thresholds may be useful to investigate other features of bubble oscillations besides the damping, but the accuracy needed for such investigations may render them impractical. However, since present theories predict a strong effect of the frequency [and hence, from Eq. (2), of the bubble radius if $\omega \sim 2\omega_0$] on the damping, a quantitative experimental verification of this dependence would lend strong support to the validity of the entire theoretical approach.

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termine the minimum value of $\xi_{in}(\omega)$.

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