Review of scattering and extinction cross-sections, damping factors, and resonance frequencies of a spherical gas bubble

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(Received 4 January 2011; revised 27 July 2011; accepted 1 August 2011)

Perhaps the most familiar concepts when discussing acoustic scattering by bubbles are the resonance frequency for bubble pulsation, the bubbles' damping, and their scattering and extinction cross-sections, all of which are used routinely in oceanography, sonochemistry, and biomedicine. The apparent simplicity of these concepts is illusory: there exist multiple, sometimes contradictory definitions for their components. This paper reviews expressions and definitions in the literature for acoustical cross-sections, resonance frequencies, and damping factors of a spherically pulsating gas bubble in an infinite liquid medium, deriving two expressions for "resonance frequency" that are compared and reconciled with two others from the reviewed literature. In order to prevent errors, care is needed by researchers when combining results from different publications that might have used internally correct but mutually inconsistent definitions. Expressions are presented for acoustical cross-sections associated with forced pulsations damped by liquid shear and (oft-neglected) bulk or dilatational viscosities, gas thermal diffusivity, and acoustic re-radiation. The concept of a dimensionless "damping coefficient" is unsuitable for radiation damping because different cross-sections would require different functional forms for this parameter. Instead, terms based on the ratio of bubble radius to acoustic wavelength are included explicitly in the cross-sections where needed. © *2011 Acoustical Society of America*. [DOI: 10.1121/1.3628321]

PACS number(s): 43.20.Fn, 43.20.Ks, 43.25.Yw, 43.30.Jx [CCC] Pages: 3184–3208

I. INTRODUCTION

The concepts of scattering (σ_s), extinction (σ_e) and absorption (σ_a) cross-sections are widely used in the context of a lightly damped spherical gas bubble pulsating linearly in a liquid about an equilibrium radius R_0, and in response to a sound pressure field of circular frequency ω.[1–3] The scattering and absorption cross-sections are defined, for a plane wave incident on a single bubble, as the ratios of the scattered and absorbed powers, respectively, to the incident intensity. The extinction coefficient is the sum of the two and is proportional to the rate of work done by the incident wave on the bubble. The following equations are often quoted for the scattering and extinction cross-sections of a bubble whose resonance frequency is $\omega_{\rm res}$:

$$\sigma_{\rm s} = \frac{4\pi R_0^2}{\left(\omega_{\rm res}^2/\omega^2 - 1\right)^2 + \delta^2} \tag{1}$$

$$\sigma_{\rm e} = \frac{4\pi R_0^2}{\left(\omega_{\rm res}^2/\omega^2 - 1\right)^2 + \delta^2}\frac{\delta}{\omega R_0/c}, \tag{2}$$

where c is the speed of sound in the liquid, and the total dimensionless damping coefficient (δ) is characterized as the sum of contributions from acoustic re-radiation ($\delta_{\rm rad}$), liquid viscosity ($\delta_{\rm vis}$), and thermal dissipation in the gas ($\delta_{\rm th}$). This might be generalized to cover a more general environment at the bubble wall by including an additional term ($\delta_{\rm other}$), applicable to marine sediments, contrast agents, foodstuffs, etc.:

$$\delta = \delta_{\rm rad} + \delta_{\rm vis} + \delta_{\rm th} + \delta_{\rm other}, \tag{3}$$

where for the present purpose, $\delta_{\rm other} = 0$ is assumed, in keeping with many studies of gas bubbles in simple liquids. Similarly, in addition to these other sources of damping, complications associated with stiffness,[4] elastic properties of bubble walls,[5,6] inertia,[7,8] multibubble effects,[9] departures from bubble sphericity,[10] and the proximity of boundaries,[10,11] which result in departures from the simple model of a single spherical gas bubble in an infinite body of water, are neglected except where stated otherwise. Examples of such complications include bubbles in tubes,[8,12–14] pores,[15,16] tissue,[17] gels,[18] fish flesh,[19–21] sediment,[22,23] and skins of methane hydrate[24,25] or polymer or lipid domestic and clinical products.[26–28] The absorption cross-section follows from the definitions as

$$\sigma_{\rm a} = \sigma_{\rm e} - \sigma_{\rm s}. \tag{4}$$

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The apparent simplicity and familiarity of this scheme, introduced by the benchmark publications of Medwin in the 1970s[29,30] (and popularized, for example, by Ref. 31), belies its potential for misinterpretation. The first indication of a problem comes from the realization that there are two choices for the dimensionless radiation damping component,[32] and the further realization that the answer to the question "which one is correct?" depends on how other terms (such as the bubble pulsation resonance frequency, ω_{res}) are defined.[33] Specifically, substitution of (3) in (1) and (2) for applications requiring both creates inconsistency at best and large errors at worst. Such applications include the widely used standard technique of characterizing the bubbles in a material by comparing the measured scattering and attenuation from a sample and relating them to the theoretical cross-sections σ_s and σ_e, typically by assuming that the ratio σ_s/σ_e equals the ratio δ_{rad}/δ. This erroneous approach relies on expressing (2) [using (3)] in the form

$$\sigma_e = \frac{4\pi R_0^2}{(\omega_{res}^2/\omega^2 - 1)^2 + (\delta_{rad} + \delta_{vis} + \delta_{th} + \delta_{other})^2} \times \frac{(\delta_{rad} + \delta_{vis} + \delta_{th} + \delta_{other})}{\delta_{rad}}, \quad (5)$$

which, unless (5) is used to *define* ω_{res}, is untenable since otherwise there is no single expression for δ_{rad} that leads to the correct expression for σ_e when used in this equation.[33] This misconception has been widespread for three decades, with (5) adopted following the pioneering (and fruitful) comparison of scattering and extinction cross-sections through comparison of (5) with (1).[34,35] In many environments, bubbles do not conform to the idealized assumptions of free non-interacting spherical bubbles with clean walls far from boundaries that are inherent in many formulations. These include bubbles rising from the seafloor,[36,37] bubbles in marine sediments,[22,38] and bubbles in food,[39] industrial or petrochemical products.[40] If discrepancies between the measured bubble properties (scatter, attenuation, damping, etc.) and the predictions of idealized theories (for σ_s, σ_e, ω_{res}, and δ, etc.), are attributed to asphericity, boundaries, or dopants on the bubble wall, etc., and indeed the values of adjustable parameters (e.g., for δ_{other}) in a model are estimated by fitting its predictions to measured data (as has been done with contrast agents[26,41]), it is important that the baseline ideal-bubble predictions are not based on misconceptions such as are found in (5).

It is shown in Sec. IV A that use in (3) of the expression $\delta_{rad} = (\omega R_0/c)(\omega_{res}^2/\omega^2)$, which is the correct form for use with Weston's scattering model,[19,42] and which therefore gives the correct result for σ_s if substituted in (1), nevertheless introduces an erroneous factor in σ_e that increases as $(\omega/\omega_{res})^2$ at frequencies above the bubble resonance if substituted in (2). The maximum size of this error (see Sec. II E 1) is quickly established by comparing the highest ensonification frequency with the resonance of the largest bubble that could be present in the sample. Such pitfalls are to be found in even the most familiar concepts used when discussing the scattering of sound by a bubble (the resonance frequency for bubble pulsation, the damping of the bubble, and the scattering and extinction acoustic cross-sections). The parameter δ is referred to by some authors as the "damping constant." The term "damping coefficient" is adopted here, reserving "damping constant" (or "damping factor" if a function of frequency),[43] denoted β, to mean half of the coefficient of the dR/dt term in the equation of motion describing the time evolution of the bubble radius $R(t)$, as for example in Eq. (62) of Sec. II C.

Linear models of bubble damping and cross-sections are key to measuring oceanic bubble size distributions and predicting their acoustic effects, for example, by the use of acoustical resonators,[44] backscatter,[45] and forward propagation,[46–48] with applications involving sonar performance,[49–51] wake acoustics,[52,53] and surf zone acoustics.[54,55] Furthermore, these concepts are not limited to ocean sound. They are routinely cited in dozens of bubble acoustics papers each year, covering fields as diverse as biomedical ultrasonics,[4,56,57] sonochemistry,[58] metamaterials,[59] and the use of ultrasound for industries involving liquid ceramics,[60] metals,[61] and foodstuffs.[39]

The purposes of this paper are: to review theoretical scattering models, with particular attention to the definitions of the damping coefficient and resonance frequency and the implications of these definitions; to outline the levels of understanding and precision required when defining and applying these terms as foundations for theoretical development or to interpret experimental measurements; and to indicate the sort of errors that can occur if insufficient care is taken. Section II summarizes the various expressions for cross-section, damping factor, and resonance frequency encountered in the literature. Several different and apparently contradictory expressions for the resonance frequency are encountered. The discrepancies in these are resolved in Sec. III by showing that different authors adopt the term "resonance frequency" to mean one of several conceptually different quantities, with potential for confusion created by them all being given the same name. Conclusions are listed in Sec. IV in the form of expressions for the various cross-sections, damping factors, and resonance frequencies.

II. SURVEY OF BUBBLE SCATTERING MODELS

This survey is ordered into three "threads," referred to as the "Wildt" thread, the "Devin" thread, and a third "nonlinear" thread. All three threads assume steady state pulsations of spherical bubbles at low frequency, such that the wavelength in the liquid medium is large compared with the bubble size ($\omega R_0/c \ll 1$), and the first two threads assume further that the bubbles undergo sufficiently small pulsations to warrant linear descriptions. The criterion for inclusion of a paper in this review, which covers the period 1945 to 2010, is that it should contain either a novel expression for the scattering cross-section (σ_s), extinction cross-section (σ_e), resonance frequency (ω_{res}) or damping factor (β), or a novel equation of motion that permits calculation of one or more of these. Bubble size, normalized relative to various different length scales, plays an important part in determining its response to sound. These length scales are described in Sec. II A, followed by the three threads in Secs. II B, II C, and II D, and in Sec. II E by a summary of the most important equations.

The two linear threads (named after the originator of the first publication in each, Wildt and Devin), each with its own derivation for the scattering cross-section, have existed separately for more than 30 years (the Devin thread is deemed to have come into existence on publication of Ref. 29), and the parallel existence of two unreconciled threads has infused the literature with many opportunities for misunderstandings. This survey explores these threads and shows how the two derivations are reconciled.

The main focus of the present review, apart from the cross-sections themselves, is on the resonance frequency and the frequency dependence of the viscous, thermal, and acoustic damping terms.

A. Bubble size and resonance

1. Bubble size regimes

The purpose of this section is to provide a framework for clarifying otherwise ambiguous statements about the properties of "small" and "large" bubbles. For example, for each of the three explicitly identified damping terms of (3), there exists a natural length scale on which the size of the bubble can be measured. These length scales are: the acoustic wavelength

$$l_{ac}(\omega) = \frac{2\pi c}{\omega}; \tag{6}$$

a viscous length scale proportional to the square root of the shear viscosity coefficient, η_S, and equal to the thickness that would characterize the microstreaming boundary layer were microstreaming to occur[1,62–64]

$$l_{vis}(\omega) = \sqrt{\frac{2\eta_S}{\rho_{liq}\omega}}, \tag{7}$$

where ρ_{liq} is the equilibrium mass density of the liquid; and the thermal diffusion length

$$l_{th}(\omega, R_0) = \sqrt{\frac{D_P(R_0)}{2\omega}} \tag{8}$$

equal to the thickness of the thermal boundary layer, which is proportional to the square root of D_P, the thermal diffusivity of the gas inside the bubble. The diffusivity is defined in terms of the equilibrium gas density ρ_{gas}, its thermal conductivity K_{gas} and its specific heat capacity at constant pressure C_P as[65]

$$D_P(R_0) \equiv \frac{K_{gas}}{\rho_{gas}(R_0)C_P}, \tag{9}$$

where the dependence on equilibrium bubble radius is caused by the influence of surface tension on the gas pressure (see p. 187 of Ref. 66). Some publications (notably Refs. 19, 67, and 68) define the diffusivity in terms of the specific heat at constant *volume* instead of at constant pressure, i.e.,

$$D_V \equiv \frac{K_{gas}}{\rho_{gas}C_P/\gamma}, \tag{10}$$

where γ is the specific heat ratio. However, Prosperetti now prefers the more common definition (9) because factors of D_V/γ simplify to D_P (A. Prosperetti, personal communication, October 2010). The definition of (9) is adopted for use throughout the present paper.

A bubble can be small or large in any of the acoustical, thermal or viscous senses, independently of the others, although attention is restricted throughout this review to acoustically small bubbles ($R_0 \ll l_{ac}$). Except where stated otherwise, the bubble radius is also assumed large compared to $4\pi\, l_{vis}^2/l_{ac}$.

Further to these, an additional length scale of relevance, denoted $R_{Laplace}$ and referred to henceforth as the "Laplace radius," is the bubble radius at which the Laplace pressure $2\,\tau/R_0$, where τ is the surface tension, is equal to the equilibrium liquid pressure P_{liq}, i.e.,

$$R_{Laplace} = 2\tau/P_{liq}. \tag{11}$$

Throughout this paper reference is made to calculations for a "standard bubble," that is, an air bubble in water at temperature 10 °C and under atmospheric pressure with $\tau = 0.072$ N/m. For such a bubble, the Laplace radius is approximately equal to 1.42 μm.

2. The Minnaert frequency and the diffusion radius

The natural frequency of a bubble that is large on both thermal and Laplace scales was derived by M. Minnaert in his 1933 classic *On Musical Air-Bubbles and the Sounds of Running Water*.[69] This natural frequency, which is denoted $f_M = \omega_M/2\pi$, where

$$\omega_M \equiv \frac{1}{R_0}\sqrt{\frac{3\gamma P_{liq}}{\rho_{liq}}}, \tag{12}$$

is referred to henceworth as the "Minnaert frequency," and is plotted vs bubble radius in Fig. 1. For a "standard bubble" of radius between 30 nm and 300 μm, the Minnaert frequency varies from about 100 MHz to 10 kHz. It differs from the true natural frequency if one or more of the following conditions are not met: $R_0 \ll l_{ac}$ (i.e., the bubbles are "acoustically small"); $R_0 \gg 4\pi\, l_{vis}^2/l_{ac}$, (i.e., the geometric mean of bubble radius and acoustic wavelength is large compared to the viscous boundary layer); $R_0 \gg l_{th}$ (i.e., the bubbles are "thermally large"); $R_0 \gg R_{Laplace}$ (i.e., the Laplace pressure is much less than the equilibrium pressure in the liquid); $\beta \ll \omega$ (the logarithmic decrement is small).

It is stressed that Fig. 1 does not plot the resonance frequency of the bubble, but rather the output of (12) (Minnaert's formulation) and the *adiabatic* resonance frequency ω_{ad}, defined in (17) below and used in Fig. 2(b) as a normalizing parameter to obtain dimensionless frequencies in different

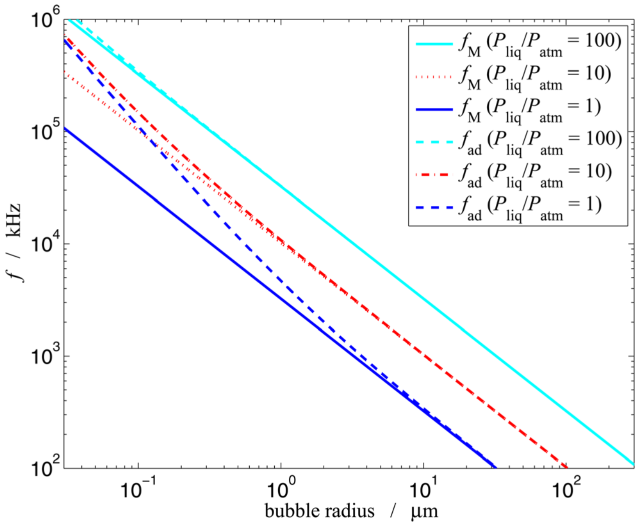

FIG. 1. (Color online) Minnaert requency $f_M = \omega_M/2\pi$ and adiabatic resonance frequency $f_{ad} = \omega_{ad}/2\pi$ vs bubble radius for a standard bubble ($P_{liq} = P_{atm}$) and for increased pressures of 10 P_{atm} and 100 P_{atm}.

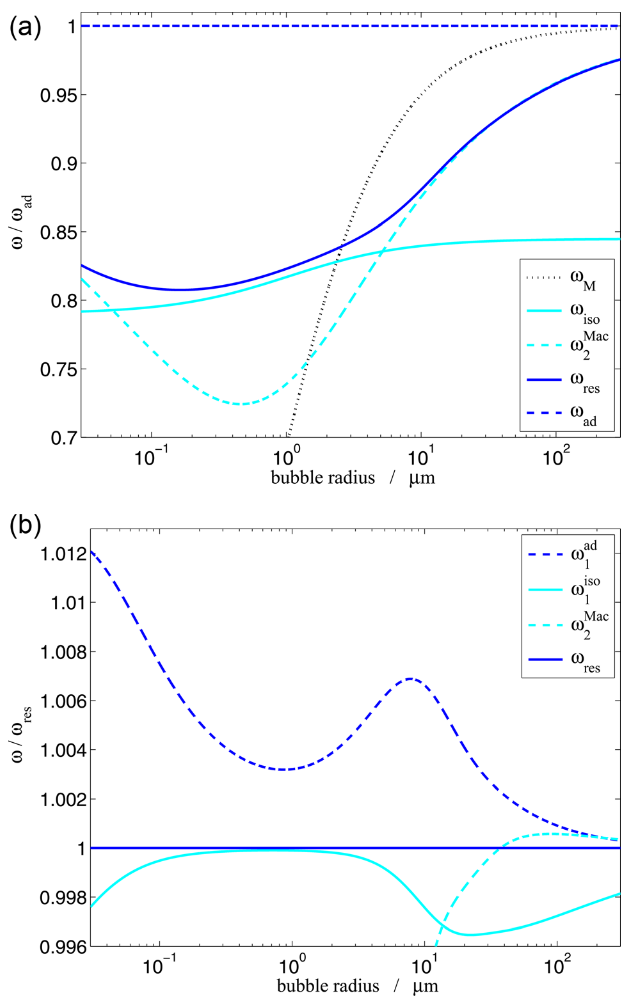

FIG. 2. (Color online) Approximations to the resonance frequency for a standard bubble, normalized by dividing (a) by the adiabatic resonance frequency ω_{ad} and (b) by the true resonance frequency ω_{res}. Individual curves are ω_M:(12); ω_{iso}:(27); ω_2^{Mac}:(26); ω_{res}:(19), converged solution; ω_{ad}: (17); ω_1^{ad}:(21); ω_1^{iso}:(28).

conditions, including those outside the range of conditions for which ω_{ad} approximates the resonance frequency.

It is useful to introduce one final length scale, the "diffusion radius," denoted R_D, as the bubble radius for which the thermal diffusion length evaluated at the Minnaert frequency, and in the absence of surface tension, would be equal to that radius. Using (8), (9), (12) and the ideal gas law relating the gas density ρ_{gas} to absolute temperature T, this definition gives

$$R_D = \frac{K_{gas} k_B T}{2 m_{gas} P_{liq} C_P} \sqrt{\frac{\rho_{liq}}{3\gamma P_{liq}}}, \tag{13}$$

where k_B is Boltzmann's constant and m_{gas} the average mass of a gas molecule. For an air bubble in water at atmospheric pressure and temperature, the diffusion radius is approximately 0.5 μm, comparable in magnitude to the Laplace radius. The implications of this coincidence in order of magnitude are considered in Sec. II A 3 e.

3. Effects of thermal bubble size on resonance frequency

a. General case. It is useful to introduce the "thermal diffusion ratio" $X(\omega, R_0)$, defined as the ratio of the bubble radius to the thermal diffusion length

$$X(\omega, R_0) \equiv \frac{R_0}{l_{th}(\omega, R_0)}. \tag{14}$$

The general case (arbitrary value of X) is considered first, followed by situations for large, intermediate and small X. For an arbitrary thermal diffusion ratio, the resonance frequency, denoted ω_{res}, is the value of ω that satisfies the equation[67,68]

$$\gamma \frac{\omega^2}{\omega_M^2} = \left(1 + \frac{R_{Laplace}}{R_0}\right) \mathrm{Re}\,\Gamma(\omega) - \frac{R_{Laplace}}{3R_0}, \tag{15}$$

where Γ is the complex polytropic index,[31,70] which takes the form[67,71,72]

$$\Gamma(\omega) = \frac{\gamma}{1 - \left\{\frac{(1+i)X/2}{\tanh[(1+i)X/2]} - 1\right\} \frac{6i(\gamma-1)}{X^2}}. \tag{16}$$

b. Thermally large bubble. For bubbles that are thermally large at resonance, the resonance frequency is approximately equal to ω_{ad}, referred to as the "adiabatic resonance frequency," and defined by[73]

$$\omega_{ad} \equiv \omega_M \sqrt{1 + \frac{R_{Laplace}}{R_0}\left(1 - \frac{1}{3\gamma}\right)}. \tag{17}$$

The approximation $\omega_{res} \approx \omega_{ad}$ holds if the diffusion ratio at resonance is large, i.e., if $X(\omega_{res}, R_0) \gg 1$. This condition is approximately equivalent to $X_{ad} \gg 1$, where

$$X_{ad} \equiv X(\omega_{ad}, R_0). \tag{18}$$

c. Bubble of intermediate thermal size. To obtain an expression for the resonance frequency of bubbles of intermediate thermal size, in the sense that order $1/X$ corrections are small but not negligible, one can write an iterative solution to (15) in the form

$$\left(\frac{\omega_N^{res}}{\omega_{ad}}\right)^2 = 1 - \left(1 - \frac{\mathrm{Re}\,\Gamma(\omega_{N-1}^{res})}{\gamma}\right)\nu, \tag{19}$$

where ν is the following dimensionless parameter related to the Laplace radius and specific heat ratio

$$\nu = \nu(R_0) \equiv \frac{1 + \dfrac{R_{Laplace}}{R_0}}{1 + \dfrac{R_{Laplace}}{R_0}\left(1 - \dfrac{1}{3\gamma}\right)}. \tag{20}$$

The exact value of ω_{res}, defined as the solution to (15), is obtained by repeated application of (19) until it converges. This converged solution is plotted (as a ratio, relative to ω_{ad}) in Fig. 2(a) for a standard bubble. Also shown is the Minnaert frequency, normalized in the same way. Other curves are explained below.

A possible choice of seed in (19) is the adiabatic resonance frequency, i.e., $\omega_0^{res} = \omega_{ad}$, which, using ω_1^{ad} to denote the resulting value of ω_1^{res} after a single iteration, gives

$$\left(\frac{\omega_1^{ad}}{\omega_{ad}}\right)^2 = 1 - \left(1 - \frac{\mathrm{Re}\,\Gamma(\omega_{ad})}{\gamma}\right)\nu. \tag{21}$$

The value of ω_1^{ad} is plotted in Fig. 2(b), normalized by ω_{res}. The approximation $\omega_{res} \approx \omega_1^{ad}$ is a good one, incurring an error of less than 1% for a standard bubble with radius exceeding 100 nm.

An approximation to (16) that is accurate for thermally large bubbles (say $X > 6$, such that $\tanh\left[(1+i)X/2\right] \approx 1$) is

$$\Gamma(\omega) \approx \frac{\gamma}{1 + \dfrac{3(\gamma-1)}{X} - \dfrac{3i(\gamma-1)}{X}\left(1 - \dfrac{2}{X}\right)}, \tag{22}$$

the real and imaginary parts of which are given by

$$\frac{\mathrm{Re}\,\Gamma(\omega)}{\gamma} \approx \frac{1 + \dfrac{3(\gamma-1)}{X}}{1 + \dfrac{6(\gamma-1)}{X} + \dfrac{18(\gamma-1)^2}{X^2}\left(1 - \dfrac{2}{X} + \dfrac{2}{X^2}\right)} \tag{23}$$

$$\frac{\mathrm{Im}\,\Gamma(\omega)}{\gamma} \approx \frac{\dfrac{3(\gamma-1)}{X}\left(1 - \dfrac{2}{X}\right)}{1 + \dfrac{6(\gamma-1)}{X} + \dfrac{18(\gamma-1)^2}{X^2}\left(1 - \dfrac{2}{X} + \dfrac{2}{X^2}\right)}. \tag{24}$$

Using this approximation, the iteration produces a Maclaurin series in powers of $1/X_{ad}$. Specifically, using the expansion

$$\frac{1 + \dfrac{3(\gamma-1)}{X}}{1 + \dfrac{6(\gamma-1)}{X} + \dfrac{18(\gamma-1)^2}{X^2}\left(1 - \dfrac{2}{X} + \dfrac{2}{X^2}\right)} = 1 - \frac{3(\gamma-1)}{X} + O\left(\frac{1}{X^3}\right) \tag{25}$$

and choosing $\omega_0^{res} = \omega_{ad}$ as seed in (19), the second iteration yields

$$\left(\frac{\omega_2^{res}}{\omega_{ad}}\right)^2 = 1 - \frac{3(\gamma-1)}{X_{ad}}\nu + \frac{9(\gamma-1)^2}{4X_{ad}^2}\nu^2 + O\left(\frac{1}{X_{ad}^3}\right). \tag{26}$$

The approximation to ω_2^{res} resulting from the sum of the first three terms of (26), denoted ω_2^{Mac}, is plotted in Fig. 2(b).

d. Thermally small bubble. For a thermally small bubble, satisfying $R_0 \ll l_{th}(\omega_{res})$, i.e., one that pulsates isothermally at resonance, the resonance frequency is obtained by putting $\Gamma = 1$ in (15), that is[73]

$$\omega_{iso} \equiv \frac{\omega_M}{\sqrt{\gamma}}\sqrt{1 + \frac{4\tau}{3P_{liq}R_0}}, \tag{27}$$

referred to as the "isothermal resonance frequency" and plotted in Fig. 2(a). The approximation $\omega_{res} \approx \omega_{iso}$ can be seen to hold for a standard bubble of radius in the range 300 nm $< R_0 < 3$ μm, which corresponds approximately to $X < 3$. A better approximation is obtained by use of ω_{iso} as seed in (19), i.e., $\omega_0^{res} = \omega_{iso}$, which gives after one iteration

$$\left(\frac{\omega_1^{iso}}{\omega_{ad}}\right)^2 = 1 - \left[1 - \frac{\mathrm{Re}\,\Gamma(\omega_{iso})}{\gamma}\right]\nu. \tag{28}$$

The error using the approximation $\omega_{res} \approx \omega_1^{iso}$ is less than 0.4% for a standard bubble with radius exceeding 30 nm.

e. Which resonance frequency? Given the various different curves plotted in Fig. 2, the question arises of which of them is appropriate for any given bubble size or range of bubble sizes. Repeated application of (19) provides a converged solution to (15) under any conditions (any value of X). The complication of an iterative method can be avoided if desired by use of simple approximations, for example if the value of X is sufficiently large (ω_2^{Mac} for $X > 5$) or sufficiently small (ω_{iso} for $X < 3$). Alternatively, ω_1^{iso} provides an excellent approximation (error $< 0.4\%$) for all sizes of standard bubble exceeding 30 nm.

Figure 3(a) shows the diffusion ratio at resonance for a standard bubble and variants, demonstrating that truly isothermal conditions at resonance are never reached for bubbles of air in water. The effect of surface tension in perturbing both the gas pressure (which stands as proxy for the oscillator stiffness[74]) and the thermal diffusion length, decreases as the equilibrium pressure in the liquid increases, since both are dependent on how large $2\tau/R_0$ is compared to P_{liq}. Also plotted [Fig. 3(b)] is $X(\omega, R_0)$ evaluated at $\omega = \omega_{ad}$ and $\omega = \omega_{iso}$, each normalized by dividing by $X(\omega_{res}, R_0)$.

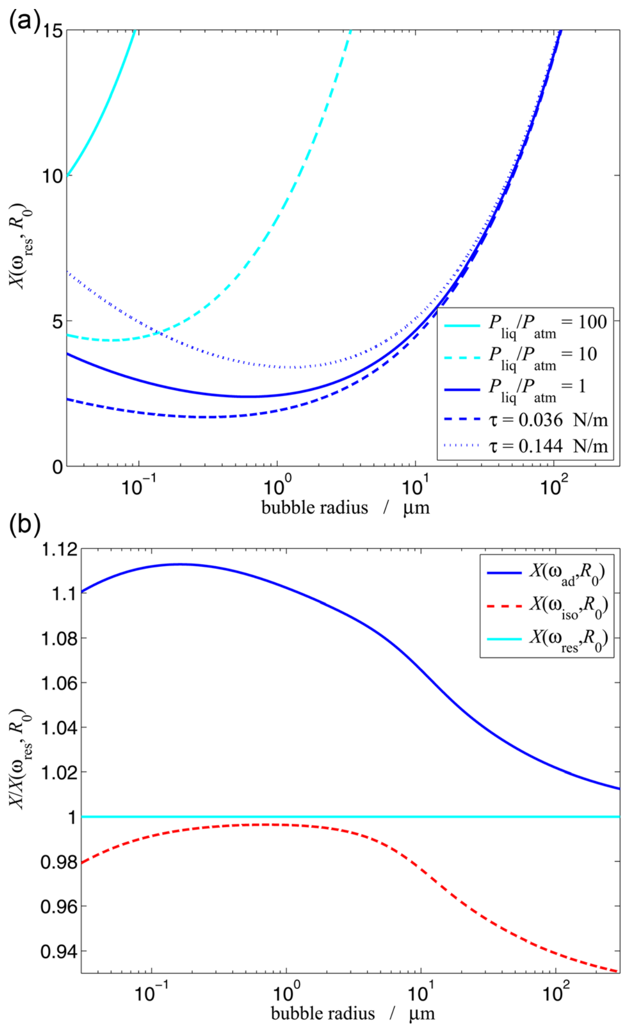

FIG. 3. (Color online) (a) Diffusion ratio $X(\omega_{res}, R_0)$ vs R_0 evaluated for a standard bubble and variants. Surface tension τ is 0.072 N/m for the first three curves in the legend, with pressure between P_{atm} and 100 P_{atm} as shown. The final two curves show the effect of halving and doubling the surface tension for $P_{liq}/P_{atm} = 1$. (b) X/X_{res} vs R_0 for a standard bubble.

Notice that l_{th} is itself a function of R_0, a feature that becomes strong when the condition $R_0 \gg R_{Laplace}$ no longer holds [Refs. 66 (p. 187), 75]. This feature arises through the effect of the Laplace pressure on D_P in (8) and contradicts the widely held belief that, as bubbles become smaller, their behavior tends ever more closely toward isothermal. More specifically, the coincidence in order of magnitude between $R_{Laplace}$ and R_D for a standard bubble (see Sec. II A 2) ensures that once the bubble is small enough to cause a strong departure from adiabatic behavior, the effect of the Laplace pressure increases, meaning that the diffusion ratio *increases* with decreasing bubble radius for small bubbles (Fig. 3). Consequently a standard bubble can never reach the isothermal limit (in the sense of $X \ll 1$) at resonance. This will be important with the expected future development of submicron bubble-based ultrasound contrast agents.[75,76]

B. Wildt thread

The detailed survey of bubble scattering models begins here. The "Wildt" thread is the first of three and encompasses results from Refs. 19, 20, 67, 77, 78, and Eqs. (25) and (58) from Ref. 33.

1. Wildt 1946

Compared with pre-Second World War knowledge, Ref. 77 shows an impressively detailed understanding of a difficult problem. Chapter 28 of that report, though anonymous, is referred to here as "Wildt 1946" because it was edited by R. Wildt (and published in 1946). Wildt 1946 considers a plane pressure wave (p_i) of amplitude A traveling toward $+\infty$ in the Cartesian x axis:

$$p_i = A \exp[i\omega(t - x/c)] \tag{29}$$

and incident on a bubble at the origin, such that the scattered field may be described by a divergent spherical pressure wave (p_s) whose amplitude is inversely proportional to distance r from the bubble center:

$$p_s = (B/r) \exp[i\omega(t - r/c)]. \tag{30}$$

If (29) and (30) accurately represent the incident and scattered waves (implying, for example, spherical symmetry and an infinite uniform inviscid liquid) then the scattering cross-section can be related to the normalized amplitude of the scattered wave $|B/A|$ through the ratio of the scattered power $4\pi r^2\ |p_s|^2/(2\rho_{liq}c)$ to the incident intensity $|p_i|^2/(2\rho_{liq}c)$, giving

$$\sigma_s = 4\pi|B/A|^2. \tag{31}$$

The ratio B/A is obtained through the following steps. First Euler's equation is applied to the scattered wave and the result evaluated at the bubble wall, yielding an equation for the volume velocity.[79] A second equation for volume velocity is found by differentiating the ideal gas law with respect to time. The two equations so derived are solved for the pressure in the interior of the bubble, which is then matched to the sum of incident and scattered pressures, evaluated at the bubble wall. The resulting equation is then rearranged for B/A, giving Wildt's result

$$\sigma_s = \frac{4\pi R_0^2}{\left(\omega_{res}^2/\omega^2 - 1\right)^2 + \delta_{Wildt}^2}, \tag{32}$$

where

$$\delta_{Wildt} \approx \varepsilon + \frac{\omega_{res}^2}{\omega^2} b_{th} + \frac{b_{vis}}{\rho_{liq} c \varepsilon}, \tag{33}$$

and ε is the dimensionless frequency

$$\varepsilon = \varepsilon(\omega) \equiv \omega R_0/c. \tag{34}$$

Wildt's scattering cross-section σ_s and damping coefficient δ_{Wildt}, calculated using (32) and (33), are plotted in Fig. 4 and Fig. 5(a), respectively. If the random error is not quantified, deviations of measured damping from predictions

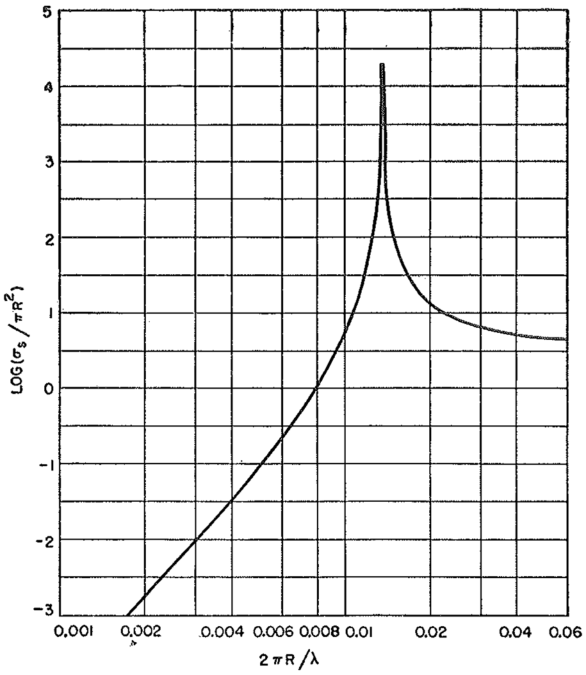

FIG. 4. Scattering cross-section of a single bubble in the form $\log_{10}[\sigma_s/(\pi R_0^2)]$, plotted vs dimensionless frequency ε (from Ref. 77).

should not be attributed to it, and indeed careful quantification of such errors shows that they can be much less than the discrepancy (see for example Fig. 6 of Ref. 80). Hence there is a more important source of discrepancy to consider. This arises because the conditions of theory (a single spherical bubble in an infinite body of liquid) are rarely realized in practice: Fig. 5(b) shows that the apparent discrepancy between the measured data points and the damping theory of Devin (solid line) are due to the radiation damping departing from its free field value as a result of the failure to achieve free field conditions during the experiment, as predicted by the dashed curve [Fig. 5(b)].[81–83] This illustrates the following more general point: The idealized conditions implied by the simple bubble scattering models described in this review (a single spherical bubbles in free field in an infinite body of liquid) are extremely difficult to produce in a controlled manner, making it necessary to understand departures from these conditions for studying even the simplest real-world scattering situations, for which a pre-requisite is a clear understanding of the single free-field bubble theory itself.

The parameter b_{vis} in (33) (denoted C_1 by Wildt) is "a constant measuring the effect of friction, which is assumed to be proportional to the radial velocity dR/dt of the bubble."[77] The parameter b_{th} (denoted β by Wildt) is introduced to take into account the "phase shift between pressure and temperature on one hand, and volume and radial velocity of the bubble on the other hand [by] inserting a complex factor $1 - \mathrm{i}b_{\mathrm{th}}$ in the right-hand side of [the equation relating the radial velocity of the bubble wall to the rate of change of incident pressure]."[77] For the extinction cross-section, Wildt 1946 gives:

$$\sigma_{\mathrm{e}} = \sigma_{\mathrm{s}}\left[1 + \frac{1}{\varepsilon}\left(\frac{\omega_{\mathrm{res}}^2}{\omega^2}b_{\mathrm{th}} + \frac{b_{\mathrm{vis}}}{\rho_{\mathrm{liq}}c\varepsilon}\right)\right]. \qquad (35)$$

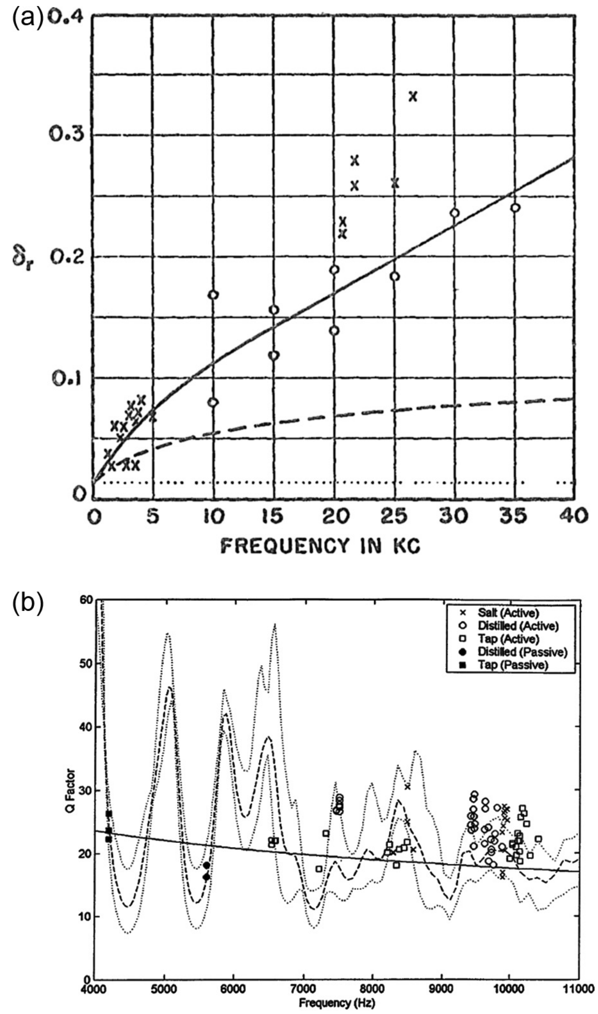

FIG. 5. (a) Damping coefficient at resonance vs resonance frequency $\omega_{\mathrm{res}}/(2\pi)$ in kilohertz. Measured values (symbols and solid curve) exceed theoretical predictions (dashed curve δ_{Wildt}). (from Ref. 77). (b) Quality factor (reciprocal of damping constant at resonance) vs resonance frequency $\omega_{\mathrm{res}}/(2\pi)$ in hertz. Measured values (symbols) are scattered around uncorrected theoretical predictions (solid black curve) due to effects of reverberation (dashed curve, the limits of uncertainty being indicated by the gray curves above and below this line). The experimental error is much less than the discrepancy between measurements and free field theory (from Ref. 82).

The relationships between the parameters b_{th} and b_{vis} used by Wildt, and physical parameters such as shear viscosity η_{S} and thermal diffusivity (via the complex polytropic index) are described in Sec. II E.

Wildt 1946 shows that for thermally large bubbles with a negligible Laplace pressure, the resonance frequency is equal to the Minnaert frequency, and states further that "For very small bubbles, with radii less than [10 μm], surface tension becomes important and the compressions and expansions of the gas in the bubble become isothermal instead of adiabatic." He also states that under these conditions ω_{res} is given by ω_{iso}, attributing this theoretical result to Spitzer,[84] although this assertion neglects the effect of surface tension on thermal diffusivity, requiring that the bubbles concerned be large on the Laplace scale and small on the thermal scale. However, as the bubble size tends to zero, the Laplace pressure increases, thus increasing the gas density and decreasing

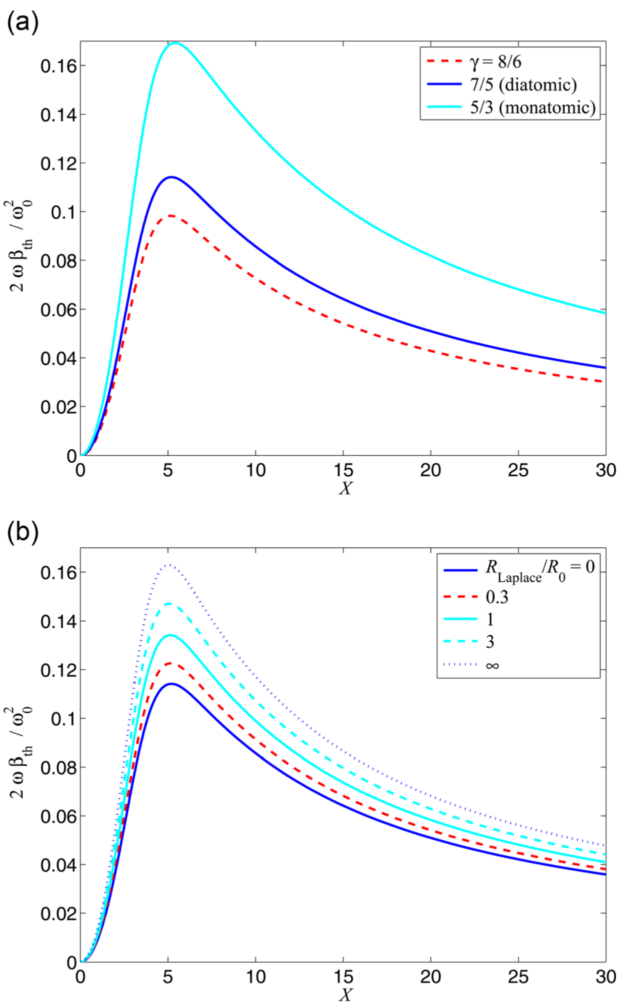

FIG. 6. (Color online) Dimensionless thermal damping parameter $2\omega\,\beta_{th}/\omega_0^2$ vs diffusion ratio X: (a) Effect of specific heat ratio and (b) Effect of dimensionless bubble radius $R_0/R_{Laplace}$.

its diffusivity and diffusion length, potentially making the bubble thermally *large* as it becomes smaller and smaller in an absolute sense [Ref. 66, p. 187]. Nevertheless, examination of Fig. 2(b) demonstrates that ω_{iso} is a good approximation to the resonance frequency for a standard bubble with radius in the range 300 nm to 3 μm.

2. Andreeva 1964

The scattering model proposed by Andreeva,[78] intended for application to a fish swimbladder (approximated as a spherical gas bubble surrounded by fish flesh), is of the form

$$\sigma_s = \frac{4\pi R_0^2}{\left(\frac{\omega_{res}^2}{\omega^2}-1\right)^2 + \left(\frac{1}{Q_{rad}}\frac{\omega_{res}}{\omega} + \frac{1}{Q_{th}}\frac{\omega_{res}}{\omega} + \frac{1}{Q_{vis}}\frac{\omega_{res}}{\omega}\right)^2}, \quad (36)$$

where the Q-factors, which Andreeva attributes to Devin[85] (see Sec. II C 1), are

$$\frac{1}{Q_{rad}} = \frac{\omega_{res} R_0}{c}, \quad (37)$$

$$\frac{1}{Q_{th}} = \frac{3(\gamma-1)}{R_0}\sqrt{\frac{D_P}{2\omega_{res}}}, \quad (38)$$

$$\frac{1}{Q_{vis}} = \frac{4\,\mathrm{Im}\,\mu}{\rho_{flesh} R_0^2 \omega_{res}^2}, \quad (39)$$

and where μ and ρ_{flesh} are the shear modulus and density of fish flesh.

Including the effect of flesh rigidity on resonance frequency here by exception [for consistency with (39)], the resonance frequency is given by

$$\frac{\omega_{res}^2}{\omega_M^2} = 1 + \frac{4\,\mathrm{Re}\,\mu}{3\gamma P_{flesh}}, \quad (40)$$

where P_{flesh} is the equilibrium pressure in the fish flesh.

3. Weston 1967

Weston[19] laid the foundation for nearly all subsequent work on scattering and absorption due to fish with a swimbladder. Based partly on Ref. 78, Weston's model provides a theoretical framework for describing scattering from large bubbles (that is, $R_0 \gg l_{th}$ and $R_0 \gg R_{Laplace}$, although $R_0 \ll l_{ac}$), accounting for both thermal and radiation damping.

Corrections due to the non-spherical shape of a fish bladder are excluded here. With this and other simplifications, Weston's expression for the scattered pressure, as described in an unpublished report [D. E. Weston, "Assessment methods for biological scattering and attenuation in ocean acoustics," BAeSEMA Report C3305/7/TR-1, April 1995], can be written

$$AR_0/B = \omega_{res}^2/\omega^2 - 1 - \mathrm{i}\delta_{Weston}, \quad (41)$$

where

$$\delta_{Weston} = \frac{1}{Q_{rad}}\frac{\omega_{res}}{\omega} + \frac{1}{Q_{th}}\left(\frac{\omega_{res}}{\omega}\right)^{5/2} + \frac{1}{Q_{vis}}\left(\frac{\omega_{res}}{\omega}\right)^2. \quad (42)$$

The Q-factors Q_{rad}, Q_{th} and Q_{vis} are given by (37), (38), and (39) (as Andreeva). It follows from (31) and (41) that

$$\sigma_s = \frac{4\pi R_0^2}{\left(\omega_{res}^2/\omega^2 - 1\right)^2 + \delta_{Weston}^2}. \quad (43)$$

The extinction cross-section is

$$\sigma_e = \sigma_s\left[1 + \frac{Q_{rad}}{Q_{th}}\left(\frac{\omega_{res}}{\omega}\right)^{7/2} + \frac{Q_{rad}}{Q_{vis}}\left(\frac{\omega_{res}}{\omega}\right)^3\right]. \quad (44)$$

4. Chapman and Plesset 1971

Chapman and Plesset[67] calculated the damping constant β for unforced pulsations, taking into account acoustic, viscous, and thermal damping. Their full method, not reproduced here, is valid for any value (large or small) of the ratio $2\pi\beta/\omega_{\mathrm{nat}}$ (the logarithmic decrement[43]), where ω_{nat} is the natural frequency. If the logarithmic decrement is small, their result simplifies to

$$\beta = \frac{R_0\omega_{\mathrm{nat}}^2}{2c} + \frac{3P_{\mathrm{gas}}}{2\rho_{\mathrm{liq}}R_0^2\omega_{\mathrm{nat}}}\mathrm{Im}\Gamma + \frac{2\eta_{\mathrm{S}}}{\rho_{\mathrm{liq}}R_0^2}, \tag{45}$$

where P_{gas} is the equilibrium pressure inside the bubble

$$P_{\mathrm{gas}} = P_{\mathrm{liq}} + \frac{2\tau}{R_0}. \tag{46}$$

Neglecting thermal and acoustic damping, the natural frequency derived by Chapman and Plesset is

$$\omega_{\mathrm{nat}} = \frac{1}{R_0}\sqrt{\frac{1}{\rho_{\mathrm{liq}}}\left(3P_{\mathrm{gas}}\kappa - \frac{2\tau}{R_0}\right) - \frac{4\eta_{\mathrm{S}}^2}{\rho_{\mathrm{liq}}^2R_0^2}}, \tag{47}$$

where κ is the (real) polytropic exponent, that is,

$$\kappa = \mathrm{Re}\,\Gamma(\omega_{\mathrm{nat}}), \tag{48}$$

the value of which is obtained by Chapman and Plesset using an iterative method and plotted vs radius for a standard bubble.

5. Love 1978

Love[20] built on Weston's work by including a more realistic model of the fish flesh surrounding its swimbladder. A subtle but important novelty is that, in contrast to the complex shear modulus introduced by Andreeva, Love modeled the fish flesh as a viscous liquid medium (with shear and bulk viscosities coefficients η_{S} and η_{B} and surface tension τ). Love's model is presented here, simplified—by extending the (liquid) flesh properties to occupy the entire medium—in order to facilitate comparisons with other models. It is the first bubble scattering model known to the present authors to introduce an explicit term associated with *bulk* viscosity. Thermal losses are included in the damping coefficient, while the resonance frequency—like Weston's—is not adjusted for thermal effects, probably because these were considered unimportant for a fish bladder. The scattering cross-section [Love's Eq. (63)] is

$$\sigma_{\mathrm{s}} = \frac{4\pi R_0^2}{\left(\frac{\omega_{\mathrm{res}}^2}{\omega^2} - 1\right)^2 + \left(2\frac{\beta_{\mathrm{th}} + \beta_{\mathrm{vis}}}{\omega} + \varepsilon\right)^2}, \tag{49}$$

with [Love's Eq. (61)]

$$2\frac{\beta_{\mathrm{vis}}}{\omega} = \frac{2}{3}\frac{4\eta_{\mathrm{S}} + 3\eta_{\mathrm{B}}}{\omega\rho_{\mathrm{liq}}R_0^2}, \tag{50}$$

$$2\frac{\beta_{\mathrm{th}}}{\omega} = \left(1 + \frac{2\tau}{\rho_{\mathrm{liq}}\omega^2R_0^3}\right)\frac{3(\gamma-1)}{X(\omega,R_0)}, \tag{51}$$

where $X(\omega,R_0)$ is the thermal diffusion ratio given by (14).

The damping factors β_{th} and β_{vis} are contributions due to thermal and viscous damping to the total damping factor β, which is defined as half of the coefficient of the $\mathrm{d}R/\mathrm{d}t$ term in the differential equation (62) describing the motion of the bubble wall $R(t)$. Love's Eq. (58) approximates the resonance frequency as ω_{ad} from (17).

6. Ainslie and Leighton 2009

a. Wildt-Weston method. The procedure followed by Ainslie and Leighton[33] to derive their Eq. (25) is identical to that of both Wildt and Weston, except for the introduction of a (complex) polytropic index[31,70] Γ to describe net heat flux across the bubble wall, and setting of the shear viscosity and surface tension to zero to aid clarity. The surface tension term is reintroduced here, giving

$$\frac{B}{A} = \frac{R_0}{\left[(1+\mathrm{i}\varepsilon)\Omega^2/\omega^2 - 1\right]\mathrm{e}^{-\mathrm{i}\varepsilon}}, \tag{52}$$

where Ω is related to the equilibrium gas pressure P_{gas}, from (46), and the complex polytropic index Γ, from (16), via

$$\frac{\rho_{\mathrm{liq}}R_0^2}{3}\Omega(R_0,\omega)^2 = \Gamma(R_0,\omega)P_{\mathrm{gas}} - \frac{2\tau}{3R_0}. \tag{53}$$

The right hand side of (53) is the bubble's bulk modulus,[86] the imaginary part of which represents the phase lag of the bubble's response to the applied pressure.

This procedure leads to the following equation for the scattering cross-section

$$\sigma_{\mathrm{s}} = \frac{4\pi R_0^2}{\left(\frac{\omega_0^2}{\omega^2} - 1 - 2\frac{\beta_{\mathrm{th}}}{\omega}\varepsilon\right)^2 + \left(2\frac{\beta_{\mathrm{th}}}{\omega} + \frac{\omega_0^2}{\omega^2}\varepsilon\right)^2}, \tag{54}$$

where

$$\omega_0(R_0,\omega) \equiv \sqrt{\mathrm{Re}\left[\Omega(R_0,\omega)^2\right]} \tag{55}$$

$$\beta_{\mathrm{th}}(R_0,\omega) \equiv \frac{\mathrm{Im}\left[\Omega(R_0,\omega)^2\right]}{2\omega}. \tag{56}$$

The frequency ω_0 is closely related to the bubble's resonance frequency ω_{res}, but it is not correct to state that ω_0 is *equal* to the resonance frequency except in conditions for which Ω is independent of frequency. Furthermore, the presence of damping can shift the resonance peak away from the undamped resonance frequency, although the size (and direction) of this shift depends on the precise definition of "resonance frequency." See Sec. III for details.

The frequency dependence of the radiation damping term in the denominator of (54) is the same as that in Weston's expression for the scattering cross-section, i.e., (42) from Sec. II B 3 above, as can be seen by setting $\beta_{\mathrm{th}} = 0$.

Equation (56) is consistent with Weston's thermal damping. Its consequences for the calculation of thermal damping are explored next. Specifically, combining it with (55) and (53) gives

$$\frac{2\omega\beta_{\rm th}}{\omega_0^2} = \frac{\mathrm{Im}\Gamma}{\mathrm{Re}\,\Gamma - \dfrac{1}{3\left(1 + R_0/R_{\rm Laplace}\right)}}, \tag{57}$$

the right hand side of which is a function of the following three parameters: diffusion ratio X, specific heat ratio γ and the ratio $R_0/R_{\rm Laplace}$. Figure 6 shows this function plotted vs X, first with γ as a variable parameter and negligible surface tension ($R_{\rm Laplace}/R_0 = 0$) [Fig. 6(a)] and then with $R_0/R_{\rm Laplace}$ as a variable parameter and fixed specific heat ratio ($\gamma = 1.4$) [Fig. 6(b)]. The complex polytropic index from Ref. 72 is used, in the form of (16).

b. Breathing mode method. A weakness of the approach pioneered in Wildt (1946) and improved by Weston is the need to assume small ε early on in the derivation, casting possible doubt on the validity of subsequent expansions in this parameter. This weakness is addressed in Ref. 33, at the expense of neglecting thermal and viscous effects, by considering the first term only in Anderson's expansion for the scattered field[87] (referred to as the "breathing mode" because of its spherical symmetry) and delaying the small ε expansion to the end.

This breathing mode derivation for the complex ratio B/A, based on Anderson's normal mode expansion, and incorporating the effect of a non-zero gas density, gives

$$\frac{A}{B}R_0 = \frac{\dfrac{\omega_0^2}{\omega^2}\varepsilon\sin\varepsilon - \left[\dfrac{3}{\xi^2}\left(1 - \dfrac{\xi}{\tan\xi}\right) - \dfrac{\omega_0^2}{\omega^2}\right]\cos\varepsilon}{\dfrac{\omega_0^2}{\omega^2}\cos\varepsilon + \left[\dfrac{3}{\xi^2}\left(1 - \dfrac{\xi}{\tan\xi}\right) - \dfrac{\omega_0^2}{\omega^2}\right]\dfrac{\sin\varepsilon}{\varepsilon}} + \mathrm{i}\varepsilon, \tag{58}$$

where

$$\xi = \frac{\omega}{\omega_0}\sqrt{\frac{\rho_{\rm gas}}{\rho_{\rm liq}}}. \tag{59}$$

Taking the squared modulus of (58) and expanding numerator and denominator separately about $\varepsilon = 0$ gives [Eq. (58) of Ref. 33]

$$\sigma_{\rm s} \approx 4\pi R_0^2 \frac{1 - \dfrac{\varepsilon^2}{3}\left(1 + 2\dfrac{\omega_0^2}{\omega^2}\right) + \dfrac{2}{15}\dfrac{\rho_{\rm gas}}{\rho_{\rm liq}}\dfrac{\omega^2}{\omega_0^2}}{\left(\dfrac{\omega_0^2}{\omega^2} - 1 - \dfrac{1}{15}\dfrac{\rho_{\rm gas}}{\rho_{\rm liq}}\dfrac{\omega^2}{\omega_0^2}\right)^2 + \varepsilon^2\dfrac{\omega_0^4}{\omega^4}}, \tag{60}$$

the denominator of which is again consistent with Weston's radiation damping, as can be seen by substituting $\rho_{\rm gas} = 0$ in (60) and comparing the result with (42). The resonance frequency based on (60) is

$$\left(\frac{\omega_0}{\omega_{\rm res}}\right)^2 = 1 - \frac{(\omega_0 R_0/c)^2}{2} + \frac{\rho_{\rm gas}}{15\rho_{\rm liq}}. \tag{61}$$

The magnitudes of the two correction terms on the right hand side of (61) are in the ratio $45k_{\rm B}T\,\mathrm{Re}\Gamma/(2m_{\rm gas}c^2)$, which by a curious numerical coincidence, for a standard bubble is close to unity.[33] The correction terms therefore approximately cancel.

C. Devin thread

The "Devin" thread encompasses results from Refs. 29, 68, and 85 and Eqs. (43) and (67) from Ref. 33. The requirement for a result to be included in the Devin thread is for it to originate from a linear differential equation relating the bubble radial displacement to the forcing pressure. This equation can be written in terms of the damping factor β and a stiffness parameter K

$$\ddot{R} + 2\beta\dot{R} + K(R - R_0) = F\mathrm{e}^{+\mathrm{i}\omega t}, \tag{62}$$

where the forcing term is

$$F = -\frac{A}{\rho_{\rm liq}R_0}. \tag{63}$$

The stiffness parameter K is a function of frequency approximately equal to ω_0^2 (see Sec. III C 2), which from (55) is the real part of Ω^2, by definition.[33]

1. Devin 1959

Devin's paper[85] is partly based on pioneering work by Pfriem,[88] who calculated the effect of a gas bubble's thermal diffusivity on its resonance frequency and damping factor. Devin developed the first comprehensive bubble damping model by extending Pfriem's calculations of thermal damping to include acoustic and viscous effects. Devin's stiffness and damping terms are described below.

a. Stiffness term (K). Devin's Eq. (65) for the stiffness parameter K in (62) is

$$K = \frac{3}{\rho_{\rm liq}R_0^2}\left(P_{\rm gas}\mathrm{Re}\,\Gamma - \frac{2\tau}{3R_0}\right). \tag{64}$$

The ratio $\gamma/\mathrm{Re}\Gamma$ (denoted α by Devin) is

$$\frac{\gamma}{\mathrm{Re}\,\Gamma} = \left[1 + \frac{9(\gamma-1)^2}{X^2}\zeta(X)^2\right]\left[1 + \frac{3(\gamma-1)}{X}\frac{\sinh X - \sin X}{\cosh X - \cos X}\right], \tag{65}$$

where

$$\zeta(X) \equiv \frac{\sinh X + \sin X - 2(\cosh X - \cos X)/X}{\cosh X - \cos X + 3(\gamma-1)(\sinh X - \sin X)/X}, \tag{66}$$

and the diffusion ratio X is given by (14).

For thermally large bubbles ($X > 10$), (65) becomes [from Devin's Eq. (58), which corrects Pfriem's Eq. (17a)]

$$\frac{\gamma}{\mathrm{Re}\,\Gamma} \approx 1 + \frac{3(\gamma-1)}{X} + \frac{9(\gamma-1)^2}{X^2}, \tag{67}$$

and Devin's Eq. (66) gives the corresponding resonance frequency as

$$\omega_{\rm res}^2 \approx \omega_{\rm M}^2\left[1 + \frac{2\tau}{R_0 P_{\rm liq}}\left(1 - \frac{1}{3\mathrm{Re}\,\Gamma}\right)\right] \times \left[1 + \frac{3(\gamma-1)}{X} + \frac{9(\gamma-1)^2}{X^2}\right]^{-1}, \tag{68}$$

where the approximation requires $R_0 \gg R_{\text{Laplace}}$ and $X \gg 1$. A practical difficulty with the use of (68) is that the terms X and Γ on the right hand side are both functions of frequency, which for consistency with the left hand side would need to be evaluated at the resonance frequency ω_{res}, making it an implicit equation, requiring iterative or graphical solution. By contrast, Eqs. (21), (26), and (28) of Sec. II A 3 are all explicit equations, for ω_1^{ad}, ω_2^{res}, and ω_1^{iso}, respectively, as their right hand sides are functions of bubble radius R_0 only (and not frequency).

For smaller values of X (say $X < 3$) (65) becomes [Devin's Eq. (59)]:

$$\frac{\gamma}{\operatorname{Re}\Gamma} \approx \gamma - \frac{X^4}{1890}\left[1 - 2.1\frac{(\gamma-1)^2}{\gamma}\right]. \tag{69}$$

The condition $X < 3$ requires the bubble radius to be less than a third of the thermal diffusion length, but this does not require the bubble to be small in an absolute sense. One reason for making this distinction is that, for fixed bubble radius, the thermal diffusion length increases with decreasing frequency. Another is that, even if interest is limited to effects at resonance, because of the effect of surface tension on gas density (and therefore on its diffusivity), for very small bubbles the value of $X(\omega_{\text{res}})$ starts to *increase* with decreasing bubble size, causing the polytropic index to recede from its isothermal value of unity (see Sec. II A).

b. Damping factor (β). Devin provided a comprehensive treatment of bubble damping by consolidating Pfriem's theoretical analysis of thermal damping, including corrections for the surface tension, and combining this with expressions for acoustic radiation and viscous damping. His equation for the damping factor β in (62) is

$$\beta = \beta_{\text{th}} + \beta_{\text{vis}} + \frac{\omega}{2}\varepsilon, \tag{70}$$

where

$$\beta_{\text{vis}} = \frac{2\eta_S}{\rho_{\text{liq}} R_0^2} \tag{71}$$

and the thermal damping factor is [Devin's Eq. (54)]

$$\beta_{\text{th}} = \frac{\omega}{2}\frac{3(\gamma-1)}{X}\zeta(X)\frac{K}{\omega^2}. \tag{72}$$

Approximate forms for thermal damping are [Devin's Eq. (55), valid for bubbles satisfying $X > 6$]

$$\beta_{\text{th}} \approx \frac{\omega}{2}\frac{3(\gamma-1)}{X}\frac{1-2/X}{1+3(\gamma-1)/X}\frac{K}{\omega^2}, \tag{73}$$

and [Devin's Eq. (56), valid for bubbles satisfying $X < 2$]

$$\beta_{\text{th}} \approx \frac{\omega}{2}\frac{\gamma-1}{30\gamma}X^2\frac{K}{\omega^2}. \tag{74}$$

The dimensionless thermal damping parameter $2\,\omega\,\beta_{\text{th}}/\omega_0^2$ is calculated using (72) and (74), with the approximation $\omega_0^2 \approx K$, and the result plotted as a function of the diffusion ratio X in Fig. 7(a). The purpose is to see how these two approximations compare with the more accurate expression obtained by combining (57) with (16) for Γ, and evaluated for the situation $\tau = 0$ in order to permit a like comparison with the other two curves. The result is $\operatorname{Im}\Gamma/\operatorname{Re}\Gamma$, which is the third curve plotted in this graph. The effect of a non-zero surface tension would be to increase the value of $2\,\omega\,\beta_{\text{th}}/\omega_0{}^2$ by an amount that depends on bubble size and liquid pressure.

The graph demonstrates that (74) is in good agreement with $\operatorname{Im}\Gamma/\operatorname{Re}\Gamma$ for small X (the error is less than 3% for $X < 2$ and less than 1% for $X < 1.5$). Similarly, (72) agrees with $\operatorname{Im}\Gamma/\operatorname{Re}\Gamma$ to within 1% for $X > 4.5$. At intermediate values of X, (57) is needed for accurate results, for example if 1% accuracy or better is desired in the range $1.5 < X < 4.5$.

Approximations for large X are compared in Fig. 7(b). For $X > 6$, (73) [Devin's Eq. (55)] is accurate to within 0.5%

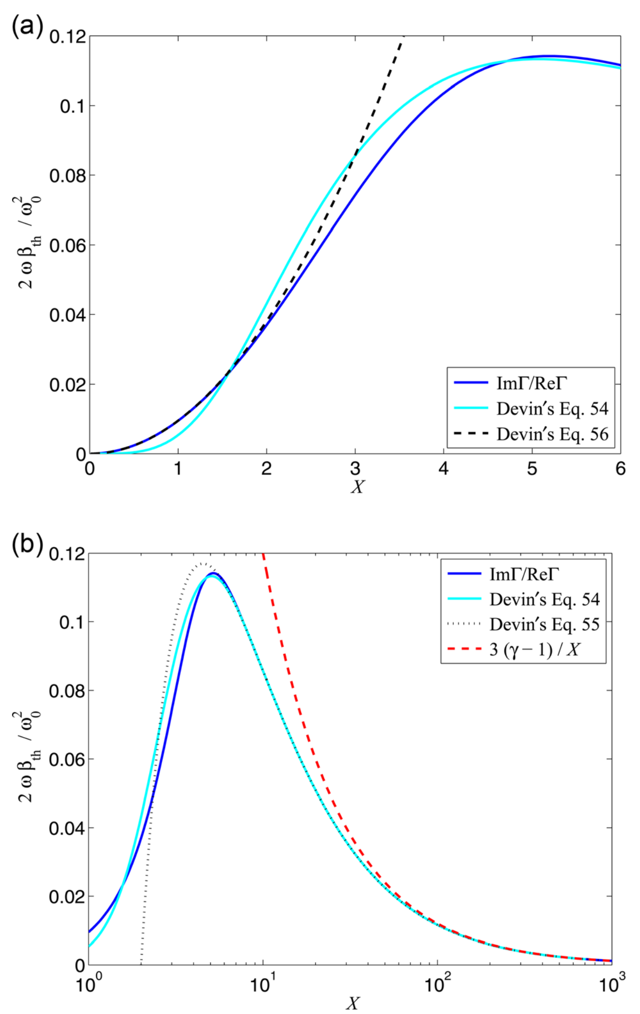

FIG. 7. (Color online) Dimensionless thermal damping parameter $2\omega\beta_{\text{th}}/\omega_0{}^2$ vs diffusion ratio: Approximations for (a) small X and (b) large X. Plots are shown for: $\operatorname{Im}\Gamma/\operatorname{Re}\Gamma$ given by Eq. (57) with $\tau = 0$; Devin's Eq. (54) [Eq. (72) of this paper]; Devin's Eq. (56) [Eq. (74) of this paper]; Devin's Eq. (55) [Eq. (73) of this paper]; $3(\gamma-1)/X$ [Eq. (75) of this paper].

of $\mathrm{Im}\Gamma/\mathrm{Re}\Gamma$ and (76), [Devin's Eq. (54)] to within 1%. The single term approximation [large X limit of (73)]

$$\beta_{\mathrm{th}} \approx \frac{\omega}{2}\frac{3(\gamma-1)}{X}\frac{K}{\omega^2}, \tag{75}$$

is accurate to first order in $1/X$, with errors exceeding 10% for $X < 30$.

2. Medwin 1977

Medwin[29] realized that Devin's results could be used to obtain expressions for scattering and extinction cross-sections that are uniformly valid across the entire range of thermal bubble sizes from isothermal to adiabatic. Substituting Devin's expressions for the damping factors (in the form used by Eller for quantifying off resonance effects[89]) into Wildt's expression for σ_s, he obtained

$$\sigma_s = \frac{4\pi R_0^2}{\left(\omega_{\mathrm{res}}^2/\omega^2 - 1\right)^2 + \delta^2} \tag{76}$$

and

$$\sigma_e = \sigma_s\frac{\delta}{\varepsilon}, \tag{77}$$

where

$$\delta = \frac{4\eta_S}{\rho_{\mathrm{liq}}R_0^2\omega} + \frac{3(\gamma-1)}{X}\zeta(X)\frac{K}{\omega^2} + \varepsilon. \tag{78}$$

Here K is the bubble stiffness, given by (64) and $\zeta(X)$ is given by (66).

3. Prosperetti 1977

Prosperetti[68] derived expressions for radiation, thermal and viscous damping, as well as for the extinction cross-section. His equation of motion for the increase in radius from equilibrium is given by (62) with

$$K = 3\frac{P_{\mathrm{gas}}}{\rho_{\mathrm{liq}}R_0^2}\mathrm{Re}\,\Gamma - \frac{2\tau}{\rho_{\mathrm{liq}}R_0^3} + \frac{\varepsilon^2}{1+\varepsilon^2}\omega^2 \tag{79}$$

and

$$\beta = 2\frac{\eta_S + \eta_{\mathrm{th}}}{\rho_{\mathrm{liq}}R_0^2} + \frac{\omega}{2}\frac{\varepsilon}{1+\varepsilon^2}. \tag{80}$$

It follows from (56) that

$$\eta_{\mathrm{th}} = \frac{\rho_{\mathrm{liq}}R_0^2}{4\omega}\mathrm{Im}\left(\Omega^2\right). \tag{81}$$

The extinction cross-section (correcting here a factor 2 error in Prosperetti's unnumbered equation for σ_e at the top of p. 19 of Ref. 68) is

$$\sigma_e = \frac{4\pi R_0^2}{\left(K/\omega^2 - 1\right)^2 + \left(2\beta/\omega\right)^2}\frac{2\beta/\omega}{\varepsilon}. \tag{82}$$

4. Ainslie and Leighton 2009

It is useful here to visit the discrepancy in the frequency dependence between Weston's radiation damping coefficient (proportional to ω^{-1}) and that of Wildt or Medwin (ostensibly proportional to ω). The discrepancy was pointed out first by Weston himself, even before his 1967 paper[42] and later by Anderson and Hampton.[22] It then went unnoticed for a further quarter of a century until it was highlighted[32] and finally resolved by Ainslie and Leighton[33] by showing that the Weston and Wildt derivations are in essence identical: their outcomes differ only because of an omitted $O(\varepsilon^2)$ term in one of the power expansions used in Ref. 77.

Starting from the differential equation of Prosperetti,[68] Ref. 33 obtains the following equation for the ratio of incident pressure to the bubble's radial velocity $\mathrm{d}R/\mathrm{d}t$

$$\frac{A}{\mathrm{d}R/\mathrm{d}t} = \mathrm{i}\rho_{\mathrm{liq}}c\varepsilon\left(\frac{K}{\omega^2} - 1 + 2\mathrm{i}\frac{\beta}{\omega}\right). \tag{83}$$

Applying Euler's equation to the scattered pressure field and eliminating the radial velocity results in

$$\frac{AR_0}{B}\exp(\mathrm{i}\varepsilon) = \frac{\omega_0^2}{\omega^2} - 1 - 2\frac{\beta_0}{\omega}\varepsilon + \mathrm{i}\left(2\frac{\beta_0}{\omega} + \frac{\omega_0^2}{\omega^2}\varepsilon\right), \tag{84}$$

from which the scattering cross-section follows as

$$\sigma_s = \frac{4\pi R_0^2}{\left(\frac{\omega_0^2}{\omega^2} - 1 - 2\frac{\beta_0}{\omega}\varepsilon\right)^2 + \left(2\frac{\beta_0}{\omega} + \frac{\omega_0^2}{\omega^2}\varepsilon\right)^2}, \tag{85}$$

which has the same form as (54), derived using Wildt's method.

An alternative (and equivalent) form to (85) (for σ_s) is given by Eq. (42) from Ref. 33,

$$\sigma_s = \frac{4\pi R_0^2\left(1+\varepsilon^2\right)^{-1}}{\left(K/\omega^2 - 1\right)^2 + \left(2\beta/\omega\right)^2}, \tag{86}$$

where[68]

$$K = \omega_0^2 + \frac{\varepsilon^2}{1+\varepsilon^2}\omega^2, \tag{87}$$

and

$$\beta = \beta_0 + \frac{\varepsilon}{1+\varepsilon^2}\frac{\omega}{2}. \tag{88}$$

The extinction cross-section obtained using the same approach is

$$\sigma_e = \sigma_s\frac{2\beta_0/\omega}{\varepsilon}\left(1 + \frac{\omega}{2\beta_0}\varepsilon + \varepsilon^2\right). \tag{89}$$

An alternative (and equivalent) form to (89) (for σ_e) is given by Eq. (64) from Ref. 33, which is identical to (82) above, demonstrating that (89) is consistent with Prosperetti's derivation.

The damping factor β_0 in (88) includes all forms of damping other than acoustic radiation. Thus, considering viscous and thermal damping it would be

$$\beta_0 = \beta_{\rm th} + \beta_{\rm vis}, \tag{90}$$

where

$$\beta_{\rm vis} = \frac{2\eta_{\rm S}}{\rho_{\rm liq} R_0^2}, \tag{91}$$

$$\beta_{\rm th} = \frac{3P_{\rm gas}}{2\rho_{\rm liq} R_0^2 \omega} \operatorname{Im} \Gamma \tag{92}$$

and Γ is given by (16).

The parameter ω_0, which through $\operatorname{Re}\Gamma(\omega)$ is a function of frequency, is given (including surface tension explicitly) by

$$\omega_0^2 = 3\operatorname{Re}\Gamma \frac{P_{\rm gas}}{\rho_{\rm liq} R_0^2} - 2\frac{\tau}{\rho_{\rm liq} R_0^3}. \tag{93}$$

In situations for which ω_0 is independent of frequency, such as for bubbles that are either thermally large or thermally small, and if β_0 is assumed to be constant, the resonance frequency is given by

$$\frac{\omega_0^2}{\omega_{\rm res}^2} = 1 - \frac{2\beta_0^2}{\omega_0^2} - \frac{\varepsilon_0^2}{2}, \tag{94}$$

where ε_0 is the dimensionless bubble radius, defined as

$$\varepsilon_0 \equiv \varepsilon(\omega_0) = \omega_0 R_0 / c \tag{95}$$

and not to be confused with the dimensionless frequency of (34).

D. Non-linear thread

The "non-linear" thread encompasses results from Refs. 5, 6, 90, and 91. The models discussed so far assume steady state spherical bubbles in the long wavelength limit ($\omega R_0/c \ll 1$), where the bubbles undergo sufficiently small amplitude pulsations to warrant linear descriptions. The basic definitions (such that in the long wavelength limit, the scattering cross-section equals the ratio of the time averaged radiated power to the incident plane wave intensity) also hold in other circumstances. One such is the time during ring-up of bubble pulsations prior to reaching steady state, for which scattering cross-section may be defined by averaging over single pulsation cycles between successive times of zero displacement,[92] becoming undefined during ringdown after the cessation of ensonification.[93] Another circumstance for which the cross-section has received particular attention is when the bubble pulsation amplitude becomes sufficiently great to generate nonlinear effects, such as the generation of higher harmonics or combination frequencies. This is in part because of the use to which such signals (which could only very rarely be generated by linear processes) can be used to distinguish between scattering from bubbles and scattering by other bodies (such as the seabed[94–96] or biological tissue[97,98]) that generate only linear scattering, or much lower levels of nonlinearity than do bubbles.[74] A typical source of nonlinearity is the amplitude dependence of the bubble stiffness.[74] Small amplitude perturbation expansions to second order have been undertaken to calculate the steady state amplitude-dependent scattering of sound at, say, the second harmonic of the ensonifying field.[99] However, emissions involving higher harmonics, or generated by driving fields or bubble responses that are too great or too brief to be modeled by perturbation expansions, generally appear in the literature as numerical solutions to the appropriate equation of motion,[100,101] or are reported as empirical observations.[97,102,103] Relating either result to some nonlinear cross-section requires further consideration, as follows.

There are several options for defining the acoustic cross-section when bubbles pulsate nonlinearly by expanding the above definition in different ways. The nonlinear cross-section can be based on the ratio of the total power scattered by a single bubble at all frequencies to the intensity of the incident plane wave, which can locally peak when that plane wave is at a frequency that is some harmonic of the bubble resonance. Figure 8 (from Ref. 104) demonstrates the results of such an approach where the nonlinearity clearly increases in the expected way with increasing drive amplitude.

However, the most popular method has been to define a cross-section for each harmonic as the power scattered within the bandwidth of the harmonic in question divided by the intensity of the incident plane wave with which the bubble is ensonified. This appeals directly to users who are characterizing how much enhancement bubbles give to scattering at a particular harmonic or combination frequency [e.g., when second-harmonic scattering from ultrasonic contrast agents (UCAs) is used to monitor blood flow]. This approach is at odds with the usual power scaling property of the linear cross-section that follows if the sources are incoherent, so

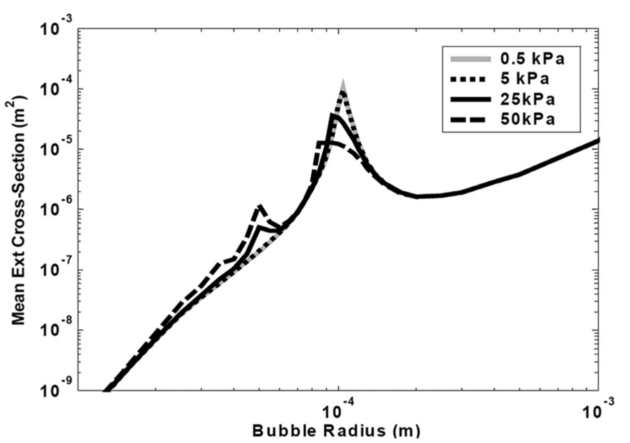

FIG. 8. Extinction cross-section for a single bubble, as a function of bubble radius, for ensonification by a 1 ms duration sinusoidal pulse of 33 kHz center frequency and zero to peak sound pressure in the range 0.5 kPa to 50 kPa. The cross-section calculated by the formulation of Ref. 93 varies over time, and the figure plots its mean value. Although the 0.5 kPa and 5 kPa lines differ (particularly close to the fundamental resonance at bubble radius 10^{-4} m), they are barely distinguishable on this scale. (from Ref. 104).

that when bubbles in a cloud scatter, the entirety of the scattered energy is found through simple addition of the cross-sections of the individual bubbles. While this property is still maintained for a given harmonic when using the above definition, the cross-sections can no longer be used to describe all the energy scattered from the cloud. Even for a single bubble, a summation of its harmonic cross-sections does not describe the whole field scattered by that bubble, because there is a fixed phase relationship between, say, the scattering at the fundamental and the scattering at the second harmonic from an individual bubble.

Despite this contradiction, the latter version of the cross-section has proved by far the most popular, probably because it appeals to the scenario of a single-frequency input generating a range of harmonic emissions from a bubble cloud (although in practice the requirement for range resolution means that the incident pulses are short enough to have significant bandwidth and to generate bubble pulsation that is dominated by ring-up and ringdown, rather than steady state). Furthermore, if the bubble population is close to monodisperse, as is often assumed with UCAs, then the measured power in each harmonic can readily be converted into an empirical cross-section for that harmonic.[105] The disadvantage of such an empirical approach is that these empirical cross-sections must vary with the amplitude of the ensonifying field and so cannot readily be transferred from one situation to another, or used as the basis for generically applicable formulations.

The drive to provide analytical cross-sections that would be generically applicable came originally from those using nonlinear scattering to detect the presence of bubbles[99,106] (since other entities in the liquid would only generate nonlinear scattering to a much smaller extent).[107–109] Perturbation expansions of a nonlinear equation of bubble dynamics were undertaken for an assumed sinusoidal driving pressure. Steady state solutions were found for the bubble wall displacement. The radius R can be assumed to take the form $R = R_0\,(1+\chi)$ with

$$\chi = \chi_0 + A_1\cos(\omega t + \varphi_{c1}) + B_1\sin(\omega t + \varphi_{s1}) + A_2\cos(2\omega t + \varphi_{c2}) + B_2\sin(2\omega t + \varphi_{s2}) \ll 1 \quad (96)$$

to examine the second harmonic emission. Sometimes the phase factors ϕ and the dc perturbation χ_0 were ignored (exponential notation is inappropriate in a description of nonlinear behavior). In other circumstances two frequencies (ω_1 and ω_2) were incident on the bubble to allow its detection through scattering of the difference frequency,[81,110,111] the relevant expansion being:

$$\begin{aligned}\chi = \chi_0 &+ A_1\cos(\omega_1 t + \varphi_{c1}) + B_1\sin(\omega_1 t + \varphi_{s1}) + A_2\cos(2\omega_1 t + \varphi_{c2}) + B_2\sin(2\omega_1 t + \varphi_{s2})\\ &+ \Psi_1\cos(\omega_2 t + \vartheta_{c1}) + \Xi_1\sin(\omega_2 t + \vartheta_{s1}) + \Psi_2\cos(2\omega_2 t + \vartheta_{c2}) + \Xi_2\sin(2\omega_2 t + \vartheta_{s2})\\ &+ \Psi_s\cos[(\omega_1+\omega_2)t + \vartheta_{cs}] + \Xi_s\sin[(\omega_1+\omega_2)t + \vartheta_{ss}]\\ &+ \Psi_d\cos[(\omega_1-\omega_2)t + \vartheta_{cd}] + \Xi_d\sin[(\omega_1-\omega_2)t + \vartheta_{sd}] \ll 1. \end{aligned} \quad (97)$$

The power scattered at each harmonic in the steady state would then be related to a cross-section. However, such expansions need to be treated with care. For example, just because the frequencies of interest in the two examples quoted above are generated by a quadratic nonlinearity, such an expansion will not capture all the energies up to second order, since higher terms will also generate energy at lower frequencies (for example, the fourth power contributes to both dc and quadratic terms through the identity $8\cos^4\omega t \equiv 3 + 4\cos 2\omega t + \cos 4\omega t$). Such an expansion approach needs a suitable nonlinear equation describing the wall response of the bubble. In the former Soviet Union, nonlinear expansions of the bubble volume, with an *ad hoc* approach to including all damping, were used,[106] while in the English language literature[99,107] radial expansions of the Rayleigh-Plesset equation were favored, which explicitly formulated the viscous losses, but no other dissipation. Reconciliation of the two approaches has been explored.[112–114]

Yang and Church[5] undertook a radial expansion of a Keller-Miksis type equation, including wall effects, for application to some ultrasonic contrast agents. This provided the lynchpin in a series of important papers expanding on this approach, as detailed below.

1. Church 1995

Church[6] derived a generalization of the Rayleigh-Plesset equation that takes into account the effect of an elastic solid layer separating a gas bubble from a surrounding viscous liquid medium. The expression derived for the scattering cross-section, obtained by linearizing the complete non-linear model, simplifies for the case of a free bubble to

$$\sigma_s = \frac{4\pi R_0^2}{\left(\omega_0^2/\omega^2 - 1\right)^2 + \left(2\beta_{vis}/\omega\right)^2}, \quad (98)$$

where ω_0 is given by (93) and β_{vis} is the viscous damping factor of (71). Church's novel derivation of this standard result laid the foundations for important later work, as described below.

2. Khismatullin 2004

Khismatullin[90] derived an expression for the scattering cross-section of a microbubble encased in a solid shell. Setting the shell thickness to zero, this expression simplifies to

$$\sigma_s = \frac{4\pi R_0^2}{\left(\frac{\omega_0^2}{\omega^2} - 1 + \frac{\varepsilon^2}{1+\varepsilon^2}\right)^2 + \left(2\frac{\beta_{vis}}{\omega} + \frac{\varepsilon}{1+\varepsilon^2}\right)^2}, \quad (99)$$

which generalizes Church's result to the case of a compressible liquid, and is consistent with (86). Khismatullin's derivation for the scattering cross-section is the first example known to the present authors of one whose starting point is the differential equation describing the motion of the bubble wall, and that results in the correct frequency dependence of the radiation damping term. At the time of writing Ref. 33, the authors were unaware of Khismatullin's work, which was consequently (and regrettably) omitted from the review element of that paper. The resonance frequency quoted by Khismatullin is

$$\omega_{res}^2 = \omega_0^2 - 2\beta_{vis}^2. \quad (100)$$

3. Yang and Church 2005

Expanding on the approach used in Ref. 6 to incorporate viscoelastic bubble wall properties in a Rayleigh-Plesset equation, Yang and Church[5] added such properties to a Keller-Miksis type equation, with viscous, acoustic radiation and thermal damping, including corrections to the bulk modulus due to surface tension and elastic forces. By linearizing a fully non-linear equation of motion, and supplementing the result with heuristic corrections for thermal and radiation damping, they obtained the most comprehensive linear model of bubble scattering known to the authors. Although Ref. 5 also incorporates terms to show the effect of tissue, these are set to zero in the present review to allow ready comparison with the other formulations. Yang and Church derived linearized differential equations, with the same form as (62) with

$$F = -\frac{A}{\rho_{liq} R_0}\left(1 + \frac{4\eta_S}{c\rho_{liq} R_0}\right)^{-1} \quad (101)$$

for the forcing term and, neglecting rigidity (and multiplying the radiation stiffness term from Eq. (24) of Ref. 5 by a factor ε^2 for consistency with Ref. 68)

$$\frac{K}{\omega_M^2} = \gamma^{-1}\left(1 + \frac{4\eta_S}{c\rho_{liq} R_0}\right)^{-1}\left[\mathrm{Re}\,\Gamma + \frac{2\tau}{3R_0 P_{liq}}(3\mathrm{Re}\,\Gamma - 1) + \frac{\rho_{liq} c^2}{3P_{liq}}\frac{\varepsilon^4}{1+\varepsilon^2}\right] \quad (102)$$

for the stiffness. Neglecting all contributions to damping other than viscosity, thermal diffusion and acoustic radiation, the damping factor simplifies to

$$\beta = \beta_{vis} + \beta_{th} + \frac{\varepsilon}{1+\varepsilon^2}\frac{\omega}{2}\left(1 + \frac{4\eta_S}{c\rho_{liq} R_0}\right)^{-1}, \quad (103)$$

where

$$\beta_{vis} = \frac{2\eta_S}{\rho_{liq} R_0^2}\left(1 + \frac{4\eta_S}{c\rho_{liq} R_0}\right)^{-1} \quad (104)$$

and (dividing the right hand side of Eq. (23b) of Ref. 5 by $\varepsilon \mathrm{Re}\,\Gamma/\mathrm{Im}\Gamma$ for consistency with Ref. 115)

$$\beta_{th} = \frac{3P_{gas}\mathrm{Im}\Gamma}{2\omega\rho_{liq} R_0^2}\left(1 + \frac{4\eta_S}{c\rho_{liq} R_0}\right)^{-1}. \quad (105)$$

The factor $(1 + 4\eta_S/c\rho_{liq} R_0)^{-1}$ in the damping, stiffness and forcing terms accounts for the possibility of the bubble not being large (in the sense that, in Sec. II D 3 only, the requirement for $4\pi\, l_{vis}^2 \ll l_{ac} R_0$ is lifted) by means of a correction to the added mass of the liquid medium.

4. Doinikov and Dayton 2006

The main thrust of Ref. 91, an analysis of the dynamics of an encapsulated gas bubble, is outside the present scope. For the special case of a free bubble, Doinikov and Dayton derived the following expression [their Eq. (57)] for the resonance frequency

$$\omega_{res}^2 = \frac{\omega_0^2}{3\varepsilon_0^2}\left\{\left[1 + 8\frac{\beta_{vis}}{\omega_0}\varepsilon_0 + 2\varepsilon_0^2\left(3 + 2\frac{\beta_{vis}^2}{\omega_0^2}\right)\right]^{1/2} - \left(1 + 4\frac{\beta_{vis}}{\omega_0}\varepsilon_0\right)\right\}. \quad (106)$$

The derivation of (106) omits terms of order ε^2 and higher, which means that such terms (order ε_0^4 terms in the curly parentheses) may be discarded from this equation without loss of accuracy. An alternative derivation that is accurate up to and including $O(\varepsilon_0^2)$ terms is presented in Sec. III C 2.

E. Which scattering or extinction cross-section?

Up to this point, this review provides a historical description of the increasing understanding with time of the physical processes involved with scattering and extinction from a single bubble undergoing spherically symmetric pulsations, and the increasing sophistication of the mathematical models used to describe the corresponding cross-sections. The purpose of this section is to offer advice on which equations are applicable if either the liquid's viscosity (Sec. II E 1) or its compressibility (Sec. II E 2) is negligible, and to describe the challenges remaining when neither is (Sec. II E 3).

1. Negligible liquid viscosity

If the liquid viscosity is negligible ($\beta_{vis} \ll \beta_{th} + \varepsilon_0\, \omega_0/2$), the damping is limited to thermal conduction across the bubble wall and acoustic radiation, in which case (85) [or (86)] and (89) apply for the scattering and extinction cross-sections, respectively, with β_0 equal to the thermal damping factor β_{th} from (92) and ω_0 calculated using (93). The result, expressed in a form designed to highlight and clarify the potential for confusion referred to in the Introduction, can be written (using $\beta_{vis} = 0$ here)

$$\sigma_s = \frac{4\pi R_0^2}{\left(\omega_0^2/\omega^2 - 1 - 2\varepsilon\beta_{th}/\omega\right)^2 + \delta_{Weston}{}^2}, \tag{107}$$

where δ_{Weston} is obtained by generalizing (42) while dropping the viscosity term, as follows:

$$\delta_{Weston} = 2\frac{\beta_{th}}{\omega} + \frac{\omega_0^2}{\omega^2}\varepsilon. \tag{108}$$

The terms ω_0, given by (93), and β_{th}, by (92), are both functions of frequency through the (complex) polytropic index Γ from (16).

The extinction cross-section is related to σ_s according to (89):

$$\frac{\sigma_s}{\sigma_e} = \frac{\varepsilon}{\delta_{Medwin} + 2\varepsilon^2\beta_{th}/\omega}, \tag{109}$$

where

$$\delta_{Medwin} \equiv 2\frac{\beta_{th}}{\omega} + \varepsilon. \tag{110}$$

These equations hold if the logarithmic decrement is small, which is equivalent to requiring δ_{Medwin} to be small. The method of Chapman and Plesset[67] is applicable to any value of the logarithmic decrement, large or small.

The ratio σ_s/σ_e, calculated using (109), is plotted in Fig. 9 (solid curves) for two different values of β_{th}/ω_0. Although both δ_{Weston} and δ_{Medwin} are correct when used with their corresponding equations, (107) and (109), it is essential that δ_{Medwin} and *not* δ_{Weston} be used in (109). This point is illustrated with a second calculation (dashed curves), obtained by replacing δ_{Medwin} with δ_{Weston} in the right hand side of (109). This second calculation underestimates the value of σ_e by a factor of $(\omega/\omega_{res})^2$ at frequencies above resonance. (Below resonance a smaller error is made, and in the opposite direction.) In order to compensate for this error, and citing a personal communication by Anderson, Ref. 38 justifies a multiplication by $(\omega_{res}/\omega)^2$ compared with AH32 (adopting the shorthand *AHn* to denote Eq. *n* from Ref. 22), with the accompanying explanation "There is no special off-resonance behavior of the radiation damping term [ε], and hence the $(\omega_0/\omega)^2$ term should be omitted." This arbitrary correction, the need for which is created by use of the ambiguous damping coefficient (δ), is avoided by making use of (85) and (89), with (4) for σ_a, which express the cross-sections instead in terms of the unambiguous damping factor (β). Doing so avoids the confusion caused by mixing expressions that imply use of δ_{Medwin} on the one hand (AH32, AH54, and AH56) and δ_{Weston} on the other (AH43 and AH55).

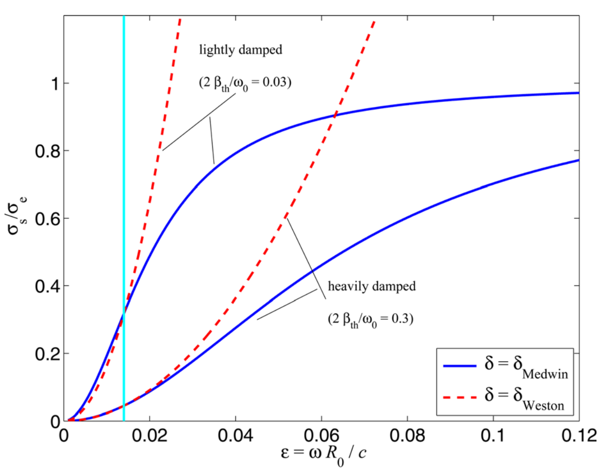

FIG. 9. (Color online) Ratio of scattering to extinction cross-sections versus dimensionless frequency ε [(109) solid lines]. Also shown (dashed lines) is the result of an incorrect calculation obtained by replacing δ_{Medwin} with δ_{Weston} in the right hand side of (109). Values used for the parameter $2\ \beta_{th}/\omega_0$ are as marked, with $\varepsilon_0 = 0.014$. Errors grow rapidly above the resonance frequency (vertical dotted line).

Comparing (57) with (33), it can be seen that the parameter b_{th} introduced by Wildt is

$$b_{th} = \frac{3P_{gas}\mathrm{Im}\Gamma}{\rho_{liq}R_0^2\omega_{res}^2}. \tag{111}$$

2. Negligible liquid compressibility

Neglecting viscosity limits the applicability of the resulting equations to bubbles that are sufficiently large for thermal or acoustic radiation losses to dominate. An alternative simplification is to neglect the liquid compressibility instead, requiring that the bubble be sufficiently small for acoustic radiation damping to be neglected ($\varepsilon \ll 2\ \beta_0/\omega$), and resulting in

$$\sigma_s = \frac{4\pi R_0^2}{\left(\omega_0^2/\omega^2 - 1\right)^2 + 4\beta_0^2/\omega^2}, \tag{112}$$

and

$$\sigma_e = \sigma_s \frac{2\beta_0/\omega}{\varepsilon}, \tag{113}$$

with β_0 given by (90) to (92), with (91) for the viscous damping factor β_{vis}. The singularity in (113) as $\varepsilon \to 0$ reflects the fact that no sound energy can be radiated, so all of the incident acoustic power is absorbed (either by thermal conduction across the bubble wall or by viscous processes in the surrounding liquid), and the ratio σ_e/σ_s tends to infinity.

Comparing (71) with (33), it can be seen that the parameter b_{vis} introduced by Wildt is

$$b_{vis} = \frac{4\eta_S}{R_0}. \tag{114}$$

These equations hold if the logarithmic decrement is small, which close to resonance is equivalent to requiring $2\ \beta_0/\omega_0$ to be small. Here, as in Sec. II E 1 above, the method of Chapman and Plesset[67] is applicable to both small and large values of the logarithmic decrement.

3. Viscous damping in a compressible liquid

The introduction of viscosity in a compressible medium brings with it three complications. The first is that both incident and scattered waves decay exponentially in a viscous medium. The assumption made so far in this paper that A and B are constants needs to be revisited. It might be justified, for example, by arranging for any measurements to be made at a sufficiently short distance from the bubble that any exponential decay may be neglected.

The second complication is the need identified by Yang and Church[5] for a viscous correction to the added mass of the liquid medium, modifying the equations for stiffness, (102), and damping factors, (103)–(105). The correction term accounts for the effect of shear viscosity (bulk viscosity is not considered).

The third is of a more fundamental nature. As the bubble pulsates, the liquid molecules in contact with it must rearrange themselves to accommodate the contractions and dilations. If the liquid is incompressible, the resulting motion is translational in nature, with layers of molecules sliding over one another to permit an increasing number of liquid molecules to come into contact with the bubble gas as it expands, and a decreasing number as it contracts. The viscous forces that act in these circumstances are controlled by shear viscosity alone.[116] Elements of a compressible liquid, on the other hand, have the freedom to accommodate the pulsating bubble by occupying more or less volume themselves (for example, by permitting more or less empty space between adjacent molecules of that liquid). Under these conditions, the viscous forces are affected also by additional contributions due to dilatational viscosity η_D,[73,117,118] also known as "second viscosity."[119] The related bulk viscosity η_B is[120]

$$\eta_B = \frac{2}{3}\eta_S + \eta_D. \tag{115}$$

The assumption of incompressible conditions allows both bulk and dilatational viscosities to be dropped from early work leading to the Rayleigh-Plesset equation.[66,114,121] Extension into other regimes such as the compressible liquids of the Herring-Keller equation[122–126] did not result in a re-introduction of bulk or dilatational viscosity. For example, in Ref. 17 it is stated that "Liquid compressibility effects do not substantially alter the effects of the… viscoelasticity of the surrounding medium." Doinikov and Dayton[91] pointed out that the dilatation term is identically zero for an incompressible medium, but did not provide a similar justification for omitting a term representing bulk or dilatational viscosity from their Eq. (31), describing the motion of a pulsating bubble in a compressible liquid. In fact, the radiation and viscous damping enter Eq. (31) of Ref. 91 by different routes, one (radiation) via the kinetic energy of a slightly compressible liquid, and the other (viscosity) via the dissipative function of a viscous, incompressible liquid. According to A. A. Doinikov (personal communication, November 2010), the introduction of compressibility in the dissipative function would result in the appearance of a bulk viscosity term in Eq. (31) of Ref. 91.

To understand the possible effects of combining viscosity with liquid compressibility, it is useful to consider the origin of the term that expresses viscous dissipation in the Rayleigh-Plesset equation [$4\eta_S\dot{R}/(\rho_{liq}R)$ if written in terms of radius[114]], readily derived from the Navier-Stokes equation in the form[118]

$$\rho_{liq}\frac{\partial \mathbf{v}}{\partial t} + \rho_{liq}(\mathbf{v}\cdot\nabla)\mathbf{v} - \sum \mathbf{F}_{ext} + \nabla P_{body} = (\eta_S + \eta_D)\nabla(\nabla\cdot\mathbf{v}) + \eta_S\nabla^2\mathbf{v}, \tag{116}$$

where P_{body} is the pressure on a boundary contained within the body of the liquid, and where the vector sum of all body forces $\sum\mathbf{F}_{ext}$ is usually assumed to be zero in bubble acoustics. If the medium is incompressible, the divergence term $\nabla\cdot\mathbf{v}$ is zero. If, further, the Navier-Stokes equation is used to describe a spherically symmetrical scattering problem, as in the present situation, $\nabla^2\mathbf{v}$ is also zero,[1] so the right hand side of (116) vanishes for any non-infinite magnitude of η_S, η_D, and η_B. In a compressible medium, for which the divergence $\nabla\cdot\mathbf{v}$ is no longer neglected, the question then moves from one of the dilatational viscosity being multiplied by a zero term (specifically $\nabla\cdot\mathbf{v}$), to the possibility that the dilatational viscosity will affect the bubble dynamics if the sum $\eta_S+\eta_D$ does not equal zero. Finite shear viscosity does not manifest itself in the dynamics of spherical bubbles in incompressible liquids through the Navier-Stokes equation. Instead, it applies a correction to the normal stress, proportional to the principle rate of strain in the radial direction, the constant of proportionality being $2\eta_S$, such that the pressure applied by the liquid on the bubble wall, denoted P_{wall}, becomes (see p. 304 of Ref. 1, and Ref. 114):

$$P_{wall} = P_{body} + \frac{4\eta_S}{R}\frac{dR}{dt}, \tag{117}$$

and it is by this boundary condition route that shear viscosity enters into the Rayleigh-Plesset equation.[1,114] However, even in spherically symmetric conditions, if the assumption of liquid incompressibility does not hold, then the simple relationship given by (117) does not hold either, and the direct translation of such terms, which neglect the dilatational viscosity, into Herring-Keller type equations needs to be critically assessed. Both Devin[85] and Chapman and Plesset[67] explicitly stated that their derivations for viscous damping neglect any effect of liquid compressibility. Devin derived his well-known viscous damping factors using (117).

The first serious attempt known to the authors to evaluate the viscous damping of a gas enclosure in a compressible medium appears in the pioneering work of Love,[20] who included the bulk viscosity of fish flesh in his model for the scattering cross-section of a swimbladder, resulting in (50). However, Love's result, originally intended to apply to scattering from a fish swimbladder, is rarely, if ever, used for gas bubbles in water. Instead, Devin's viscous damping term, (71), is typically added heuristically to treatments of compressibility in an inviscid medium.[5,91]

The question of consistency between the two models of viscous damping due to Devin[85] for an incompressible medium and that of Love[20] for a compressible one is now addressed. Comparing (71) with (50) reveals an apparent factor 2/3 difference between the coefficient of the η_S term in the damping models of Love and Devin, giving the impression of an inconsistency. However, before drawing such a conclusion, it is important to consider the relationship between the shear (η_S), bulk (η_B) and dilatational (η_D) viscosity coefficients of a viscous fluid (115), subject to the condition[127]

$$\eta_B \geq 0. \tag{118}$$

It has been pointed out to us (R. H. Love, personal communication, November 2010) that substituting (115) in (50) gives

$$\beta_{vis} = \frac{2\eta_S + \eta_D}{\rho_{liq} R_0^2}, \tag{119}$$

which has the same η_S coefficient as (71), implying that had Devin considered his dilatational viscosity to be zero, the apparent contradiction between (71) and (50) would be resolved. However, the present authors do not perceive this assumption in Devin's derivation.

Stokes[117] argued that for some fluids (e.g., monatomic gases[128]) the bulk viscosity might be zero or negligible (referred to henceforth as the "Stokes condition"), in which case it follows from (115) that

$$\eta_D = -\frac{2}{3}\eta_S, \tag{120}$$

which can only be zero if the shear viscosity vanishes. It is known that (120) does not hold for water,[119,128,129] and that the bulk viscosity makes an important contribution to absorption of ultrasound in water[130] and in other liquids.[131,132] Liebermann[130] reports that for water, the ratio η_D/η_S is approximately equal to 2.2, a value confirmed by modern measurements.[133]

For the theories of Devin and Love to be consistent, Love's version must simplify to Devin's in the limit of an incompressible liquid medium. As they differ only by a term proportional to η_D, consistency requires η_D to vanish in the incompressible limit.[134] Regardless of whether this might hypothetically be the case, the dilatational viscosity of water (a compressible medium),[119,128,129] or of any viscous liquid satisfying the Stokes condition[117,118] is not zero, so in practice (71) and (50) result in different values of β_{vis}. Therefore, even if the two theories are somehow consistent, they cannot *both* provide a correct description of viscous damping of a gas bubble either in water or in any viscous liquid satisfying the Stokes condition.

III. RESONANCE FREQUENCY REVISITED

A. Introduction

Ever since Minnaert's pioneering work,[69] the natural frequency of a single spherical bubble, and the related resonance frequency, have been among the most widely used concepts in the development of an increasingly sophisticated understanding of linear bubble acoustics. Advances are described by Refs. 1, 5, 6, 19, 20, 22, 29, 33, 66–68, 73, 77, 85, 86, 88, 90, 91, 115, and 135–137. Despite this central role, examination of the expressions for ω_{res} in Sec. II reveals that there is no single widely accepted expression for the resonance frequency. For example, (100) (see also Ref. 73) includes a viscosity dependent correction to the square of the resonance frequency that reduces it by $2\beta_{vis}^2$ from its nominal value of ω_0^2, whereas (94), originally from Ref. 33, features a correction of the same magnitude but in the opposite direction. Similarly, Houghton's viscosity correction[136] has the same sign as that of Khismatullin, but only half its magnitude.

The present authors are not aware of any previously given explanation for the above-mentioned discrepancies. This section will show that the differences are entirely due to the use in different publications of different definitions of resonance frequency. Given the importance of the use of a consistent set of definitions and assumptions in any analysis, this section considers the options. Several definitions are possible, although the choice made by individual authors is rarely elaborated on. In this section, four alternative interpretations of the term "resonance frequency" are examined, all consistent with the 1994 American National Standards Institute (ANSI) definition.[138] These are preceded by Sec. III B on the natural frequency of unforced oscillations. The section ends with a summary (Sec. III H) of which equations are appropriate in different circumstances.

B. Natural frequency of unforced oscillations

The vibration frequency of a system undergoing unforced oscillations is known as its "natural frequency." In the following, this quantity is denoted ω_{nat}.

1. Constant stiffness and damping factor

Damping is modeled by introducing a force proportional to $\dot{R}$ in the differential equation for $R(t)$:

$$\ddot{R} + 2\beta\dot{R} + K(R - R_0) = 0, \tag{121}$$

where β is the damping constant. The solution is

$$R - R_0 = a\exp(\mathrm{i}\omega_{nat}t)\exp(-\beta t), \tag{122}$$

where (if K is also constant)

$$\omega_{nat} = \sqrt{K - \beta^2} \tag{123}$$

and the complex constant a is determined by the boundary conditions.

2. Application to a bubble (frequency dependent stiffness and damping factor)

If K and β are functions of frequency, as is in general the case for a pulsating bubble, the natural frequency satisfies the equation

$$\omega_{nat}^2 = K(\omega_{nat}) - \beta(\omega_{nat})^2, \tag{124}$$

which (if β_0, ω_0 are both constant) can be written as the following quadratic in ω^2

$$\frac{5\varepsilon_0^2}{4\omega_0^2}\left[1+O\left(\varepsilon_0^2\right)\right]\omega_{\mathrm{nat}}^4+\left[1+\frac{\varepsilon_0\beta_0}{\omega_0}-2\varepsilon_0^2\left(1-\frac{\beta_0^2}{\omega_0^2}\right)\right]\omega_{\mathrm{nat}}^2 - \left(\omega_0^2-\beta_0^2\right)=0, \tag{125}$$

where ω_0, β_0 and ε_0 are given by (55) (with Γ equal to a constant such as 1 or γ), (90) and (95), respectively. Noting that in the limit of $\varepsilon_0 \to 0$, $\omega_{\mathrm{nat}}{}^2$ is equal to $\omega_0{}^2 - \beta_0{}^2$, and retaining all terms up to $O(\varepsilon_0{}^2)$ in (125), the natural frequency is given by

$$\omega_{\mathrm{nat}}^2=\left(\omega_0^2-\beta_0^2\right)\left[1-\frac{\beta_0}{\omega_0}\varepsilon_0+\frac{1}{4}\left(3+\frac{\beta_0^2}{\omega_0^2}\right)\varepsilon_0^2+O\left(\varepsilon_0^3\right)\right], \tag{126}$$

which in the limit of small ε_0 simplifies to the expression derived by Houghton[136] for the "frequency of radial pulsation." It is also consistent with (47), the expression derived by Chapman and Plesset for the natural frequency. The correction terms proportional to ε_0 and $\varepsilon_0{}^2$ are due to the frequency dependence of the stiffness and damping terms in (124), through (87) and (88).

C. Frequency of displacement resonance

The resonance frequency of a system in forced oscillation is the frequency at which "any change in the frequency of excitation results in a decrease in the response of the system."[138] In the present section, this is interpreted as the frequency of maximum displacement amplitude for fixed forcing pressure amplitude, and denoted ω_R.

The particular solution to (62) is

$$R-R_0=\frac{F\mathrm{e}^{\mathrm{i}\omega t}}{K+2\mathrm{i}\beta\omega-\omega^2}. \tag{127}$$

The general solution is the sum of the particular solution and the unforced result from (122).

1. Constant stiffness and damping factor

Differentiating the expression for the squared magnitude of the displacement amplitude

$$|R-R_0|^2=\frac{F^2}{\left(K-\omega^2\right)^2+4\beta^2\omega^2}, \tag{128}$$

and setting the result to zero gives the following simple expression for the displacement resonance frequency

$$\omega_R^2=K-2\beta^2. \tag{129}$$

This is the resonance frequency that maximizes the bubble wall displacement for fixed forcing pressure amplitude

2. Application to a bubble (frequency dependent stiffness and damping factor)

Now consider $K(\omega)$ and $\beta(\omega)$ as variables, in which case the condition for a displacement resonance becomes

$$\frac{1}{2}\frac{\mathrm{d}}{\mathrm{d}\omega}\frac{F^2}{|R-R_0|^2}=\left(\omega^2-K\right)\left(2\omega-\frac{\mathrm{d}K}{\mathrm{d}\omega}\right) +4\omega\beta\left(\beta+\omega\frac{\mathrm{d}\beta}{\mathrm{d}\omega}\right)=0. \tag{130}$$

Rearranging (130), retaining terms to $O(\varepsilon^2)$ and using the approximations

$$K\approx\omega_0^2+\left(\varepsilon_0/\omega_0\right)^2\omega^4 \tag{131}$$

$$\beta\approx\beta_0+\left(\varepsilon_0/2\omega_0\right)\omega^2, \tag{132}$$

such that

$$\mathrm{d}K/\mathrm{d}\omega\approx 4\left(\varepsilon_0/\omega_0\right)^2\omega^3 \tag{133}$$

$$\mathrm{d}\beta/\mathrm{d}\omega\approx\left(\varepsilon_0/\omega_0\right)\omega, \tag{134}$$

it follows that

$$\left(1+4\frac{\varepsilon_0\beta_0}{\omega_0}\right)\omega^2=\omega_0^2-2\beta_0^2-\varepsilon_0^2\left(2-\frac{3\omega^2}{2\omega_0^2}\right)\omega^2+O\left(\varepsilon_0^3\omega^2\right). \tag{135}$$

Substituting

$$\omega^2=\omega_0^2-2\beta_0^2+O\left(\omega^2\varepsilon_0\right) \tag{136}$$

in the right hand side of (135) and using ω_R to denote the solution to the resulting equation for ω, it follows that

$$\frac{\omega_R^2}{\omega_0^2}=\left(1-2\frac{\beta_0^2}{\omega_0^2}\right)\left[1-4\frac{\beta_0}{\omega_0}\varepsilon_0-\frac{1}{2}\left(1-26\frac{\beta_0^2}{\omega_0^2}\right)\varepsilon_0^2+O\left(\varepsilon_0^3\right)\right]. \tag{137}$$

To first order in ε_0 this equation is consistent with (106). It is not consistent to second order because not all order ε^2 terms are retained in the derivation of Ref. 91. To zeroth order in ε_0 it is also consistent with Ref. 73 [specifically, with (100)]. This observation explains the difference between (100) and Houghton's result, which was for the natural frequency (Sec. III B 2).

D. Frequency of velocity resonance

In the present section, the ANSI definition of "resonance frequency"[138] is interpreted as the frequency of maximum amplitude of the bubble wall velocity for fixed forcing pressure amplitude and denoted ω_u.

Differentiating (127) with respect to time and defining input impedance Z as

$$Z\equiv p_{\mathrm{i}}/(\mathrm{d}R/\mathrm{d}t), \tag{138}$$

it follows that

$$\mathrm{i}\omega\frac{Z}{\rho_{\mathrm{liq}}R_0}=\omega^2-K-2\mathrm{i}\beta\omega. \tag{139}$$

1. Constant stiffness and damping factor

The rate of change of input impedance with frequency is given by

$$\frac{\mathrm{d}|Z|^2}{\mathrm{d}\omega} = 2\rho_{\mathrm{liq}}^2 R_0^2 \omega \left(1 - \frac{K}{\omega^2}\right)\left(1 + \frac{K}{\omega^2}\right), \quad (140)$$

and the maximum radial wall velocity occurs when this quantity is zero. Thus, if K and β are both independent of frequency

$$\omega_u = \sqrt{K}. \quad (141)$$

2. Application to a bubble (frequency dependent stiffness and damping factor)

The general condition for velocity resonance is

$$\frac{\mathrm{d}}{\mathrm{d}\omega}\frac{|Z|^2}{\rho_{\mathrm{liq}}^2 R_0^2} = \frac{2}{\omega}\left[\left(\omega^2 - K\right)\left(1 + \frac{K}{\omega^2} - \frac{1}{\omega}\frac{\mathrm{d}K}{\mathrm{d}\omega}\right) + 4\beta\omega\frac{\mathrm{d}\beta}{\mathrm{d}\omega}\right] = 0, \quad (142)$$

which can be written in the form

$$\omega^4 = K^2 + \left(\omega^2 - K\right)\omega\frac{\mathrm{d}K}{\mathrm{d}\omega} - 4\beta\omega^3\frac{\mathrm{d}\beta}{\mathrm{d}\omega}, \quad (143)$$

the solution of which is the velocity resonance frequency ω_u. Using (131) to (134), the right hand side of this equation is equal to $\omega_0^4\,[1 + O(\varepsilon_0)]$. Substituting this result back into the right hand side of (143), it follows that

$$\frac{\omega_u^4}{\omega_0^4} = 1 - 4\frac{\beta_0}{\omega_0}\varepsilon_0 + O\left(\varepsilon_0^2\right). \quad (144)$$

E. Frequency of bubble pressure resonance

In the present section, the ANSI definition of "resonance frequency"[138] is interpreted as the frequency of maximum amplitude of the internal bubble pressure for fixed forcing pressure amplitude and denoted ω_p.

The complex time-varying acoustic pressure p_b (assumed to be spatially uniform inside the bubble) is related to the incident wave amplitude A according to [see Eq. (20) from Ref. 33]

$$\frac{p_\mathrm{b}}{A} = \frac{(1 + \mathrm{i}\varepsilon)\left(\omega_0^2 + 2\mathrm{i}\omega\beta_0\right)}{(1 + \mathrm{i}\varepsilon)\left(\omega_0^2 + 2\mathrm{i}\omega\beta_0\right) - \omega^2}\exp(\mathrm{i}\omega t), \quad (145)$$

and therefore

$$\left|\frac{A}{p_\mathrm{b}}\right|^2 = \frac{1 + \left(\frac{4\beta_0^2}{\omega_0^2} - 2 + \varepsilon_0^2\right)\frac{\omega^2}{\omega_0^2} + \left(1 + \varepsilon_0\frac{2\beta_0}{\omega_0}\right)^2\frac{\omega^4}{\omega_0^4}}{1 + \left(\frac{4\beta_0^2}{\omega_0^2} + \varepsilon_0^2\right)\frac{\omega^2}{\omega_0^2} + \varepsilon_0^2\frac{4\beta_0^2}{\omega_0^2}\frac{\omega^4}{\omega_0^4}}. \quad (146)$$

The condition for (bubble pressure) resonance is that the right hand side of (146) be independent of frequency, which results in the quadratic equation (neglecting terms of order $\varepsilon_0{}^3$ and higher)

$$\left[\frac{4\beta_0^2}{\omega_0^2} + \varepsilon_0^2\left(1 + 2\frac{4\beta_0^2}{\omega_0^2}\right)\right]\frac{\omega^4}{\omega_0^4} + 2\frac{\omega^2}{\omega_0^2} - 2\left(1 - 2\varepsilon_0\frac{2\beta_0}{\omega_0} + 4\varepsilon_0^2\frac{4\beta_0^2}{\omega_0^2}\right) = 0, \quad (147)$$

with solution

$$\frac{\omega_p^2}{\omega_0^2} = \frac{\sqrt{1 + 2\left[4\beta_0^2/\omega_0^2 + \left(1 + 8\beta_0^2/\omega_0^2\right)\varepsilon_0^2\right]} - 1}{4\beta_0^2/\omega_0^2 + \left(1 + 8\beta_0^2/\omega_0^2\right)\varepsilon_0^2} + O\left(\varepsilon_0^3\right). \quad (148)$$

The right hand sides of (145) and (146), and the first term of that of (148) are symmetrical to interchange of ε_0 and $2\beta_0/\omega_0$, implying that the pressure amplitude inside the bubble is insensitive to whether damping is caused by acoustic radiation or by some other mechanism.

F. Frequency of far-field pressure resonance

In the present section, the ANSI definition of "resonance frequency"[138] is interpreted as the frequency that maximizes the amplitude of the far-field pressure for fixed forcing pressure amplitude, and denoted ω_σ. Ainslie and Leighton[33] showed that this frequency is given by

$$\frac{\omega_0^2}{\omega_\sigma^2} = 1 - \frac{2\beta_0^2}{\omega_0^2} - \frac{\varepsilon_0^2}{2}. \quad (149)$$

This explains the differences compared with Houghton's result for the natural frequency (Sec. III B 2) and that of Khismatullin for the displacement resonance frequency (Sec. III C 2), respectively. Thus, all of the differences mentioned in Sec. III A are explained by the use by different authors of different *definitions* of resonance frequency.

The right hand side of (149) exhibits the same symmetry property as (145) and (146), as does the right hand side of (84), from which it is derived. Therefore, the amplitude of the far-field scattered pressure exhibits the same insensitivity to the damping mechanism as does the bubble pressure.

G. Resonance frequency for frequency-dependent stiffness

Consider a velocity resonance with $\beta\,\mathrm{d}\beta/\mathrm{d}\omega = 0$. If ε_0 is neglected, then (142) simplifies to

$$\omega^2 = \omega_0^2(\omega), \quad (150)$$

with (93) for ω_0. The solution of (150) is discussed in Sec. II A 3. For similar treatments of the resonant bubble radius at a fixed frequency, see Refs. 86 and 139.

H. "Which resonance frequency?" revisited

Equations for a bubble's natural pulsation frequency are described in Sec. III B. The natural frequency is the pulsation frequency of an unforced bubble and, if β_0 and ω_0 are independent of frequency, is given by (126). In general, however, both β_0 and ω_0 are functions of frequency, through (90) to (92) and through (93), respectively.

The selection of comparable equations for the resonance frequency is complicated by multiple possible definitions of that term, as described in Secs. III C to III G. The answer to the question "Which equation should I use?" depends on the application. If one's purpose is to evaluate any one of the acoustical cross-sections σ_s, σ_e, or σ_a, there is no need to evaluate the resonance (or natural) frequency at all. Instead it is sufficient to evaluate the parameter ω_0 at the frequency of interest using (93) and substitute the result into the appropriate equation for the cross-section (Sec. II E).

The various definitions depend on which physical property is chosen to quantify the amplitude of a bubble's pulsations for a fixed incident plane wave amplitude. The chosen resonance can then be used to determine the correct formula to use in place of the generic resonance frequency ω_{res} to apply the findings of Sec. II to the case in hand. The frequency of maximum far-field response (149) and maximum wall displacement (137) are both in use, although the choice is rarely made explicit. The consequence of choosing the frequency that maximizes the far-field pressure when a maximum wall displacement was intended (or vice versa) would be a fractional error of approximately $2\,\beta_0^2/\omega_0^2$ in the design frequency of (say) a drug delivery system.

The resonance frequencies for maximum wall velocity and maximum (internal) bubble pressure are derived in Sec. III and given by (144) and (148), respectively. All of these expressions for the resonance frequency require ω_0 to be independent of frequency, in which situation all definitions considered result in the same value for the undamped resonance frequency, equal to ω_0. The presence of damping increases some resonance frequencies and decreases others. Most of the expressions derived also require the dimensionless bubble radius ε_0 (defined, for situations in which ω_0 is a constant, as the dimensionless frequency evaluated at $\omega = \omega_0$) to be small, the non-acoustic damping factor β_0 to be independent of frequency (but not necessarily small) and the gas density to be negligible.

The effect of relaxing the requirement for constant ω_0 is considered in Sec. II A 3 (see also Sec. III G), where the effect on the velocity resonance of varying the polytropic index between its isothermal and adiabatic limits is explored. Good approximations for the resonance frequency of bubbles whose radius is between 30 nm and 300 μm, under the conditions considered in this review, are $\omega_{res} \approx \omega_1^{ad}$ (21) and $\omega_{res} \approx \omega_1^{iso}$ (28). Finally, the effect of non-zero gas density on the far-field pressure resonance with $\beta_0 = 0$ is given by (61).

IV. CONCLUSIONS

The apparent simplicity of the widely used concepts of bubble cross-sections, resonance frequency and damping is illusory. There exist multiple definitions for their component terms, some of which are contradictory, leading to a risk of large error if inconsistent combinations are used.

A. Scattering, extinction, and absorption cross-sections

Equations for the scattering and extinction cross-sections for a gas bubble in a compressible inviscid or an incompressible viscous liquid are summarized in Sec. II E. The absorption cross-section is given by (4).

As yet unresolved problems for a compressible viscous medium are described in Sec. II E 3. For example, the models of Devin[85] and Love[20] are inconsistent with one another when applied to a gas bubble in an incompressible viscous liquid, a situation for which ostensibly both seem applicable.

Use of the dimensionless damping coefficient δ results in ambiguity, partly because different researchers adopt different definitions for this quantity, and partly because these definitions are rarely made explicit. Sometimes more than one definition is used in the same publication. For this reason the equations in Sec. II E are cast in this paper either in terms of damping factor β, or with an accompanying definition for δ.

B. Natural and resonance frequencies

The natural frequency of Sec. III B is given by (123) or (126). For the resonance frequency, multiple definitions are in use, depending on which physical property is chosen to quantify the amplitude of a bubble's pulsations for a fixed incident plane wave amplitude. Each definition results in a different equation for the resonance frequency (see Sec. III for details).

ACKNOWLEDGMENTS

This work was sponsored in part by the Defence Research and Development Department of the Netherlands Ministry of Defence. Helpful correspondence with K. Baik, C. C. Church, A. A. Doinikov, R. H. Love and A. Prosperetti, and the constructive and detailed comments of two anonymous reviewers, are gratefully acknowledged.

NOMENCLATURE

A	amplitude of incident plane wave
A_1, A_2	dimensionless constants in (96) and (97)
B	amplitude of scattered spherical wave multiplied by distance from center of symmetry
B_1, B_2	dimensionless constants in (96) and (97)
b_{th}	constant in (35) (β in the notation of Wildt 1946; related to thermal damping)
b_{vis}	constant in (35) (C_1 in the notation of Wildt 1946; related to viscous damping)
c	speed of sound in liquid
C_P	specific heat capacity at constant pressure (gas)
D_P	gas diffusivity defined by (9)
D_V	gas diffusivity defined by (10)
F	coefficient of $e^{+i\omega t}$ in (62), representing the forcing term proportional to amplitude A of incident pressure wave.
$K(\omega)$	coefficient of $(R - R_0)$ in (62), representing stiffness to mass ratio
k_B	Boltzmann's constant
K_{gas}	thermal conductivity (gas)
l_{ac}	acoustic length scale (wavelength), (6)
l_{th}	thermal diffusion length, (8)

l_{vis} length scale associated with viscous forces, (7)
m_{gas} average mass of a gas molecule
p acoustic pressure
P total pressure
p_b time-varying complex acoustic pressure within the bubble gas (assumed to be spatially uniform)
P_{body} total instantaneous pressure in body of liquid medium
P_{flesh} equilibrium pressure in fish flesh
P_{gas} equilibrium pressure inside gas bubble
p_i acoustic pressure of incident plane wave
P_{liq} equilibrium pressure of liquid
p_s acoustic pressure of scattered spherical wave
P_{wall} total instantaneous pressure applied by liquid to bubble wall
Q Q factor
Q_{rad} Q factor (acoustic radiation damping)
Q_{th} Q factor (thermal damping)
Q_{vis} Q factor (viscous damping)
r distance from center of bubble
$R(t)$ instantaneous bubble radius
R_0 equilibrium bubble radius
R_D diffusion radius, defined by (13)
$R_{Laplace}$ Laplace radius, defined by (11)
t time
T absolute temperature
u_s acoustic particle velocity associated with scattered spherical wave
$\mathbf{v}$ acoustic particle velocity
$X(\omega, R_0)$ diffusion ratio, defined as the ratio of bubble radius to thermal diffusion length; see (14)
X_{ad} diffusion ratio evaluated at the adiabatic resonance frequency ω_{ad}
Z input impedance at bubble wall, defined by (138)
β damping factor, defined as half of the coefficient of dR/dt in (62)
β_0 sum of non-acoustic contributions to the damping factor β
β_{th} contribution to the damping factor β due to thermal conduction across the bubble wall
β_{vis} contribution to the damping factor β due to liquid viscosity
γ specific heat ratio (gas)
Γ complex polytropic index
δ dimensionless damping coefficient
δ_{Medwin} dimensionless damping coefficient defined as $2\ \beta/\omega$
δ_{other} contributions to δ other than those resulting from acoustic radiation, viscosity, and thermal dissipation, which are included specifically in δ_{rad}, δ_{vis}, and δ_{th}, respectively
δ_{rad} contribution to dimensionless damping coefficient due to acoustic radiation
δ_{th} contribution to dimensionless damping coefficient due to thermal diffusion in the gas bubble
δ_{vis} contribution to dimensionless damping coefficient due to liquid viscosity
δ_{Weston} dimensionless damping coefficient defined as $2\ \beta_0/\omega + \varepsilon\ \omega_0^2/\omega^2$
δ_{Wildt} dimensionless damping coefficient defined as $\mathrm{Im}(A\ R_0/B)$
ε dimensionless frequency $\omega R_0/c$
ε_0 dimensionless bubble radius $\omega_0 R_0/c$, defined by (95); this notation is only used when ω_0 is independent of frequency, making ε_0 a function of radius only
$\zeta(X)$ function of X defined by (66)
η_B bulk viscosity coefficient (liquid)
η_D dilatational viscosity coefficient (liquid)
η_S shear viscosity coefficient (liquid)
η_{th} equivalent thermal viscosity coefficient; see (80) and (81)
κ real polytropic exponent, defined by (48)
μ shear modulus of fish flesh
ν dimensionless function of bubble radius defined by (20)
ρ_{flesh} equilibrium mass density of fish flesh
ρ_{gas} equilibrium mass density of gas
ρ_{liq} equilibrium mass density of liquid
σ_a absorption cross-section
σ_e extinction cross-section
σ_s scattering cross-section
$\varphi_{c1}, \varphi_{c2}, \varphi_{s1}, \varphi_{s2}$ phase terms in expansion for change in bubble radius (96) and (97)
χ dimensionless change in bubble radius in (96) and (97)
χ_0 dimensionless constants in (96) and (97)
ω angular frequency of driving acoustic field
ω_0 frequency dependent parameter, closely related to the resonance frequency and defined by (55)
ω_1^{ad} approximation to resonance frequency obtained by a single iteration of (19), using ω_{ad} as seed
ω_1^{iso} approximation to resonance frequency obtained by a single iteration of (19), using ω_{iso} as seed
ω_2^{Mac} approximation to resonance frequency equal to the sum of the first three terms in the Maclaurin expansion obtained after two iterations of (19), using ω_{ad} as seed; see (26)
ω_{ad} adiabatic resonance frequency; see (17)
ω_{iso} isothermal resonance frequency; see (27)
ω_M Minnaert frequency multiplied by 2π; see Eq. (12)

ω_{nat} natural frequency
ω_p resonance frequency (bubble pressure)
ω_R resonance frequency (radial displacement)
ω_{res} resonance frequency (generic); the expression for this parameter depends on whether the resonance is for a maximum response in velocity, pressure, or some other parameter; this notation is used to indicate that the precise nature of the maximum is unspecified
ω_u resonance frequency (radial velocity)
ω_σ resonance frequency (far-field pressure)
Ω frequency dependent parameter, closely related to the resonance frequency and defined by (53)

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