Near resonant bubble acoustic cross-section corrections, including examples from oceanography, volcanology, and biomedical ultrasound

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(Received 27 June 2008; revised 23 June 2009; accepted 23 June 2009)

The scattering cross-section σ_s of a gas bubble of equilibrium radius R_0 in liquid can be written in the form $\sigma_s = 4\pi R_0{}^2/[(\omega_1{}^2/\omega^2 - 1)^2 + \delta^2]$, where ω is the excitation frequency, ω_1 is the resonance frequency, and δ is a frequency-dependent dimensionless damping coefficient. A persistent discrepancy in the frequency dependence of the contribution to δ from radiation damping, denoted δ_{rad}, is identified and resolved, as follows. Wildt's [*Physics of Sound in the Sea* (Washington, DC, 1946), Chap. 28] pioneering derivation predicts a linear dependence of δ_{rad} on frequency, a result which Medwin [Ultrasonics **15**, 7–13 (1977)] reproduces using a different method. Weston [Underwater Acoustics, NATO Advanced Study Institute Series Vol. **II**, 55–88 (1967)], using ostensibly the same method as Wildt, predicts the opposite relationship, i.e., that δ_{rad} is *inversely* proportional to frequency. Weston's version of the derivation of the scattering cross-section is shown here to be the correct one, thus resolving the discrepancy. Further, a correction to Weston's model is derived that amounts to a shift in the resonance frequency. A new, corrected, expression for the extinction cross-section is also derived. The magnitudes of the corrections are illustrated using examples from oceanography, volcanology, planetary acoustics, neutron spallation, and biomedical ultrasound. The corrections become significant when the bulk modulus of the gas is not negligible relative to that of the surrounding liquid.

 [DOI: 10.1121/1.3180130]

PACS number(s): 43.20.Ks, 43.30.Pc, 43.35.Bf, 43.35.Zc [ADP] Pages: 2163–2175

I. INTRODUCTION

Gas bubbles play an important role in the generation, scattering, and absorption of sound in a liquid.[1] Applications include performance prediction for search sonar or underwater telemetry, acoustical oceanography, medical and industrial ultrasound, and volcanology. The acoustic properties of bubbles are generally well understood, to the extent that acoustical measurements are sometimes used to determine characteristics of bubble clouds such as their size distribution.[2–5] Such acoustical characterization of bubble properties requires a firm foundation in theory.

The purpose of this article is to highlight and resolve a discrepancy that exists at the heart of the currently accepted theory of bubble acoustics. The scattering cross-section, σ_s, of a small spherical bubble of equilibrium radius R_0, undergoing forced linear pulsations at angular frequency ω, is commonly written in the form

$$\sigma_s = \frac{4\pi R_0{}^2}{(\omega_1{}^2/\omega^2 - 1)^2 + \delta^2}, \tag{1}$$

where ω_1 is the bubble's pulsation resonance frequency and δ is a dimensionless frequency-dependent parameter known variously as the loss factor, damping constant, or damping coefficient. The term "damping coefficient" is adopted here throughout. The value of δ at resonance is equal to the reciprocal of the Q-factor.

In Sec. II two models for δ are described that differ in the frequency dependence of the damping due to acoustic re-radiation in the free field[6] (known as "radiation damping"). In one of these, published by Wildt[7] and Medwin,[2,8] the radiation damping coefficient is directly proportional to frequency, whereas in the other, published by Andreeva[9] and Weston,[10] the proportionality is an inverse one. This discrepancy has hitherto gone largely unnoticed, to the extent that the authors know of only three publications that mention it.[11–13]

In Sec. III two different derivations for σ_s are provided, with particular attention to establishing the correct frequency dependence of δ for small bubbles. It is shown that the discrepancy is caused in part by ambiguity in the *definition* of δ, and three alternative (though not equivalent) definitions for this parameter are suggested, which can be expressed in terms of the unambiguous damping factor β. The first of the two derivations, which includes thermal damping using a generalization of Weston's method, leads to Eq. (25), including a correction term that is not present in Weston's original formulation. The second, starting from Prosperetti's[14] equation of motion, leads to Eq. (43), of identical form to Eq. (25), and also including the new correction term. This second derivation, which permits a more general damping factor, leads further to new expressions for the resonance frequency [Eq. (47)] and extinction cross-section [Eq. (67)]. The resulting expression for σ_s is compared with a reference solution due to Anderson,[15] which, unlike the other models considered, has no restriction on bubble size, and serves as ground truth for the situation involving only acoustic radiation losses. In Sec. IV the persistence of the discrepancy and its consequences for the extinction cross-section are discussed. (The Andreeva–Weston model, shown in Sec. III to be the correct one, was last used in the open literature, to the best of the authors' knowledge, more than 40 years ago,[10,11] whereas the incorrect formulation is widely promulgated through standard reviews.[1,16–18]) This is followed in Sec. V by a description of scenarios in which the magnitude of the required correction is not negligible, and conclusions are summarized in Sec. VI.

II. SCATTERING CROSS-SECTION: PUBLISHED RESULTS

In this section some previously published results for the scattering cross-section of a single bubble are considered, stripping them of all forms of damping other than radiation damping. Thus, except where stated otherwise, the effects of viscosity (of the liquid) and thermal conduction (in the gas) are neglected. Surface tension at the boundary is also neglected. These assumptions are made for simplicity and clarity, in order to highlight the discrepancy in the radiation damping term. The publications form two groups, each adopting a different model for the frequency dependence of the radiation damping term.

A. Wildt–Medwin (WM) model

The first relevant publication is the volume edited by Wildt,[7] Chap. 28 of which presents, for the first time, a detailed description of the response of a bubble to ensonification through resonance. Reference 7 offers a clear physical insight into the important physical mechanisms that give rise to damping at and around the resonance frequency.

Wildt's derivation suggests that if acoustic re-radiation is the only loss mechanism, then δ has a linear dependence on frequency. Specifically, Wildt's formula for the radiation damping coefficient is

$$\delta_{\mathrm{WM}}(\omega) = \frac{R_0}{c}\omega, \tag{2}$$

where c is the speed of sound in the surrounding liquid. Equation (2) is used by Medwin[8] to describe the frequency dependence of radiation damping in Eq. (1), and further promulgated by its use in landmark papers[12,19,20] and standard reviews.[1,16–18] The use of $\delta_{\mathrm{WM}}(\omega)$ from Eq. (2) in place of the radiation damping coefficient in Eq. (1) is referred to as constituting the WM model.

B. Andreeva–Weston (AW) model

A form of the damping coefficient that is less well known is derived by Weston[10] and appears for the first time

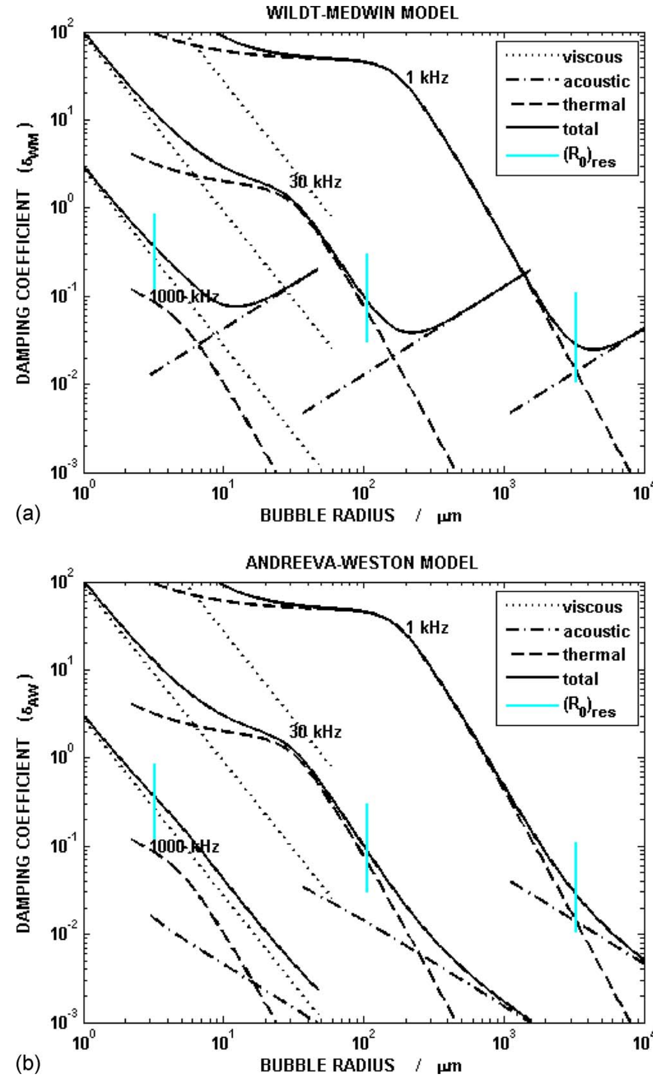

FIG. 1. (Color online) Theoretical damping coefficient vs equilibrium bubble radius for air bubbles in water at atmospheric pressure (0.1 MPa) for acoustic frequencies 1, 30, and 1000 kHz, calculated using (a) the Wildt-Medwin model and (b) the Andreeva–Weston model. The total damping is the black solid line and the contribution due to acoustic radiation is shown as a dash-dotted line (-.-). The remaining curves are for viscous and thermal damping, as indicated by the legend. The resonant bubble radius [denoted $(R_0)_{\mathrm{res}}$] is marked for each frequency using a vertical gray line (cyan online). This quantity is calculated here as the value of the equilibrium bubble radius $R_0=(R_0)_{\mathrm{res}}$ that satisfies the condition $\omega_0(R_0,\omega)=\omega$, where ω_0 is given by Eq. (23) [generalized to incorporate surface tension effects (Ref. 14)].

(without derivation) in the work of Andreeva.[9] Although Weston does not introduce the variable δ explicitly, his derivation of σ_s results in an expression that is consistent with Eq. (1) only if the radiation damping coefficient is *inversely* proportional to frequency, that is, if δ is replaced with δ_{AW}, given by

$$\delta_{\mathrm{AW}}(\omega) = \frac{\omega_1{}^2 R_0}{c}\omega^{-1}. \tag{3}$$

The use of $\delta_{\mathrm{AW}}(\omega)$ from Eq. (3) in place of the radiation damping coefficient in Eq. (1) is referred to as constituting the AW model. A graph similar to Fig. 1 from Medwin's paper,[8] showing the variation in the total damping coefficient with bubble radius R_0 at three frequencies, is presented here as Fig. 1(a), using Eq. (2) for the radiation damping coefficient, proportional to R_0. Figure 1(b) shows the damping coefficient plotted in the same way, except that the contribution from acoustic radiation is calculated using Eq. (3). Both graphs include viscous and thermal damping, as well as the effects of surface tension. The discrepancy between WM and AW models of radiation damping is apparent from the lower right portion of each solid curve, where the radiation damping term is dominant.

Apart from by Weston himself,[11,21] use of the AW model is rare. The only other publication the authors know of is that of Anderson and Hampton,[12] which presents three different equations for σ_s: first, their Eq. (54) (attributed by Anderson and Hampton[12] to Spitzer[22] and abbreviated here as AH-54), which Anderson and Hampton state is not valid away from resonance, and is identical to the WM model; second, a corrected version, AH-55, which is identical to the AW model; and third, AH-56, which includes additional effects due to viscous and thermal damping and forms the basis of their subsequent calculations of bubble attenuation vs frequency. Once stripped of these extra complications, AH-56 reduces not to AW but to WM, making AH-55 and AH-56 mutually inconsistent.

III. SCATTERING CROSS-SECTION: THEORY

Spherically symmetrical pulsations of a single gas bubble of negligible density in an infinite volume of liquid are considered. Except in Sec. III E the bubble radius is assumed to be small compared with the acoustic wavelength in the liquid. Perturbations to the bubble's radius are assumed small, permitting use of the methods of linear acoustics.

A. Damping factor (β)

Following Morfey,[23] the expression "damping factor" is used in this paper to refer to the parameter β in the equation of motion

$$\ddot{X} + 2\beta\dot{X} + \omega_{\mathrm{nat}}{}^2 X = 0, \tag{4}$$

where the dots represent time derivatives and ω_{nat} is the undamped natural frequency. In the following Eq. (4) is applied to the bubble, with X representing the departure of the bubble's radius (R) from its equilibrium value ($X=R-R_0$). Considering further a sinusoidal forcing term of angular frequency ω, and in general permitting β to vary with frequency, $\beta \to \beta(\omega)$, the equation of motion then becomes

$$\ddot{R} + 2\beta(\omega)\dot{R} + K(\omega)(R - R_0) = F(\omega)e^{i\omega t}, \tag{5}$$

in which R is understood to be a complex variable and β, K are real parameters that are independent of time, representing resistive and elastic forces, respectively. Like β, the parameters K and F can also be functions of the forcing frequency ω. The effect of inertia is included in the forcing term, so that

$$Fe^{i\omega t} = -\frac{4\pi R_0{}^2}{m_{\mathrm{RF}}^{\mathrm{rad}}} P_F(t), \tag{6}$$

where P_F is the external forcing pressure, $m_{\mathrm{RF}}^{\mathrm{rad}}$ is the radiation mass in the radius-force frame,[1] equal to three times the displaced liquid mass

$$m_{\mathrm{RF}}^{\mathrm{rad}} = 4\pi\rho_0 R_0{}^3, \tag{7}$$

and ρ_0 is the equilibrium density of the liquid.

The particular solution to Eq. (5) is

$$R = R_0 + \frac{F}{K - \omega^2 + 2i\beta\omega} e^{i\omega t}, \tag{8}$$

and hence

$$|R - R_0|^2 = \frac{|F|^2/\omega^4}{(K/\omega^2 - 1)^2 + (2\beta/\omega)^2}. \tag{9}$$

If the term $2\beta/\omega$ is small, then the maximum response occurs when ω is equal to $\sqrt{K}$, which means that K may be approximated in Eq. (9) by the square of the resonance frequency ($K \approx \omega_1{}^2$). If this substitution is made, the similarity between the denominators of Eq. (9) (the radial excursion) and Eq. (1) (the scattering cross-section) makes it tempting to assume further that δ is equal to $2\beta/\omega$, but is it correct to do so? It turns out there is no simple answer to this question, as the true relationship between δ and β depends on the precise definition of δ, which is explored further below.

B. Derivation of the damping coefficient (δ), based on Weston

The following derivation follows the method of Weston,[10] generalized by replacing the specific heat ratio (γ) with a complex polytropic index (denoted Γ). Consider a plane wave p_i of pressure amplitude A and angular frequency ω:

$$p_i = A \exp[i\omega(t - x/c)], \tag{10}$$

incident on a spherical bubble placed with its center at the origin such that the factor F in Eq. (5) is given by

$$F = -\frac{A}{\rho_0 R_0} \tag{11}$$

and

$$P_F = p_i(x=0) = A \exp(i\omega t). \tag{12}$$

Assume that the scattered wave p_s is a spherical one of amplitude $|B|/r$, such that

$$p_s = (B/r)\exp[i\omega(t - r/c)], \tag{13}$$

where r is the distance from the origin. The scattering cross-section σ_s can then be defined in terms of the ratio B/A as

$$\sigma_s \equiv 4\pi|B/A|^2, \tag{14}$$

Weston's derivation for this ratio is now followed. Euler's equation relates p_s to the radial component of particle velocity u:

$$-\frac{\partial p_s}{\partial r} = \rho_0 \frac{\partial u}{\partial t}. \tag{15}$$

Substituting Eq. (13) into Eq. (15) relates the particle velocity in the liquid at distance r from the bubble center to the acoustic pressure associated with the spherical radiated field, $p_s(r)$, giving

$$u(r) = \left(1 - \frac{i}{\omega r/c}\right)\frac{p_s(r)}{\rho_0 c}, \tag{16}$$

which, when evaluated at the bubble wall, relates the scattered amplitude to the rate of change of bubble volume V (assuming that departures from the bubble's rest radius are small)

$$\frac{B}{R_0{}^2}(1 + i\omega R_0/c)\exp[i\omega(t - R_0/c)] = \frac{i\omega\rho_0}{4\pi R_0{}^2}\frac{\mathrm{d}V}{\mathrm{d}t}. \tag{17}$$

An expression for $\mathrm{d}V/\mathrm{d}t$ can be found by differentiating the polytropic relationship between pressure and volume (PV^Γ =constant, where Γ is the complex polytropic index of the air bubble[17,24]). If the hydrostatic pressure is P_0 and the acoustic pressure inside the bubble is p_b, such that the total interior pressure is P_0+p_b, the result is

$$\mathrm{d}V/\mathrm{d}t = -i\omega p_b V_0/\Gamma P_0, \tag{18}$$

where V_0 is the unperturbed bubble volume. Substituting for $\mathrm{d}V/\mathrm{d}t$ in Eq. (17) and rearranging for the interior acoustic pressure

$$p_b = 4\pi\frac{\Gamma P_0 B}{\rho_0 V_0 \omega^2}(1 + i\omega R_0/c)\exp[i\omega(t - R_0/c)]. \tag{19}$$

In the absence of surface tension, this pressure must equal the sum of the incident and scattered fields [using Eqs. (10) and (13)]:

$$p_b = \left(A + \frac{B}{R_0}e^{-i\omega R_0/c}\right)e^{+i\omega t}. \tag{20}$$

Equating the right hand sides of Eqs. (19) and (20), and solving for the ratio B/A, the result is (introducing the shorthand ε for $\omega R_0/c$)

$$\frac{B}{A} = \frac{R_0}{[(1+i\varepsilon)\Omega^2/\omega^2 - 1]e^{-i\varepsilon}}, \tag{21}$$

where Ω is a complex parameter given by the equation

$$\Omega(R_0,\omega)^2 = 4\pi\frac{\Gamma(R_0,\omega)P_0 R_0}{\rho_0 V_0(R_0)}, \tag{22}$$

the real part of which is closely related to the pulsation resonance frequency. Specifically, if Γ is real and independent of frequency (which occurs for both isothermal and adiabatic pulsations), then Ω is equal to the bubble's natural frequency. For example, in the adiabatic case Γ and Ω are equal to the specific heat ratio and the Minnaert frequency,[25] respectively.

To proceed further, the following variables are defined

$$\omega_0(R_0,\omega) \equiv \sqrt{\mathrm{Re}[\Omega(R_0,\omega)^2]} \tag{23}$$

and

$$\beta_{\mathrm{th}}(R_0,\omega) \equiv \frac{\mathrm{Im}[\Omega(R_0,\omega)^2]}{2\omega}. \tag{24}$$

The variable ω_1 was introduced earlier as the "resonance frequency" but not properly defined. The new variable ω_0 plays a similar role and can be thought of as a more rigorously defined version of the same variable, though it is more closely related to the natural frequency than the resonance frequency. Wherever the symbol ω_0 is used below, it is always in the sense of Eq. (23).

With these definitions, from Eq. (21) the following generalization of Weston's expression for the scattering cross-section is obtained:

$$\sigma_s = \frac{4\pi R_0{}^2}{\left(\frac{\omega_0{}^2}{\omega^2} - 1 - 2\frac{\beta_{\mathrm{th}}}{\omega}\varepsilon\right)^2 + \delta_{\mathrm{AW}}(\omega)^2}, \tag{25}$$

where

$$\delta_{\mathrm{AW}}(\omega) \equiv \mathrm{Im}\,\frac{p_i(0)}{p_s(R_0)} = 2\frac{\beta_{\mathrm{th}}}{\omega} + \frac{\omega_0{}^2}{\omega^2}\varepsilon. \tag{26}$$

If thermal effects are neglected (implying that $\omega_0=\omega_{\mathrm{nat}}$ and $\beta_{\mathrm{th}}=0$), Equation (25) simplifies to the AW model with ω_1 equal to ω_{nat}. The correction term $-2\beta_{\mathrm{th}}\varepsilon/\omega$ in the denominator of Eq. (25) [amounting to a fractional correction to the resonance frequency of $\beta_{\mathrm{th}}(\omega_0)R_0/c$] is new.

Wildt's derivation makes the same assumptions and follows an almost identical procedure as Weston's, so why does it result in a different expression for δ [Eq. (2)]? A close look at Wildt's derivation reveals a subtle error on p. 462. The error occurs in the step from Wi-17 to Wi-22, where the abbreviation Wi-n indicates Eq. (n) from Ref. 7. Specifically, although Wi-13, Wi-16, and Wi-17 are correct to first order in ε, a missing *second* order term is required for the step to Wi-22. To illustrate the nature and importance of this missing term, the expansion $\exp(-i\varepsilon)=1-i\varepsilon-\varepsilon^2/2+O(\varepsilon^3)$ is substituted in Eq. (21) to obtain

$$\frac{B}{A} = \frac{R_0}{\Omega^2/\omega^2 - 1 + \varepsilon^2(\Omega^2/\omega^2+1)/2 + i\varepsilon + O(\varepsilon^3)}. \tag{27}$$

Substituting Eq. (27) in Eq. (14) gives Eq. (25) to this order of accuracy, again consistent with the AW model.

One might conclude from this that Wildt's equation for δ [Eq. (2)] is incorrect. Indeed, if δ is defined through Eq. (1), that would seem to be the only *possible* conclusion. In order to be unambiguous, however, such a definition of δ requires the resonance frequency ω_1 to be defined first. Both national[26] and international[27] standards provide different definitions of resonance frequency to choose from, depending on the type of resonance (peak response of, for example, scattered pressure, or bubble wall velocity, or displacement) each leading to a different δ. The most obvious choice for the present purpose would be to define the resonance frequency as the frequency that maximizes σ_s, but this choice leads to an internal contradiction, because this frequency *cannot* be equal to ω_1 in Eq. (1) unless the derivative $\delta'(\omega)$ vanishes at $\omega=\omega_1$. The issue of the resonance frequency is addressed in Sec. III D, but here a second interpretation is considered, based on Wildt's implied definition (from Wi-32) as the imaginary part of the ratio of R_0 to the scattering amplitude. Denoting this quantity δ_{Wildt}:

$$\delta_{\mathrm{Wildt}}(\omega) \equiv \mathrm{Im}\,\frac{R_0}{B/A}, \tag{28}$$

so that

$$\delta_{\mathrm{Wildt}}(\omega) = \mathrm{Im}\left(\frac{p_i(0)}{p_s(R_0)}e^{-i\varepsilon}\right). \tag{29}$$

With this definition it follows [by substituting Eq. (21) into Eq. (28)] that

$$\begin{aligned}\delta_{\mathrm{Wildt}}(\omega) &= \left(2\frac{\beta_{\mathrm{th}}}{\omega} + \frac{{\omega_0}^2}{\omega^2}\varepsilon\right)\cos\varepsilon - \left(\frac{{\omega_0}^2}{\omega^2} - 1 - 2\frac{\beta_{\mathrm{th}}}{\omega}\varepsilon\right)\sin\varepsilon \\ &= 2\frac{\beta_{\mathrm{th}}}{\omega} + \varepsilon + \frac{\beta_{\mathrm{th}}}{\omega}\varepsilon^2 - \frac{1}{6}\left(2\frac{{\omega_0}^2}{\omega^2} + 1\right)\varepsilon^3 + O(\varepsilon^4)\end{aligned} \tag{30}$$

and

$$\sigma_s = \frac{4\pi {R_0}^2}{\left(\frac{{\omega_0}^2}{\omega^2} - 1\right)^2 + {\delta_{\mathrm{Wildt}}}^2 + \left(\frac{{\omega_0}^4}{\omega^4} - 1\right)\varepsilon^2 + O(\varepsilon^2 {\delta_{\mathrm{Wildt}}}^2)}, \tag{31}$$

replacing Wi-34, which is missing the term $({\omega_0}^4/\omega^4 - 1)\varepsilon^2$.

The physics underlying the source of these discrepancies is illustrated by Eq. (16). The ratio of the local scattered pressure field to the local particle velocity contains both real and imaginary parts [through substitution of Eq. (13) into Eq. (15)]. At the limit of $r\to\infty$, this ratio is real and equal to the impedance of a plane wave, the pressure and velocity are in phase, and indeed locally the wavefront appears planar at $r\to\infty$. At the limit of $r\to 0$, they would be $\pi/2$ out of phase, but this limit cannot be achieved because the bubble wall prevents one tracking back from $r\to\infty$ to $r\to 0$. On such a track the magnitude and phase of the ratio of p_s to u changes from the $r\to\infty$ value, the phase difference increasing towards $\pi/2$ without reaching it. The discrepancy lies in the order to which one approximates by how much the magnitude and phase of the ratio differs from the $r\to 0$ value. This is made clear by the way the $\varepsilon=\omega R_0/c$ terms enter the derivation of Prosperetti:[14] the amplitude term $\varepsilon/(1+\varepsilon^2)^{1/2}$ is retained to second order (equations 3.102, 3.105, and 4.190 of Ref. 1).

It is shown above that, after correcting the error in Wildt's derivation, the results of Wildt[7] and Weston[10] are consistent. It now remains to investigate how Medwin,[2] who uses the damping model of Devin,[28] independently reproduces Wildt's original (uncorrected) result, thus creating a second discrepancy, this time between Weston[10] and Medwin.[2,8] Prosperetti's[14] formulation is now used to address this remaining discrepancy.

C. Alternative derivation of the damping coefficient, based on Prosperetti

Building on the work of Smith,[29] Devin[28] derives an equation of motion for the bubble volume that includes effects of viscous, thermal, and acoustic damping. Prosperetti[14] derives an equation of motion for the bubble radius, including $O(\varepsilon^2)$ correction terms to Devin's equation, that can be written (in the present notation) as

$$\ddot{R} + 2\beta\dot{R} + K(R - R_0) = -\frac{A}{\rho_0 R_0}e^{+i\omega t}, \tag{32}$$

where (neglecting effects of surface tension for consistency with Sec. III B)

$$K = {\omega_0}^2 + \frac{\varepsilon^2}{1+\varepsilon^2}\omega^2, \tag{33}$$

$$\beta = \beta_0 + \frac{\varepsilon}{1+\varepsilon^2}\frac{\omega}{2} \tag{34}$$

and β_0 is the contribution to the damping factor β from mechanisms other than acoustic radiation. [For example, the combined contribution from shear viscosity μ and thermal losses would be $\beta_0=\beta_{\mathrm{th}}+2\mu/\rho_0 {R_0}^2$ (Refs. 14 and 30).]

The input impedance Z is defined as the ratio of incident pressure (at $x=0$) to the particle velocity at $r=R_0$:

$$Z \equiv \frac{p_i(x=0)}{u_s(r=R_0)} = i\rho_0 c\varepsilon\left(\frac{K}{\omega^2} - 1 + 2i\frac{\beta}{\omega}\right). \tag{35}$$

An expression for the scattered pressure can then be derived in terms of β by use of Euler's equation (at $r=R_0$):

$$u_s(R_0) = -i\frac{1+i\varepsilon}{\rho_0 c\varepsilon}p_s(R_0). \tag{36}$$

Eliminating u_s from Eqs. (35) and (36) gives

$$\frac{p_i(0)}{p_s(R_0)} = -i\frac{1+i\varepsilon}{\rho_0 c\varepsilon}Z, \tag{37}$$

which can be written as

$$\frac{p_i(0)}{p_s(R_0)} = \frac{{\omega_0}^2}{\omega^2} - 1 - 2\frac{\varepsilon\beta_0}{\omega} + i\left(2\frac{\beta_0}{\omega} + \frac{{\omega_0}^2}{\omega^2}\varepsilon\right). \tag{38}$$

Substitution of Eq. (38) in Eq. (29) results in an equation identical to Eq. (30) except with β_{th} replaced by the more general β_0.

Calculating the scattering cross-section with radiation damping only [i.e., set $\beta_0=0$ in the squared modulus of Eq. (38)] the result is Eq. (1), with $\omega_1=\omega_0$ and Eq. (3) for the damping coefficient, once again in agreement with the AW model, which therefore must be the correct one.

Medwin[2,8] defines δ as

$$\delta_{\mathrm{Medwin}}(\omega) \equiv \frac{2\beta}{\omega}, \tag{39}$$

and then uses this expression for δ in Eq. (1), implicitly (and incorrectly) assuming that it is equal to the imaginary part of the pressure ratio [Eq. (38)]. Substituting Prosperetti's result for β in Eq. (39) gives

$$\delta_{\mathrm{Medwin}}(\omega) = \frac{2\beta_0}{\omega} + \frac{\varepsilon}{1+\varepsilon^2} = 2\frac{\beta_0}{\omega} + \varepsilon - \varepsilon^3 + O(\varepsilon^5). \quad (40)$$

Comparison with the imaginary part of Eq. (38) demonstrates that this assumption results in an unwanted factor $\omega^2/\omega_0^{\,2}$ in the radiation damping term, coincidentally reproducing Wildt's result for σ_s and reinforcing the erroneous impression that Medwin's and Wildt's expressions for the scattering cross-section are both correct. To lowest order in ε and β_0, the damping coefficients δ_{Wildt} and δ_{Medwin} are equal:

$$\delta_{\mathrm{Medwin}} = \delta_{\mathrm{Wildt}} - \frac{\beta_0}{\omega}\varepsilon^2 - \frac{1}{6}\left(5 - 2\frac{\omega_0^{\,2}}{\omega^2}\right)\varepsilon^3 + O(\varepsilon^4). \quad (41)$$

A convenient form for σ_s that follows from Eqs. (35) and (37) [with Eq. (39)] is

$$\sigma_s = \frac{4\pi R_0^{\,2}(1+\varepsilon^2)^{-1}}{(K/\omega^2 - 1)^2 + \delta_{\mathrm{Medwin}}^{\,2}}, \quad (42)$$

or (equivalently)

$$\sigma_s = \frac{4\pi R_0^{\,2}}{\left(\frac{\omega_0^{\,2}}{\omega^2} - 1 - 2\frac{\beta_0}{\omega}\varepsilon\right)^2 + \left(2\frac{\beta_0}{\omega} + \frac{\omega_0^{\,2}}{\omega^2}\varepsilon\right)^2}. \quad (43)$$

Equation (43) is derived from Prosperetti's equation of motion. It is identical in functional form to Eq. (25), which is derived using the generalization of Weston's method that was outlined in Sec. III B. The only difference between them is the appearance in Eq. (43) of the more general β_0 instead of β_{th} for the non-acoustic damping factor.

Comparison of Eq. (40) (with $\beta_0=\beta_{\mathrm{th}}$) with Eq. (26) then gives

$$\delta_{\mathrm{Medwin}} = \delta_{\mathrm{AW}} + \frac{\varepsilon}{1+\varepsilon^2} - \varepsilon\frac{\omega_0^{\,2}}{\omega^2}. \quad (44)$$

D. Effect of radiation damping on the resonance frequency

The resonance frequency predicted by the AW and MW models and by Eq. (43) are now compared. Specifically, the WM and AW models are considered in the form

$$\sigma_{\mathrm{WM}} = \frac{4\pi R_0^{\,2}}{\left(\frac{\omega_0^{\,2}}{\omega^2} - 1\right)^2 + \left(2\frac{\beta_0}{\omega} + \varepsilon\right)^2} \quad (45)$$

and

$$\sigma_{\mathrm{AW}} = \frac{4\pi R_0^{\,2}}{\left(\frac{\omega_0^{\,2}}{\omega^2} - 1\right)^2 + \left(2\frac{\beta_0}{\omega} + \frac{\omega_0^{\,2}}{\omega^2}\varepsilon\right)^2}. \quad (46)$$

In all three cases, the ratio $\sigma_s/R_0^{\,2}$ is a function of the three parameters ω/ω_0, ε_0, and β_0/ω_0, where $\varepsilon_0=\omega_0 R_0/c$.

For a pressure resonance, the resonance frequency can be defined[26,27] as the frequency at which the magnitude of the mean square scattered pressure response (proportional to σ_s) is maximized. Adopting this definition and denoting the corresponding resonance frequencies ω_{43}, ω_{WM}, and ω_{AW}, gives the result

$$\left(\frac{\omega_0}{\omega_{43}}\right)^2 = 1 - \frac{2\beta_0^{\,2}}{\omega_0^{\,2}} - \frac{\varepsilon_0^{\,2}}{2}, \quad (47)$$

$$\left(\frac{\omega_0}{\omega_{\mathrm{WM}}}\right)^2 = 1 - \frac{2\beta_0^{\,2}}{\omega_0^{\,2}} + \frac{\varepsilon_0^{\,2}/2}{1 - 2\beta_0^{\,2}/\omega_0^{\,2}} + O(\varepsilon_0^{\,4}) \quad (\varepsilon_0^{\,2} \ll 1) \quad (48)$$

and

$$\left(\frac{\omega_0}{\omega_{\mathrm{AW}}}\right)^2 = 1 - \frac{2\beta_0^{\,2}}{\omega_0^{\,2}} - \frac{\varepsilon_0^{\,2}}{2} - 2\frac{\varepsilon_0\beta_0}{\omega_0}. \quad (49)$$

No approximation is involved in the derivation of either Eq. (47) [from Eq. (43)] or Eq. (49) [from Eq. (46)], except for the assumption that both β_0 and ω_0 are independent of frequency. That of Eq. (48) [from Eq. (45)] requires the further assumption that $\varepsilon_0^{\,2}$ is small.

The main point here is that the $O(\varepsilon_0^{\,2})$ term in Eq. (48) has the wrong sign. This sign error, which can be traced back to the incorrect frequency dependence in the radiation damping term of the WM model, implies that the WM model systematically underestimates the resonance frequency by a fraction of order $\varepsilon_0^{\,2}$. If a measurement of the resonance frequency were used to estimate the bubble radius, the WM model would lead to a bias of the same order in the inferred radius.

E. Comparison with breathing mode solution for $\beta_0=0$

The scattering amplitude for the breathing mode of a spherical gas bubble of arbitrary radius (i.e., without restriction on the magnitude of ε_0) is now evaluated for the case when $\beta_0=0$. It has already been shown theoretically that (of the models considered so far) AW [or Eq. (43)] is the correct version. The purpose of the present section is to show that there exists a regime in which ε is large enough for the discrepancy to become an issue, while remaining small enough for the derivation to hold. The amplitude of the scattered wave associated with the pulsating or "breathing" mode (denoted B_{bm}) is determined by[15] (see also Ref. 16)

$$\frac{AR_0}{B_{\mathrm{bm}}(x)} = \frac{\frac{\omega_0^{\,2}}{\omega^2}\varepsilon\sin\varepsilon - \cos\varepsilon[1-\xi(x)]}{\frac{\omega_0^{\,2}}{\omega^2}\cos\varepsilon + \frac{\sin\varepsilon}{\varepsilon}[1-\xi(x)]} + i\varepsilon, \quad (50)$$

where

$$\xi(x) \equiv \frac{\omega_0^{\,2}}{\omega^2} + 1 - \frac{3}{x^2}(1 - x\cot x), \quad (51)$$

$$x = \frac{\omega}{\omega_0}\sqrt{\frac{\rho_g}{\rho_0}}, \quad (52)$$

and ρ_g is the equilibrium gas density. The scattering cross-section for the breathing mode is introduced as

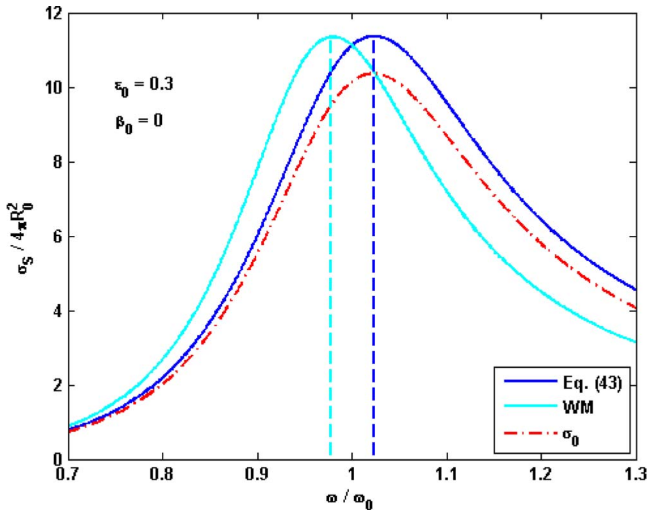

FIG. 2. (Color online) Theoretical pressure response (normalized scattering cross-section, $\sigma_s/4\pi R_0{}^2$) vs dimensionless frequency ω/ω_0. Black solid line (blue online): Eq. (43); gray line (cyan online): Eq. (45); dash-dot line: Eq. (54) with Eq. (50) and x=0. Damping coefficients at resonance are ε_0=0.3 and β_0=0.

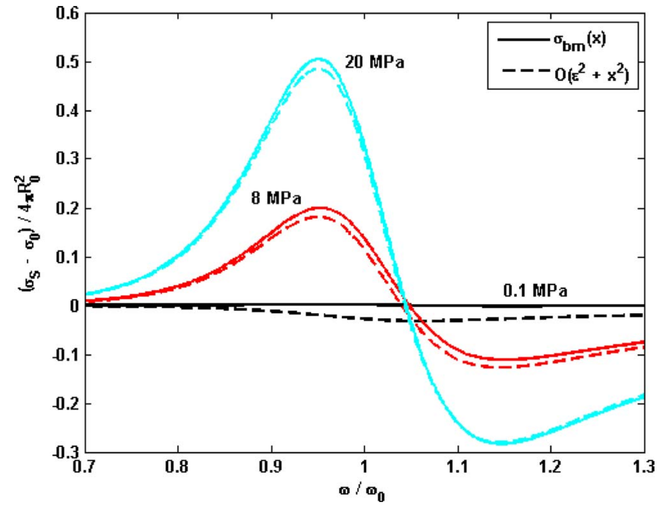

FIG. 3. (Color online) Difference in normalized scattering cross-section $(\sigma_s-\sigma_0)/4\pi R_0{}^2$ vs dimensionless frequency ω/ω_0, for an air bubble in water at pressure as marked, calculated using Anderson's expression for the breathing mode [Eq. (53), with Eq. (50) for B_{bm}, solid lines] and the approximation [Eq. (58), dashed lines]. The corresponding density ratios ρ_g/ρ_0 are 0.00125 (for a pressure of 0.1 MPa), 0.100 (8 MPa), and 0.250 (20 MPa). The radiation damping coefficient at resonance is ε_0=0.3.

$$\sigma_{\mathrm{bm}}(x) \equiv 4\pi |B_{\mathrm{bm}}(x)/A|^2, \tag{53}$$

with B_{bm} given by Eq. (50), and the variable σ_0 is defined as this cross-section evaluated with zero gas density

$$\sigma_0 \equiv \sigma_{\mathrm{bm}}(0). \tag{54}$$

This quantity is plotted in Fig. 2 for the case ε_0=0.3 and compared with the approximations WM and Eq. (43) for the same case, and with β_0=0, for consistency with Anderson's model [Eq. (43) and AW are identical for this case]. This graph is valid for any gas of negligible density in any liquid. As expected from the sign error in Eq. (48), WM underestimates the resonance frequency (this is caused by the error in radiation damping), whereas Eq. (43) gives the correct resonance frequency. Both WM and Eq. (43) overestimate the maximum breathing mode scattering cross-section. This anomaly is explained below, in the discussion following Eq. (58).

The effect of departures from zero gas density are now considered by plotting the difference between σ_{bm} and σ_0 in Fig. 3 (solid curves). This graph shows that an increase in gas density reduces the resonance frequency, an effect that can be understood by considering the behavior of σ_{bm} for small values of the ratio ρ_g/ρ_0 as follows. Equation (50) can be approximated, if the gas density is small, using

$$\xi(x) \approx \frac{\omega_0{}^2}{\omega^2} - \frac{x^2}{15}, \tag{55}$$

such that

$$\frac{AR_0}{B_{\mathrm{bm}}} \approx \frac{\dfrac{\omega_0{}^2}{\omega^2}\varepsilon \sin \varepsilon - \cos \varepsilon \left(1 - \dfrac{\omega_0{}^2}{\omega^2} + \dfrac{1}{15}\dfrac{\rho_g}{\rho_0}\dfrac{\omega^2}{\omega_0{}^2}\right)}{\dfrac{\omega_0{}^2}{\omega^2}\cos \varepsilon + \dfrac{\sin \varepsilon}{\varepsilon}\left(1 - \dfrac{\omega_0{}^2}{\omega^2} + \dfrac{1}{15}\dfrac{\rho_g}{\rho_0}\dfrac{\omega^2}{\omega_0{}^2}\right)} + i\varepsilon. \tag{56}$$

If ε is also small, expanding to $O(\varepsilon^2)$ gives

$$\frac{AR_0}{B_{\mathrm{bm}}} \approx \frac{\dfrac{\omega_0{}^2}{\omega^2} - 1 - \dfrac{1}{15}\dfrac{\rho_g}{\rho_0}\dfrac{\omega^2}{\omega_0{}^2} + \dfrac{\varepsilon^2}{2}\left(1 + \dfrac{\omega_0{}^2}{\omega^2}\right)}{1 + \dfrac{1}{15}\dfrac{\rho_g}{\rho_0}\dfrac{\omega^2}{\omega_0{}^2} - \dfrac{\varepsilon^2}{6}\left(1 + 2\dfrac{\omega_0{}^2}{\omega^2}\right)} + i\varepsilon. \tag{57}$$

Using Eq. (57) for the amplitude of the breathing mode yields the approximate result

$$\sigma_{\mathrm{bm}} \approx 4\pi R_0{}^2 \frac{1 - \dfrac{\varepsilon^2}{3}\left(1 + 2\dfrac{\omega_0{}^2}{\omega^2}\right) + \dfrac{2}{15}\dfrac{\rho_g}{\rho_0}\dfrac{\omega^2}{\omega_0{}^2}}{\left(\dfrac{\omega_0{}^2}{\omega^2} - 1 - \dfrac{1}{15}\dfrac{\rho_g}{\rho_0}\dfrac{\omega^2}{\omega_0{}^2}\right)^2 + \varepsilon^2\dfrac{\omega_0{}^4}{\omega^4}}. \tag{58}$$

This expression is evaluated and the difference relative to σ_0 plotted in Fig. 3 (dashed lines) to enable comparison with its counterpart evaluated without these approximations. The graph is calculated for an air bubble in water at a temperature of 283 K. It applies more generally to any gas in any liquid with the density ratios stated in the figure caption. Neglecting the gas density in Eq. (58) results in the AW model for the denominator and an order ε^2 correction in the numerator. This correction to the numerator explains the amplitude anomaly of Fig. 2, without affecting the frequency of resonance.

The resonance frequency associated with Eq. (58) (denoted ω_{58}), to lowest order in $\varepsilon_0{}^2$ and ρ_g/ρ_0, is given by

$$\left(\frac{\omega_0}{\omega_{58}}\right)^2 = 1 - \frac{\varepsilon_0{}^2}{2} + \frac{\rho_g}{15\rho_0}. \tag{59}$$

By a curious numerical coincidence, the two correction terms in Eq. (59) approximately cancel for a bubble of air in water at atmospheric pressure. The precise ratio for an ideal gas at temperature T is

$$\frac{15\varepsilon_0{}^2\rho_0}{2\rho_g} = \frac{45k_B T}{2m_g c^2}\,\mathrm{Re}\,\Gamma, \tag{60}$$

where k_B is Boltzmann's constant $(1.381\times10^{-23}\ \mathrm{J/K})$ and m_g is the average mass of an air molecule $(4.82\times10^{-26}\ \mathrm{kg})$. Using round values of $T=300$ K and $c=1500$ m/s gives approximately 0.81 $\mathrm{Re}(\Gamma)$ for the right hand side, i.e., between 0.8 (for isothermal pulsations) and 1.2 (for adiabatic ones).

It is argued above that the radiation damping can dominate if ε is sufficiently large. This statement seems to conflict with the initial assumption (made in order to satisfy the requirement of uniform pressure at the bubble wall) that ε is "small." Some experimental conditions have forced pragmatic solutions where the tractable approach has been to apply formulations known to be derived assuming that $\varepsilon\to0$, but where this is not the case in practice. The need to ensonify across the range of bubble pulsation resonances to obtain a size distribution for bubbles in a population spanning orders of magnitude, probably meant that Leighton *et al.*[5] worked at up to $\varepsilon\sim0.2$. The application of a two-frequency technique by Newhouse and Shankar[31] probably generated exposure exceeding $\varepsilon\sim2$.

Nevertheless, it is shown above that there exists a regime in which the ε^2 terms are needed, while the derivation remains valid. Prosperetti[14] also argues that $O(\varepsilon^2)$ terms are not only justified, but *necessary* in the regime when $\varepsilon/2\pi$ is small but ε is of order 1. A further counter-argument is one of principle, as follows. A derivation that purports to be accurate to order ε^2 must include *all* terms of that order. Some of the terms might be negligible for some conditions, but the correct way to identify the circumstances in which they may be legitimately neglected is to derive the formally correct solution and only then consider which terms to omit.

F. Confusion caused by the use of dimensionless δ in σ_s

As the preceding text shows, the use of a dimensionless damping coefficient without adequate definition creates confusion. It would not be correct to state categorically that one or other expression for the dimensionless damping coefficient is right or wrong, because any can be perceived as being correct in complying with each respective definition (giving rise, in the present notation, to δ_{Wildt}, δ_{Medwin}, and δ_{AW}). However, the same ambiguity does not apply to the definition of the scattering cross-section, so one *can* make such a statement about σ_s. Thus, the WM model for σ_s [Eq. (45)] is missing a term of order ε^2 in the denominator. This is the same order as δ^2 itself, making it a correction to leading order in the damping term, translating to the sign error in the corresponding correction term in the expression for the resonance frequency ω_{WM} [Eq. (48)]. The confusion can be mitigated by avoiding use of the dimensionless coefficient δ, replacing it with the unambiguous damping factor β as in Eq. (43).

IV. PERSISTENCE OF THE DISCREPANCY, AND THE EXTINCTION CROSS-SECTION

A. Persistence of the discrepancy: The example of ultrasound contrast agents

The result identified as incorrect by the analysis of Sec. III originates from early research related to search sonar[7] and is now in widespread use in acoustical oceanography. (References 18 and 32 are recent examples taken from many possible candidates). A more recent application that is now explored in more detail arises in biomedical acoustics,[33] namely, in the study of ultrasound contrast agents (UCAs), i.e., microbubbles used to enhance the contrast of ultrasound images. This application is chosen because it illustrates how this previously unchallenged discrepancy has been exported to another field and because the case of UCAs provides a convenient demonstration involving the relationship between the extinction and scattering cross-sections. The narrow bubble size range of UCAs promoted a technique of fitting the measured ultrasonic scatter to models of the scattering and extinction cross-sections, in order to produce empirical estimates of, say, the elasticity[34] or frictional losses in the bubble wall[35] in order to determine the mechanical properties of the stabilized bubble wall. Having an incorrect expression for one of the fixed parameters (radiation damping) is unsatisfactory, especially because since its pioneering introduction in the early 1990s,[34–36] the approach became widely used around the world, with the incorrect formulation appearing in dozens of research papers and reviews.[33,37,38] Additional difficulties are exemplified by experiments with UCAs to measure the attenuation of the ultrasonic signal and then compare these data with the computed extinction cross-section for the bubbles. These difficulties are described below.

B. Confusion caused by the use of dimensionless δ in σ_e

The accepted formula for the extinction cross-section (σ_e) of a bubble can be written as[1,2,8,16]

$$\sigma_e = \sigma_s \frac{\delta}{\delta_{\mathrm{rad}}}, \tag{61}$$

where σ_s is given by Eq. (1) and the denominator,

$$\delta_{\mathrm{rad}} = \delta - 2\beta_0/\omega, \tag{62}$$

is the contribution to the damping coefficient from radiation damping alone. Of the various possible definitions for δ though, the following questions are now posed: which one should be used in (a) the right hand side of Eq. (62), (b) the numerator of Eq. (61), and (c) the expression for σ_s [Eq. (1)]?

To answer these questions the definition of the extinction cross-section is considered as the ratio of the mean rate of work done on the bubble to the mean intensity of the incident plane wave. This definition leads to

$$\sigma_e = -\frac{8\pi\rho_0 c R_0{}^2}{|p_i|^2}\,\overline{\mathrm{Re}(p_i)\mathrm{Re}(p_i/Z)}, \tag{63}$$

and hence

$$\sigma_e = \frac{4\pi R_0^{\,2}}{(K/\omega^2-1)^2+\delta_{\mathrm{Medwin}}^{\;2}}\frac{\delta_{\mathrm{Medwin}}}{\varepsilon}, \tag{64}$$

which can be written as

$$\sigma_e = \sigma_s \frac{\delta_{\mathrm{Medwin}}}{\varepsilon}(1+\varepsilon^2). \tag{65}$$

Equation (65) shows that the answer to both (a) and (b) is δ_{Medwin} (or the approximately equivalent δ_{Wildt}), while the correct answer to (c) is shown in Sec. III to be δ_{AW}.

Therefore, if the dimensionless damping coefficient is used to encompass the losses, then one is faced with the unsatisfactory conclusion that, unless correction terms are applied to the currently accepted equations for these cross-sections, there is no single definition of δ that gives the correct result for both σ_s and σ_e, i.e., correct substitution of Eq. (1) into Eq. (61) requires use of both δ_{Medwin} and δ_{AW}:

$$\sigma_e = \frac{4\pi R_0^{\,2}}{(\omega_1^{\,2}/\omega^2-1)^2+\delta_{\mathrm{AW}}^{\;2}}\frac{\delta_{\mathrm{Medwin}}}{\delta_{\mathrm{Medwin}}-2\beta_0/\omega}. \tag{66}$$

The confusion is eliminated by expressing the cross-sections in terms of β_0 (and ε) instead of δ, i.e., using Eq. (43) for the scattering cross-section, with ω_0 given by Eq. (23), and

$$\sigma_e = \sigma_s \frac{2\beta_0/\omega}{\varepsilon}\left(1+\frac{\omega}{2\beta_0}\varepsilon+\varepsilon^2\right) \tag{67}$$

for the extinction term.

V. EXAMPLE APPLICATIONS

In many circumstances, the contribution from damping due to radiation losses (without which the various scattering models described in Sec. III are in agreement) is small relative to thermal or viscous damping, so the magnitude of any error produced by the choice of an incorrect model is small. The purpose of this section is to discuss the conditions for which the radiation damping might be large enough to cause a significant effect, including numerical examples from a wide range of applications. Since the effect increases as the bulk modulus of the gas becomes no longer insignificant compared to that of the surrounding (possibly bubbly) liquid, these examples cover not only the acoustic monitoring of domestic bubbly products with high void fractions but also of bubbly liquids in extreme conditions (e.g., in coolant, fuel lines, or engineering for deep-ocean and extraterrestrial environments).

A. When are radiation losses large?

The examples considered for Figs. 2 and 3 involve acoustic radiation but no other form of damping. The value of ε^2 at resonance is proportional to the ratio of the bulk modulus of the gas bubble B_{bubble} to that of the surrounding liquid B_{medium}. Because of the relatively low pressure at the sea surface, the underwater acoustics literature is concerned mostly with damping dominated either by thermal conduction (in the case of large bubbles) or viscosity (small ones). In these circumstances the compressibility of the gas bubble is far greater than that of the surrounding liquid, leading to a strong resonance with low radiation loss (small ε). The ratio $B_{\mathrm{bubble}}/B_{\mathrm{medium}}$ will increase if B_{medium} is reduced. For example, if the medium surrounding the bubble in question is itself a bubbly liquid, the medium becomes more compressible than the bubble-free liquid. Examples are found in ship wakes, white caps caused by breaking waves, foams, bubble clouds generated by therapeutic ultrasound, sparging, or as found in the production of metals, pharmaceuticals, foodstuffs, or domestic products, for which void fractions can exceed 1%.[39,40] The ratio will also increase as B_{bubble} increases with increasing static pressure. If the depth of a bubble in the ocean is increased to a few hundred metres, the radiation damping can be dominant, as in the case of a fish bladder,[9] the lung of a deep diving whale or a methane vent at the seabed[41] (noting the additional complication of hydrate formation). Most models of bubble resonance assume that a bubble-free liquid surrounds the bubble in question, and to get large absolute effects (ε^2 of order 0.1) under such circumstances, for a bubble of air in water the static pressure needs to increase to several hundred megapascals, which is not achievable in oceans on earth. Even if such a high static pressure were to exist, the air inside the bubble would liquefy unless the temperature were also increased. For this reason the acoustics of bubbles at high temperature and pressure in a volcano are considered,[42,43] since their presence might influence or indicate eruptions and outgassing hazard.[44–48] Figure 4 illustrates the presence of a high void fraction in a sample of volcanic rock.

FIG. 4. (Color online) Photograph (by TGL) of bubbles in set lava at Timanfaya, Lanzarote. The image is 117 mm wide.

Below some examples are considered. While in some earlier sections of the paper, non-acoustic forms of damping were neglected for clarity, it is important to include these in the quantitative calculations of Sec. V B.

B. Numerical examples including non-acoustic damping ($\beta_0 \neq 0$)

Figure 5 shows σ_s plotted vs frequency for WM, AW, and Eq. (43), using numerical values of ε_0=0.3 and $2\beta_0/\omega_0$=0.3. Also plotted (vertical dashed lines) are the resonance frequencies associated with each model, as predicted by Eqs. (47)–(49). Figure 5 shows that if the values of ε_0 and β_0 are realized, significant differences arise not just between the WM and AW models but also between both of

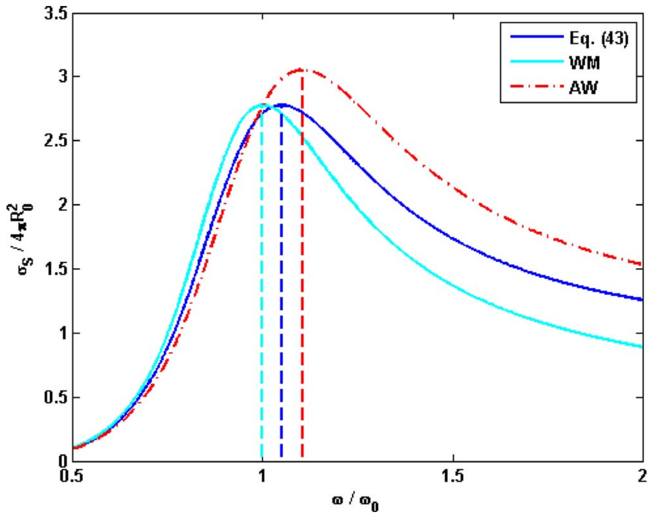

FIG. 5. (Color online) Theoretical pressure response (normalized scattering cross-section) vs dimensionless frequency ω/ω_0 calculated using WM [Eq. (45)], AW [Eq. (46)] and Eq. (43). Damping coefficients are ε_0=0.3 and $2\beta_0/\omega_0$=0.3.

these and Eq. (43). Taking Eq. (47) as a reference on the grounds that the derivation of Eq. (43) makes fewest approximations of the three models considered, it can be seen that neither AW nor WM are wholly accurate: WM underestimates and AW overestimates the resonance frequency [see Eqs. (48) and (49)] by $\varepsilon_0{}^2/2(1-2\beta_0{}^2/\omega_0{}^2)$ and $\varepsilon_0\beta_0/\omega_0$, respectively. The graph applies to any negligible density gas in any liquid.

There are many combinations of temperature and pressure that can give rise to the chosen input values of ε_0 and β_0. To illustrate the diversity, the following generic scenarios are considered:

(1) Case A: air bubbles at atmospheric pressure (e.g., in ship wakes and breaking waves,[49] foodstuff,[39] and cement paste[40]) and the multiphase reactors used in chemical, biochemical, environmental, pharmaceutical, or petrochemical industries.[50]
(2) Case B: methane bubbles in seawater at a depth of order 1000 m (notwithstanding the formation of hydrates at this depth)[41,51,52] (or deep water blowout[53]).
(3) Case C: carbon dioxide bubbles at high temperature and pressure in a volcano.[44–47,54–56]
(4) Case D: nitrogen bubbles in ethane lake on Titan.[57,58]
(5) Case E: helium bubbles in liquid mercury (neutron spallation target).[59–64]

For these gas-liquid mixtures the following notation is introduced (see Table I):

(a) subscript g denotes properties of the gas (e.g., ρ_g for the gas density),
(b) subscript w denotes properties of the host liquid (e.g., c_w for speed of sound in the bubble-free liquid), and
(c) subscript m denotes properties of the gas-liquid mixture (e.g., B_m for its bulk modulus).

Figure 5 is applicable to each of these scenarios. The properties of the gas-liquid mixture are specified by means of the gas bulk modulus B_g, mixture density ρ_m, specific heat ratio γ of the gas, and temperature T. These four parameters may be chosen freely for any given value of ε_0 and β_0/ω_0. Once chosen, the first two of them (B_g and ρ_m) determine the bulk modulus B_m

$$B_m = \frac{3B_g}{\varepsilon_0{}^2} \tag{68}$$

and the sound speed c_m of the mixture (column 6)

$$c_m = \sqrt{\frac{B_m}{\rho_m}}. \tag{69}$$

The bubble radius R_0 can take any value. Once chosen, its value determines the undamped natural frequency (column 7)

$$\omega_0 = R_0{}^{-1}\sqrt{\frac{3B_g}{\rho_m}} \tag{70}$$

and "equivalent" viscosity, which is defined as (column 8)

$$\mu_{\mathrm{eq}} \equiv R_0\sqrt{3B_g\rho_m}\frac{\beta_0}{2\omega_0}. \tag{71}$$

Thus, μ_{eq} is the shear viscosity that would be required, if viscosity were the only non-acoustic damping mechanism, to achieve the non-acoustic damping factor β_0. For example, a bubble radius of 1 mm gives a resonance frequency ($\omega_0/2\pi$) of between 2 kHz (case C) and 32 kHz (case E) and equivalent viscosity between 1.5 Pa s (case A) and 39 Pa s (case C).

The gas density corresponding to these conditions is shown in the last column [$(\rho_g)_{\mathrm{ad}}$, calculated from B_g assuming adiabatic pulsations]

TABLE I. Defining parameters (in bold) for cases A (air bubbles in wake), B (methane vent), C (carbon dioxide bubbles in molten lava), D (nitrogen bubbles in ethane lake on Titan), and E (helium bubbles in mercury spallation target). Parameters in remaining columns are calculated using the expressions given in the text.

Case	B_g (MPa)	ρ_m (kg m^{-3})	γ	T (K)	c_m (m s^{-1})	$\omega_0 R_0$ (m s^{-1})	$\mu_{\mathrm{eq}}R_0^{-1}$ (Pa s mm^{-1})	$(\rho_g)_{\mathrm{ad}}$ (kg m^{-3})
A	**0.14**	**1 000**	**7/5**	**280**	68.3	20.5	1.54	1.25
B	**10**	**1 000**	**4/3**	**280**	577	173	13.0	51.9
C	**35**	**2 600**	**4/3**	**1470**	670	201	39.2	95.2
D	**0.21**	**630**	**7/5**	**95**	38.9	31.6	1.49	5.36
E	**0.60**	**13 200**	**5/3**	**300**	105	11.7	11.6	0.581

$$(\rho_g)_{\text{ad}} = \frac{m_g B_g}{\gamma k_B T}. \tag{72}$$

Together with the information from the table, this value can be used to estimate the required void fraction using

$$U = \frac{\rho_w - \rho_m}{\rho_w - \rho_g}, \tag{73}$$

provided that the bubble-free liquid density ρ_w is known. To do so, Wood's equation[65] is used in the form (accepting the use of this low-frequency approximation for the purposes of this first-order calculation)

$$\left(\frac{1}{B_g} - \frac{1}{B_m}\right)\rho_w^{\,2} + \left(\frac{\rho_g}{B_m} - \frac{\rho_m}{B_g}\right)\rho_w + \frac{\rho_m - \rho_g}{c_w^{\,2}} = 0. \tag{74}$$

Using this equation it is found that the required void fraction is approximately 3% for all cases considered, even though the static pressure (as given by B_g/γ) varies by more than two orders of magnitude. The physical reason for this result is that, unless the void fraction is very low, increasing the pressure increases the bulk modulus of the gas-liquid mixture surrounding the bubble in question almost as much as it does that of the gas bubble. This can be seen more clearly by writing Eq. (73) in the form

$$U = \frac{\varepsilon_0^{\,2}/3 - B_g/B_w}{1 - B_g/B_w}. \tag{75}$$

For the cases considered, B_g is small compared with B_w, from which it follows that $U \approx \varepsilon_0^{\,2}/3 = 0.03$. The difficulty of applying Wood's equation if the bubbles are at or near resonance is recognized,[66,67] but the above simple analysis suggests that large values of ε_0 are unlikely to be achieved by high pressure alone.

VI. SUMMARY AND CONCLUSIONS

The dimensionless damping coefficient δ introduced by Wildt[7] and developed further by Medwin[2] are considered. Particular attention is paid to the role of radiation damping in determining the through-resonance frequency dependence of the scattering cross-section σ_s of a single spherical gas bubble in terms of the parameter ε, defined as the product of acoustic wave number and bubble radius. Specific conclusions are as follows

(1) Published theoretical results of Andreeva[9] and Weston[10] for σ_s are not consistent with those of Wildt and Medwin. The AW model, which has not been used in open literature for more than 40 years, is correct to order ε^2 in the denominator of σ_s. The WM model, which is in widespread use, is missing a term of this order and thus requires a leading order correction to the damping term.

(2) A generalization of Weston's derivation is used to obtain a new expression for σ_s [Eq. (25)], which simplifies to the AW model if non-acoustic damping is neglected. The same equation is then derived by application of Euler's equation to the input impedance obtained from Prosperetti's equation of motion, leading to Eq. (43). This second approach leads also to a new equation for the extinction cross-section σ_e [Eq. (67)].

(3) The magnitude of the effect, as measured by the difference in resonance frequency between the different scattering models, though often small, can become significant in some realistic conditions. It is of order 10% if ε and $2\beta_0/\omega$ are both equal to 0.3 at resonance (and is proportional to ε^2). This requires either a bubble under very high pressure (comparable with the bulk modulus of the surrounding liquid) or a bubble in a highly compressible liquid. Possibilities explored include a ship wake at atmospheric pressure, methane vents under pressure at the seabed, carbon dioxide bubbles in molten lava and helium bubbles in a neutron spallation target. For all cases considered, the required void fraction according to Wood's equation is close to 3% (i.e., $\varepsilon_0^{\,2}/3$.)

(4) The zeroth order term from Anderson's expansion for the scattering cross-section of a fluid sphere of arbitrary radius and density is simplified and used to confirm the accuracy of the AW model for the case with gas density and non-acoustic damping coefficient both negligible.

(5) Three different definitions of δ are considered, denoted δ_{AW} [defined by Eq. (26)], δ_{Wildt} [Eq. (28)], and δ_{Medwin} [Eq. (39)]. Of these, δ_{AW} is required in Eq. (1) to obtain the correct frequency dependence for σ_s, while either δ_{Wildt} or δ_{Medwin} (and *not* δ_{AW}) must be used in Eq. (65) for σ_e. Unless correction terms are applied to the currently accepted equations for σ_s and σ_e, there is no single definition of δ that gives the correct result for both cross-sections.

(6) In situations for which acoustic radiation is the main form of damping (e.g., in water under high static pressure), the WM model underestimates the resonance frequency. Its use to infer the bubble radius from an acoustical measurement would therefore lead to a systematic bias.

ACKNOWLEDGMENTS

One of the authors (M.A.A.) acknowledges formative discussions with Dr. D. E. Weston concerning the interaction of sound with fish swimbladders. He regrets now that none of these were about the frequency dependence of radiation damping. He also thanks Dr. A. J. Robins for his encouragement to pursue the discrepancy between the WM and AW models of radiation damping. Constructive comments by two anonymous reviewers helped us improve the final manuscript. This work was sponsored in part by the Defence Research and Development Department of the Netherlands Ministry of Defence (M.A.A) and in part by the UK Engineering and Physical Sciences Research Council (EPSRC), the UK Natural Environment Research Council (NERC), the UK Science and Technology Facilities Council (Rutherford Appleton Laboratory), and the Oak Ridge National Laboratories, Tennessee, Spallation Neutron Source (ORNL is managed by UT-Battelle, LLC, under contract DE-AC05-00OR22725 for the U.S. Department of Energy) (T.G.L).

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