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Analysis of Radial Oscillations of Gas Bubbles in Newtonian or Viscoelastic Mediums under Acoustic Excitation

by

Yuning Zhang

Thesis

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Declarations

I herewith declare that this thesis contains my own work conducted under the supervision of Prof. Shengcai Li. No part of the work in this thesis was previously submitted for a degree at another university.

Nomenclature

Roman letters (alphabetical order)

b_{th}	thermal dissipation coefficient
c	concentration of the gas in the liquid
c_g	speed of sound in the gas
c_l	speed of sound in the liquid
C_0	saturation concentration of the gas in the liquid
C_i	initial uniform concentration of the gas in the liquid and also the concentration of the gas in the liquid at infinity
C_s	concentration of the gas in the liquid at the bubble wall
D	diffusion constant
$D_{g,p}$	thermal diffusivity of the gas defined at constant pressure
$D_{g,v}$	thermal diffusivity of the gas defined at constant volume
G	shear modulus
h	enthalpy
H	enthalpy at the bubble wall
k	ratio of the liquid and the gas thermal conductivities

k_g	thermal conductivity of the gas
$k_{\mathbf{H}}$	Henry's constant
k_l	thermal conductivity of the liquid
Ma	Mach number of bubble wall
M_g	molecular weight of the gas in the bubble
p	non-dimensional deviation from equilibrium value of the pressure in the gas
$p(R_0, t)$	non-dimensional pressure deviation from equilibrium pressure at the gas side of bubble wall
P	local value of the pressure in the gas
P_0	ambient pressure
P_A	acoustic pressure amplitude
P_{A_1}	amplitude of external sound field of frequency ω_1
P_{A_2}	amplitude of external sound field of frequency ω_2
P_{in}	instantaneous pressure at the gas side of bubble wall
$P_{in,eq}$	equilibrium pressure at the gas side of bubble wall
P_{rad}	radiation pressure
P_T	threshold of acoustic pressure amplitude of rectified diffusion
r	radial coordinate
R	instantaneous bubble radius

$\dot{R}$	first derivative of the instantaneous bubble radius
$\ddot{R}$	second derivative of the instantaneous bubble radius
R_0	equilibrium bubble radius
R_g	universal gas constant
t	time
T_∞	ambient temperature in the liquid
T_g	local temperature in the gas
T_l	local temperature in the liquid
u	gas velocity
$\mathbf{u}$	velocity of the liquid
x	non-dimensional perturbation of the instantaneous bubble radius
$\dot{x}$	first time derivative of x
$\ddot{x}$	second time derivative of x

Greek letters (alphabetical order)

β_{ac}	acoustic damping constant
β_{th}	thermal damping constant
β_{tot}	total damping constant
β_{vis}	viscous damping constant

γ	ratio of specific heats of gas
γ_{rr}	strain
δ_{ac}	non-dimensional acoustic damping constant
δ_{th}	non-dimensional thermal damping constant
δ_{tot}	non-dimensional total damping constant
δ_{vis}	non-dimensional viscous damping constant
ε	non-dimensional amplitude of driving sound field
η	non-dimensional deviation from equilibrium value of the density in the gas
θ_g	non-dimensional deviation from equilibrium value of the temperature in the gas
θ_l	non-dimensional deviation from equilibrium values of the liquid temperature
κ	polytropic exponent
λ_g	wavelength in the gas
λ_l	wavelength in the liquid
μ_l	viscosity of the liquid
μ_{th}	effective thermal viscosity
ρ	local value of the density in the gas
ρ_g	density of the gas
ρ_l	density of the liquid

σ surface tension coefficient

τ_{rr} stress in the radial direction

φ a function related with the solution of bubble interior problem

Φ a simplified function related with the solution of bubble interior problem

ω angular frequency of the driving sound field with single frequency

ω_0 natural frequency

ω_1 one angular frequency of the driving sound field of dual frequency

ω_2 another angular frequency of the driving sound field of dual frequency

ω_r resonance frequency

Abstract

Acoustic cavitation plays an important role in a broad range of biomedical, chemical and oceanic engineering problems. For example, kidney stone can be crushed into the powder (being discharged naturally) by the acoustic cavitation generated by carefully controlled focused ultrasonic beams. Therefore, the prediction of generation of acoustic cavitation is essential to the aforementioned emerging non-invasive technique for kidney stone crushing. The objective of this PhD program is to study the generation of acoustic cavitation (e.g. through rectified mass diffusion across bubble interface) theoretically in the Newtonian fluids (e.g. water) or viscoelastic mediums (e.g. human soft tissue) under acoustic excitation of single or dual frequency. The compressibility and the viscosity of the liquid, heat and mass transfer across bubble-medium interface are all considered in this study.

During this PhD program, the established works in the literature on the above topic have been re-examined. More physically general formulas of natural frequency and damping of gas bubble oscillations in Newtonian or viscoelastic mediums has been derived and further employed for solving the problem of bubble growth under acoustic field (i.e. rectified mass diffusion).

For rectified mass diffusion of gas bubbles in Newtonian liquids, the predictions have been improved for high-frequency region of megahertz and above. Effects of medium viscoelasticity and dual-frequency acoustic excitation on rectified mass diffusion have also been studied. To facilitate the fast growth of bubble under acoustic field, dynamic-frequency and dual-frequency techniques have been proposed and demonstrated.

Key words: acoustic cavitation, gas bubbles, rectified mass diffusion, liquid compressibility, radial oscillations, damping mechanism, thermal effects, viscoelasticity

Chapter 1

Introduction and background

This chapter reviews relevant literature of acoustic cavitation and explains the objectives of this PhD research program.

Under the support of Engineering and Physical Sciences Research Council (EPSRC) Warwick Innovative Manufacturing Research Centre (WIMRC) phase II major project "Non-Surgical Cavitation-Effect Destruction of Kidney Stones", a non-surgical technique for crushing kidney stones based on cavitation induced by carefully controlled focused ultrasonic beam is being developed by the Cavitation Research Group (University of Warwick, UK). Comparing with current extracorporeal shock wave lithotripter (ECSWL), this technique can crush the stones into powder being discharged naturally; thus minimizes the negative effects.

The predictions of the bubble growth under ultrasonic waves (i.e. generation of acoustic cavitation) are essential to the above project. The frame work for solving this problem has been set up several decades ago. The previous investigations detailed in the literature were mainly focused on gas bubble dynamics under low- and single-frequency acoustic excitation (e.g. 20 kHz). However, in most of present and emerging applications, gas bubbles usually experience ultrasonic (e.g. megahertz and above) and multiple-frequency (two, three or more frequencies involved) excitation. Furthermore, micro- even nano-sized bubbles are often involved in applications; and such cases will further expand dramatically in the future. Therefore, a re-visit to those established works has been performed in this thesis with a focus on high-frequency regions and multiple-frequency acoustic excitation. Theoretical analysis on rectified mass diffusion has also been extended for gas bubble growth in viscoelastic mediums. Based on our analysis, two techniques (dynamic-frequency and dual-frequency approaches) for facilitating the fast growth of bubble under acoustic excitation have been proposed and demonstrated.

1.1 Radial oscillations of gas bubbles

This section briefly reviews published works on solving the radial oscillations of gas bubbles in liquids. For details, readers are referred to reviews by e.g. Plesset and Prosperetti (1977), Neppiras (1980), Prosperetti (1984a; 1984b), Feng and Leal (1997), Brenner et al. (2002) and Coussios and Roy (2008) or textbooks by e.g. Young (1989) and Brennen (1995). The basic equation for the motion of a spherical bubble in an infinite domain of liquid was mainly developed by Rayleigh (1917), Plesset (1949), Noltingk and Neppiras (1950), Neppiras and Noltingk (1951) and Poritsky (1952). This equation is generally called "Rayleigh-Plesset equation" for short (Brennen, 1995, Chap. 2.2). Shu (1952) examined Poritsky's results and proved that without surface tension, the collapse time of the bubble will be infinite if the non-dimensional viscosity is above a critical value.

In Rayleigh-Plesset equation, the effect of liquid compressibility is ignored. To account for liquid compressibility, radiation pressure has been proposed by Crandall (1926, p.120-124) and used by others (e.g. Chapman and Plesset, 1971; Prosperetti, 1977) for linear cases. The expressions of damping and natural frequencies of gas bubbles in liquids shown in Prosperetti (1977) have been widely cited in the literature till now. Equations of bubble motion with liquid compressibility considered have been intensively studied by researchers, mainly including Herring's equation (Herring, 1941; Trilling, 1952), Keller's equation (Keller and

Kolodner, 1956; Epstein and Keller, 1971; Keller and Miksis, 1980) and Gilmore’s equation (Gilmore, 1952). Prosperetti and Lezzi (1986) found that there exists one-parameter family of equations of bubble motion to the first order of bubble wall Mach number, which incorporates Herring’s equation and Keller’s equation as special cases. They pointed out that equations close to Keller’s form but written by enthalpy is the most accurate one in the test cases even better than Gilmore’s equation. Lezzi and Prosperetti (1987) further found that Gilmore’s equation is not accurate to the second order of bubble wall Mach number and its success reported in Hickling and Plesset (1964) is due to the use of enthalpy.

The damping mechanisms (viscous, thermal and acoustic damping) and natural frequency of bubbles are primary parameters for bubble dynamics, which can be obtained based on linearization of the equation of bubble motion. Many researchers contribute to the understandings of these topics including Minnaert (1933), Smith (1935), Pfriem (1940), Devin (1959), Plesset and Hsieh (1960), Eller (1970), Shima (1970), Chapman and Plesset (1971) and Prosperetti (1977). For thermal effects, the work by Prosperetti (1977) has laid down a systematic frame work by allowing non-uniform pressure in the bubbles and temperature variations in the gases and liquids. Crum (1983) has shown that the expressions related with thermal effects in Devin (1959) can be reduced from those in Prosperetti (1977). Experimental verification has been done by Crum (1983) through measuring the polytropic exponents for different gases. The expressions of damping constants and natural frequency obtained by Devin (1959) and Prosperetti (1977) have been highly cited by other researchers and are prevalent in the literature. Formulas in Prosperetti (1977) are usually used by researchers for validations e.g. Prosperetti (1991) and Preston et al. (2007).

Equations of bubble motion in viscoelastic mediums have also been studied by many researchers. For review of this topic, readers are referred to a recent book by Brujan (2010). Here, we only focused our discussions on a recent work by Yang and Church (2005). By using linear Voigt model as constitutive equation, a generalized Keller's equation was obtained by Yang and Church (2005). After linearization, the formulas for damping constants and natural frequency are obtained and compared with those in Newtonian fluids given by Prosperetti (1977).

1.2 Rectified mass diffusion

Rectified (mass) diffusion serves as an important mechanism for dissolution or growth of bubbles in acoustic field. Due to the interdisciplinary nature of this phenomenon, it attracts many researchers from various background e.g. acoustics, biomedical engineering and sonochemistry. For detailed reviews of this topic, readers are referred to Plesset and Prosperetti (1977), Crum (1984) and Ashokkumar et al. (2007). Rectified diffusion serves as a paramount factor in many physical processes, such as bubble sonoluminescence (Gaitan et al., 1992; Brenner et al., 1996; Roberts and Wu, 1998), formation of gas bubbles with visible size under ultrasound *in vivo* (Crum and Hansen, 1982b; Crum et al. 1987), evaluation of human diver and marine mammal safety (Crum and Mao, 1996; Houser et al., 2001; Crum et al., 2005; Ilinskii et al., 2008), generation of free radical in sonochemistry (Okada et al., 2009), enhancement of transdermal transport of a variety of different molecules (termed as "sonophoresis") (Lavon et al., 2007), ultrasonic degassing of hydrogen bubbles in molten aluminum alloys (Naji Meidani and Hasan, 2004), volcanic eruptions (Sturtevant et al., 1996 and Brodsky et al., 1998; Ichihara and Brodsky, 2006) and behavior of nanobubbles at solid-liquid interface under ultrasound irradiation (Brotchie and Zhang, 2011).

Generally, there are three effects involved in the process of rectified diffusion (Fyrillas and Szeri, 1994, p.381):

1. During bubble oscillations, the interface area of bubble for mass transportation changes.
2. During bubble oscillations, the volume of bubble, internal pressure and gas concentration in bubble change. According to the Henry's law, the concentration of dissolved gas at the bubble interfaces changes either.
3. The bubble volume oscillations also generate radial motion of liquids with velocity field which is inversely proportional to the square of radial coordinate. This velocity field will influence the gas diffusion through the convection terms in diffusion equation.

It has been shown that all three effects are necessary for an adequate description of this phenomenon.

This subject has been studied by researchers over half a century. This phenomenon was firstly observed by Harvey et al. (1944) and theoretically analyzed by Blake (1949) with the effect of area change included only. Hsieh and Plesset (1961) considerably improved the predictions by including the convection term in the diffusion equation comparing with the experiments (Strasberg, 1959; 1961). Eller and Flynn (1965) proposed an approach to account nonlinear pulsation of bubble under acoustic field based on thin-diffusion layer approximation initially used by Plesset and Zwick (1952). Eller and Flynn (1965) uncoupled the diffusion equation and the equation of bubble motion based on the fact that gas diffusion across bubble interface is very limited during a single cycle of bubble oscillation. Safar (1968) found that approaches of Hsieh and Plesset (1961) and Eller and Flynn (1965) are identical if the inertial terms are not neglected in Hsieh and Plesset (1961). Eller-Flynn's framework was adopted by many researchers, e.g. Eller (1969; 1972; 1975), Crum (1980), Crum and Hansen (1982a), Church (1988a; 1988b) and Roberts and Wu (1998). Fyrillas and Szeri (1994) proposed more physically general formulas for this problem. For details, readers are referred to Fyrillas and Szeri (1994; 1995; 1996). Crum (1980) reported a fairly good agreement between predictions and experiments except for the addition of surface active agents. The effect of surface active agent on rectified diffusion was further experimentally investigated by Lee et al. (2005), Leong et al. (2010) and Leong et al. (2011). Recently, the effect of temperature on rectified diffusion has been studied by Webb et al. (2010) and Webb et al. (2011).

For analytic approach, Crum and Hansen (1982a) derived first generalized equations by introducing the polytropic exponent and all damping terms (viscous, thermal and acoustic damping) obtained by Devin (1959) and Eller (1970). The analytic approach of rectified diffusion has been successfully employed to explain bubble growth under ultrasound *in vivo* (e.g. Crum and Hansen, 1982b; Crum *et al.*, 1987), evaluate human diver and marine mammal safety (Crum and Mao, 1996) and predict transdermal transport of molecules (Lavon et al., 2007). Equations in Crum and Hansen (1982a) have been widely cited in reviews (e.g. Crum, 1984) and books (e.g., Leighton, 1994, Chap. 4.4.3; Brennen, 1995, Chap. 4.9).

All the above works deal with the rectified diffusion of gas bubbles in Newtonian fluids. According to the literature review, no work has been done for rectified diffusion of gas bubbles in non-Newtonian fluids (e.g. viscoelastic mediums).

1.3 Gas bubble dynamics under acoustic field of multiple frequencies

The effect of acoustic cavitation can be enhanced by multiple-frequency acoustic excitation. Previously, this multiple-frequency technique has been successfully applied for bubble size measurement (Newhouse and Shankar, 1984), fluid pressure measurement (Shankar et al., 1986), contrast imaging (Wu and Tsao, 2003; Wu et al., 2005), optimization of acoustic scattering (Wyczalkowski and Szeri, 2003), enhancement of cavitation effect in sonochemistry (Feng et al., 2002; Avvaru and Pandit, 2008) and enhancement of bubble sonoluminescence (Holzfuss et al., 1998; Ketterling and Apfel, 2000; Hargreaves and Matula, 2000; Krefting et al., 2002; Kanthale et al., 2008).

For the theoretical studies, analytic solutions for the radial oscillations of gas bubbles in liquids under dual-frequency acoustic excitation have been obtained by Newhouse and Shankar (1984) with liquid compressibility excluded. For numerical simulations, readers are referred to the recent work by Kanthale et al. (2007). According to the literature review, the mass transfer across bubble-medium interface during the radial oscillations of gas bubbles in liquids is currently ignored by published works.

1.4 Objectives of the thesis

In this thesis, physically general formulas for predictions of generation of acoustic cavitation in Newtonian or viscoelastic mediums under acoustic excitation have been derived. Published works involving this topic have been re-examined. Dynamic-frequency and dual-frequency techniques have been proposed and demonstrated for facilitating the fast growth of acoustic bubbles. The whole thesis is organized as follows.

Chapter 2: More physically general formulas for damping mechanisms and thermal effects of radial oscillations of gas bubbles in the liquids are proposed. The valid regions for formulas shown in the literature have been investigated and defined. Formulas in the literature are compared and discussed with ours through several demonstrating examples.

Chapter 3: Analytical solutions for the rectified mass diffusion during radial oscillations of gas bubbles in the Newtonian fluids are derived for more general cases and compared with those in the literature. Based on above formulas derived by us, the advantage of dynamic-frequency approach is shown by comparing with constant-frequency approach.

Chapter 4: More physically general formulas of damping and natural frequency for the radial oscillations of gas bubbles in viscoelastic mediums are derived and compared with previous works. Analytical solutions for the rectified mass diffusion during radial oscillations of gas bubbles in the viscoelastic mediums are also derived. The predictions based our formulas are compared with experimental data and previous predictions in the literature based on formulas of rectified mass diffusion in Newtonian fluids.

Chapter 5: Analytical solutions for the rectified mass diffusion during radial oscillations of gas bubbles in the liquids under acoustic field of dual frequency are derived. Some demonstrating examples are given and compared with those under acoustic field of single frequency, showing the advantage of the dual-frequency technique.

Chapter 6: The contributions obtained from this PhD programme to the further understanding of the subjects are summarized and possible future works are outlined.

Chapter 2

Damping mechanisms and thermal effects for radial oscillations of gas bubbles in liquids

In this chapter, the damping mechanisms and thermal effects for radial oscillations of gas bubbles in liquids are investigated. Parts of Chap. 2.1 and 2.3 have been published in Zhang and Li (2012). Parts of Chap. 2.2.5 and 2.2.7 have been published in Zhang and Li (2010a). The primary assumptions employed in this chapter are

1. The gas and liquid are both Newtonian fluids.
2. The bubble is spherical.
3. Only radial motion of bubble is considered.

If not specified, the constants shown in Appendix A are used for calculations in this chapter.

2.1 Natural frequency and damping for radial oscillations of gas bubbles in liquids

In this section, expressions for damping constants and natural frequency of linear radial oscillations of gas bubbles in liquids will be derived. Prosperetti and Lezzi (1986) proposed a one-parameter family of equations of bubble motion to the first order of bubble-wall Mach number (the ratio between bubble wall velocity and speed of sound), which treats Keller's equation (Keller and Kolodner, 1956; Keller and Miksis, 1980) as a special case. For small oscillations considered here, expressions of damping constant and natural frequency will be the same based on any equation of bubble motion which falls into this one-parameter family. For the proof of this, readers are referred to Appendix B. Here, Keller's equation is chosen as the equation of bubble motion and a more appropriate expression of Keller's equation is used (Prosperetti and Lezzi, 1986, p.466). Comparing with derivations shown in literature (e.g. Devin, 1959; Prosperetti, 1977), a more physically general derivations are given here. Comments on other previous works and comparisons with formulas in literature will be given in Chap. 2.3. Here, oscillations of spherical gas bubbles with small amplitude in infinite liquids are considered. Keller's equation (Keller and Kolodner, 1956; Keller and Miksis, 1980; Prosperetti and Lezzi, 1986) can be written as

$$\left(1-\frac{\dot{R}}{c_l}\right)R\ddot{R}+\frac{3}{2}\left(1-\frac{\dot{R}}{3c_l}\right)\dot{R}^2=\left(1+\frac{\dot{R}}{c_l}\right)\frac{p_{ext}(R,t)-p_s(t)}{\rho_l}$$

$$+\frac{R}{\rho_l c_l}\frac{d[p_{ext}(R,t)-p_s(t)]}{dt}, \quad (2.1)$$

where

$$p_{ext}(R,t)=P_{in}-\frac{2\sigma}{R}-\frac{4\mu_l}{R}\dot{R}; \quad (2.2)$$

$$p_s(t)=P_0\left[1+\varepsilon e^{i\omega t}\right]. \quad (2.3)$$

Here, R is the instantaneous bubble radius; the overdot denotes the time derivatives; c_l is the undisturbed speed of sound in the liquid (Prosperetti and Lezzi, 1986); ρ_l is the density of the liquid; *t* is the time; P_{in} is the instantaneous pressure at the gas side of bubble wall; σ is the surface tension coefficient; μ_l is the viscosity of the liquid; P_0 is the ambient pressure; ε is the non-dimensional amplitude of driving sound field; ω is the angular frequency of driving sound field.

For comparisons, framework in Prosperetti (1977) for thermal effects will be followed but the equation of bubble motion based on radiation pressure (P_{rad}) being replaced by Keller's equation. Notations in Prosperetti (1977) will also be retained the same such as

$$R=R_0(1+x),$$

$$P_{in}=P_{in,eq}+P_0 p(R_0,t),$$

$$P_{in} = P_{in,eq}\left(R_0 / R\right)^{3\kappa} - 4\mu_{th}\dot{x},$$

$$P_{in,eq} = P_0 + \frac{2\sigma}{R_0}.$$

Here, R_0 is the equilibrium bubble radius; x is the non-dimensional perturbation of instantaneous bubble radius, which is of the same order of ε ; $p(R_0,t)$ is the non-dimensional deviation from the equilibrium pressure at the gas side of bubble wall; $P_{in,eq}$ is the equilibrium pressure at the gas side of bubble wall; μ_{th} is the effective thermal viscosity; κ is the polytropic exponent. For small oscillations, ε (or x) <<1 and we omit the 2nd and higher orders of ε (or x). Based on above formulas, we obtain the following relation,

$$P_0 p(R_0,t) = P_{in,eq}\left[\left(R_0 / R\right)^{3\kappa} - 1\right] - 4\mu_{th}\dot{x} \approx -3\kappa P_{in,eq} x - 4\mu_{th}\dot{x}. \qquad (2.4)$$

Substituting above relation into Eq. (2.1) results in an inhomogeneous equation for a harmonic oscillator (the oscillating bubble being studied),

$$\ddot{x} + 2\beta_{tot}\dot{x} + \omega_0^2 x = -\alpha_0\varepsilon\left(1 + i\omega R_0 / c_l\right)e^{i\omega t} / M, \qquad (2.5)$$

where

$$\alpha_0 = \frac{P_0}{\rho_l R_0^2};$$

$$M = 1 + \frac{R_0}{c_l}\frac{4\left(\mu_l + \mu_{th}\right)}{\rho_l R_0^2}.$$

In Eq. (2.5), β_{tot} is the total damping constant; ω_0 is the natural frequency. The total damping constant (β_{tot}) is

$$\beta_{tot} = \beta_{vis} + \beta_{th} + \beta_{ac},$$

where

$$\beta_{vis} = 2\mu_l / \rho_l R_0^2 M \; ; \tag{2.6}$$

$$\beta_{th} = 2\mu_{th} / \rho_l R_0^2 M \; ; \tag{2.7}$$

$$\beta_{ac} = \frac{R_0}{2c_l}\omega_0^2 , \tag{2.8}$$

representing the viscous, thermal and acoustic damping constants respectively. The acoustic damping constant (Eq. (2.8)) derived by us is different with expressions derived by others. For details, readers are referred to Chap. 2.3. The natural frequency of the harmonic oscillator is

$$\omega_0^2 = \frac{1}{M}\left(\frac{3\kappa P_{in,eq}}{\rho_l R_0^2} - \frac{2\sigma}{\rho_l R_0^3} \right). \tag{2.9}$$

Note that here μ_l, μ_{th} and liquid compressibility make contributions to natural frequency through M in Eq. (2.9) though these contributions are relatively small[1].

Forced oscillations of bubbles will enter stationary oscillations once the transient term (the solution for the corresponding homogenous equation) in the solution approaches to zero. For details, readers are referred to Appendix C. For most cases of bubble oscillations, the contribution from the transient term dies quickly. For example, for a bubble of radius 1 μm driven by an external force at angular frequency $\omega = 10^7$ sec^{-1} with

[1] Taking an air bubble oscillating in water as an example, M varies between 1 and 1.03 if 10^{-2}m$\geq R_0 \geq 10^{-7}$m and 10^8 sec$^{-1} \geq \omega \geq 10^4$ sec^{-1}.

non-dimensional amplitude $\varepsilon = 0.1$, the ratio of amplitudes between the transient term and the stationary term (i.e. the particular integral for the inhomogeneous equation) reduces down to 0.1 at t=1.15 μs (i.e. t=1.83T, T is the period of acoustic field). Therefore, transient term only plays an important role during the initial several cycles of gas bubble oscillations. For the rectified mass diffusion, the large timescale effects after thousands of bubble oscillations are of interest. Hence, the oscillation to be analyzed can be simplified as a stationary one such as appearing in the previous studies (e.g. Prosperetti, 1977). Here, for the stationary forced oscillations driven by sound field of single frequency (ω), μ_{th} in Eq. (2.7) and κ in Eq. (2.9) can be determined by solving the bubble interior following the approach in Prosperetti (1977). For details, readers are referred to Chap. 2.2. Though Keller's equation has been used previously to replace the radiation pressure as used by Prosperetti (1977), the resultant expressions for μ_{th} and κ are the same[1] (Prosperetti, 1977),

$$\mu_{th} = \frac{1}{4}\omega\rho_g R_0^2 \,\mathrm{Im}\,\varphi; \tag{2.10}$$

$$\kappa = \frac{1}{3}\left(\omega^2\rho_g R_0^2 / P_{in,eq}\right)\mathrm{Re}\,\varphi. \tag{2.11}$$

Here, ρ_g is the gas density; φ is a function relating to the solution of bubble interior problem. For the expressions of φ, readers are referred to Chap. 2.2.2.

[1] Noticing that expressions of μ_{th} and κ in Prosperetti (1977) were determined by comparing solutions of two methods: one based on polytropic model with effective thermal damping; the other based on solution of bubble interior.

For a wide range of parameters, $M \approx 1$ (referring to note 1 in p.17). Thus, Eq. (2.5) reduces to

$$\ddot{x} + 2\beta_{tot}\dot{x} + \omega_0^2 x = -\alpha_0 \varepsilon \left(1 + i\omega R_0 / c_l\right) e^{i\omega t}. \tag{2.12}$$

Here ω_0 and β_{tot} reduce to

$$\omega_0^2 = \frac{3\kappa P_{in,eq}}{\rho_l R_0^2} - \frac{2\sigma}{\rho_l R_0^3}; \tag{2.13}$$

$$\beta_{tot} = \frac{2\left(\mu_l + \mu_{th}\right)}{\rho_l R_0^2} + \frac{R_0}{2c_l}\omega_0^2. \tag{2.14}$$

This expression for natural frequency is well cited (e.g. Plesset and Prosperetti, 1977), in which the contribution of compressibility (through M) disappears. However, the contribution of compressibility to the total damping constant β_{tot} does not disappear completely since the β_{ac} term still remains.

Furthermore, if $c_l \to \infty$ (corresponding to $\frac{\dot{R}}{c_l}$ and $\frac{R_0}{c_l} \to 0$), i.e. incompressible cases, Eq. (2.1) reduces to the Rayleigh-Plesset equation. And Eq. (2.12) reduces to

$$\ddot{x} + \left[\frac{4(\mu_l + \mu_{th})}{\rho_l R_0^2}\right]\dot{x} + \omega_0^2 x = -\alpha_0 \varepsilon e^{i\omega t}, \tag{2.15}$$

identical to the cases where the Rayleigh-Plesset equation replaces the radiation pressure in Prosperetti (1977). Naturally, the acoustic damping disappears since liquid compressibility is not considered in Rayleigh-Plesset equation.

Here, formulas for free gas bubble oscillations can also be reduced from above derivations. Let μ_l =0, μ_{th} =0, ε =0 and σ =0 then Eq. (2.12) reduces to

$$\ddot{x}+\frac{3\kappa P_0}{\rho_l R_0 c_l}\dot{x}+\frac{3\kappa P_0}{\rho_l R_0^2}x=0\,. \tag{2.16}$$

which is the Eq. (28) in Keller and Kolodner (1956). Bubble radius as a function of time predicted by Eq. (2.16) agrees well with experiments (Keller and Kolodner, 1956, Fig.7). Keller and Miksis (1980) re-evaluated the work in Keller and Kolodner (1956) by including liquid viscosity but found that its influence is not distinguishable.

If μ_{th} =0 and ε =0, Eq. (2.5) reduces to

$$\left[1+\frac{R_0}{c_l}\frac{4\mu_l}{\rho_l R_0^2}\right]\ddot{x}+\left[\frac{4\mu_l}{\rho_l R_0^2}+\frac{R_0}{c_l}\omega_0^2\right]\dot{x}+\omega_0^2 x=0\,, \tag{2.17}$$

which is identical with Eq. (13) in Shima (1970) based on Gilmore's equation (Gilmore, 1952). For details, readers are referred to Appendix D. This is not surprising because during the linearization process of Gilmore's equation, the variation of sound speed will reduce to a constant (c_l) and a second-order term of sound speed in Gilmore's equation has been neglected. Therefore, formulas for damping based on Gilmore's equation are identical with those obtained using Keller's equation.

Comparing the expressions of acoustic damping for the forced (i.e. Eqs. (2.5) and (2.8)) and the free (i.e. Eq. (2.17)) gas bubble oscillations, one can find that they are identical. This finding is different from previous understanding (referring to Chap. 2.3).

2.2 Thermal effects

For the closure of the model, solution of φ in the expressions of μ_{th} and κ (Eqs. (2.10) and (2.11)) relating to thermal effects will be given and discussed in this section.

2.2.1 Equations for gas bubbles in liquids

In order to solve φ in Eqs. (2.10) and (2.11), Prosperetti (1977) further considered following equations for bubble interior, which will be introduced in this section. The deviations of quantities (e.g. density, pressure and temperature) from their equilibrium values are assumed to be small and we denote

$$\rho = \rho_g \left(1+\eta\right);$$

$$P = P_0\left(1 + 2\sigma / R_0 P_0 + p\right);$$

$$T_g = T_\infty\left(1 + \theta_g\right);$$

$$T_l = T_\infty\left(1 + \theta_l\right).$$

Here, ρ, P and T_g are the local values of density, pressure and temperature in the gas respectively; T_∞ is the ambient temperature; η, p and θ_g are the non-dimensional deviations from the equilibrium values of density, pressure and temperature in the gas respectively; T_l is the local temperature in the liquid; θ_l is the non-dimensional deviation from the equilibrium value of the liquid temperature. The equation of state of the gas is

$$P = \left(R_g / M_g\right)\rho T_g .$$

Here, R_g is the universal gas constant; M_g is the molecular weight of the gas in the bubble. Then, we obtain

$$p = \left(1 + 2\sigma / R_0 P_0\right)\left(\theta_g + \eta\right).$$

The equations of conservation of mass and momentum in the gas are

$$\frac{\partial \eta}{\partial t} + \frac{1}{r^2}\frac{\partial\left(r^2 u\right)}{\partial r} = 0\,; \tag{2.18}$$

$$\frac{\partial u}{\partial t} + \frac{P_0}{\rho_g}\frac{\partial p}{\partial r} = 0\,. \tag{2.19}$$

Here, u is the gas velocity; r is the radial coordinate. The equations of conservation of energy in the gas and the liquid are

$$\frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2\frac{\partial \theta_g}{\partial r}\right) + \frac{\left(P_0 + 2\sigma / R_0\right)}{k_g T_\infty}\frac{\partial \eta}{\partial t} = \frac{1}{D_{g.v}}\frac{\partial \theta_g}{\partial t}\,; \tag{2.20}$$

$$\frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2\frac{\partial\theta_l}{\partial r}\right)=\frac{1}{D_l}\frac{\partial\theta_l}{\partial t}\,. \tag{2.21}$$

Here, k_g is the gas thermal conductivity; $D_{g,v}$ is the thermal diffusivity of the gas defined at constant volume; D_l is the thermal diffusivity of the liquid. The boundary conditions can be given as follows:

Continuity of temperature at gas-liquid interface ($r = R_0$)

$$\theta_l\Big|_{(r=R_0,t)}=\theta_g\Big|_{(r=R_0,t)}\,. \tag{2.22}$$

Continuity of heat flux at gas-liquid interface ($r = R_0$)

$$-k_l\frac{\partial\theta_l}{\partial t}\Big|_{(r=R_0,t)}=-k_g\frac{\partial\theta_g}{\partial t}\Big|_{(r=R_0,t)}\,. \tag{2.23}$$

Here, k_l is the thermal conductivity of the liquid.

Continuity of velocity at gas-liquid interface ($r = R_0$)

$$u\Big|_{(r=R_0,t)}=dR/dt\,. \tag{2.24}$$

2.2.2 Prosperetti's analysis

In this section, the analysis by Prosperetti (1977) for the solution of equations of gas bubbles in liquids (Eqs. (2.18-2.21)) with boundary conditions (Eqs. (2.22-2.24)) is introduced. The main assumption used by Prosperetti (1977) is that the gas bubble is spherical. The validities of this assumption will be further discussed in Chap. 2.2.7. Non-uniform pressure in the gas bubbles, temperature variations in the liquids and the gas bubbles are all allowed. The equations of mass, momentum and energy conservation equations in the gas and the energy conservation equation in the liquid were solved together with boundary conditions. Three non-dimensional parameters have been defined by Prosperetti (1977):

$$G_1 = M_g D_{g,v} \omega / \gamma R_g T_\infty ; \tag{2.25}$$

$$G_2 = \omega R_0^2 / D_{g,v} ; \tag{2.26}$$

$$G_3 = \omega R_0^2 / D_l . \tag{2.27}$$

Here, γ is the ratio of specific heats of the gas. G_1 reflects the ratio between mean free path and the wavelength in the gas (Prosperetti, 1977); G_2 reflects the ratio between bubble radius and thermal penetration depth (Prosperetti, 1977).

To close the model, expression of φ is required in Eqs. (2.10) and (2.11). Prosperetti (1977) determined the expression of φ by solving Eqs. (2.18-2.21) with boundary conditions (Eqs. (2.22-2.24)). Here, the solution of φ given by Prosperetti (1977) will be directly cited here. For details, readers are referred to Prosperetti (1977). Following Prosperetti (1977), φ in Eqs. (2.10) and (2.11) can be expressed by

$$\varphi = \frac{kf\left(\Gamma_2 - \Gamma_1\right) + \lambda_2 \Gamma_2 - \lambda_1 \Gamma_1}{kf\left(\lambda_2 \Gamma_1 - \lambda_1 \Gamma_2\right) - \lambda_1 \lambda_2 \left(\Gamma_2 - \Gamma_1\right)} , \tag{2.28}$$

where

$$\beta_{1,2} = \left(\frac{1}{2} \gamma G_2 \left\{ i - G_1 \pm \left[\left(i - G_1 \right)^2 + 4iG_1 / \gamma \right]^{1/2} \right\} \right)^{1/2} ; \tag{2.29}$$

$$\lambda_i = \beta_i \coth \beta_i - 1 \quad i\text{=}1,2\,; \tag{2.30}$$

$$\Gamma_{1,2} = i + G_1 \pm \left[\left(i - G_1 \right)^2 + 4iG_1 / \gamma \right]^{1/2}; \tag{2.31}$$

$$f = 1 + \left(1+i\right) \left(\frac{1}{2} G_3 \right)^{1/2}; \tag{2.32}$$

$$k = k_l / k_g\,. \tag{2.33}$$

Therefore, k is the ratio of the liquid and the gas thermal conductivities.

Using the definition of G_1 (Eq. (2.25)) and G_2 (Eq. (2.26)) and equations of state of gas ($P_{in,eq} = R_g \rho_g T_\infty / M_g$), μ_{th} and κ can also be expressed as

$$\mu_{th} = \frac{P_{in,eq} \gamma G_1 G_2}{4\omega} \operatorname{Im} \varphi\,; \tag{2.34}$$

$$\kappa = \frac{1}{3} \gamma G_1 G_2 \operatorname{Re} \varphi\,. \tag{2.35}$$

In order to simplify above expressions, Prosperetti (1977) further assumed that the angular frequency of acoustic field is limited (corresponding to G_1<<1 based on Eq. (2.25)). Thus Eqs. (2.29) and (2.31) become

$$\beta_1 = \left(1+i\right) \left(\frac{1}{2} \gamma G_2 \right)^{1/2} \left\{ 1 + \frac{1}{2} i \left[(\gamma - 1)/\gamma \right] G_1 + O(G_1^2) \right\}; \tag{2.36}$$

$$\beta_2 = \left(G_1 G_2 \right)^{1/2} \left\{ i + \frac{1}{2} \left[(\gamma - 1)/\gamma \right] G_1 + O(G_1^2) \right\}; \tag{2.37}$$

$$\Gamma_1 = 2\left(i + G_1/\gamma \right) + O(G_1^2)\,; \tag{2.38}$$

$$\Gamma_2 = 2\left[(\gamma - 1)/\gamma \right] G_1 + O(G_1^2)\,. \tag{2.39}$$

Based on the fact that the magnitude of quantity *kf* is a large number in most cases, Prosperetti (1977) further simplified the expression of φ (Eq. (2.28)) as follows

$$\varphi = \Theta\left[1+\left(kf\right)^{-1} E_1 + O\left(kf\right)^{-2}\right], \tag{2.40}$$

where

$$\Theta = \frac{\Gamma_1 - \Gamma_2}{\lambda_1\Gamma_2 - \lambda_2\Gamma_1}; \tag{2.41}$$

$$E_1 = \frac{\Gamma_1\Gamma_2}{\Gamma_1 - \Gamma_2}\frac{\left(\lambda_1 - \lambda_2\right)^2}{\lambda_1\Gamma_2 - \lambda_2\Gamma_1}. \tag{2.42}$$

If all $(kf)^{-1}$ term with the first order and above are neglected, Eq. (2.40) reduces to

$$\varphi \approx \Theta. \tag{2.43}$$

In section Ⅱ of Prosperetti (1977), κ values for the range of G_1=10^{-9} to 10^{-5} have been evaluated based on Eqs. (2.30), (2.36)-(2.39), (2.41) and (2.43). The thermal damping constant (β_{th}) for the range of ω=10^2 to 10^9 sec^{-1} (corresponding to G_1=2.46×10^{-8} to 2.46×10^{-1} for air bubbles) has also been evaluated in section Ⅲ of Prosperetti (1977) based on Eqs. (2.28), (2.30), (2.32)-(2.33) and (2.36)-(2.39).

2.2.3 Devin's analysis

In this section, analysis of thermal effects shown in Devin (1959) will be introduced. It should be noted that Devin's work on this topic is largely based on Pfriem (1940). The following assumptions were used by Devin (1959):

1. Liquid temperature adjacent the bubble interface does not change so the liquid behaves as a heat reservoir (Devin, 1959, p.1657). Therefore, the equation of energy conservation in liquids was not solved in Devin (1959).
2. The density and specific heats of the gas are regarded as constants (Devin, 1959, p.1657).
3. Pressure in the gas bubbles is uniform. Therefore, pressure in the gas is only a function of time rather than radial coordinate (Devin, 1959, p.1657).
4. The boundary conditions at the bubble interface and the bubble center are given as follows: at the center of the bubble, the changes of temperature must be finite and the gradient of change in temperature must be zero; at the bubble-liquid interface, the changes of temperature must be zero and the gradient of change in temperature must be finite (Devin, 1959, p.1658).
5. The oscillations of pressure, bubble volume and temperature are assumed to be small (Devin, 1959, p.1657).

Based on above assumptions, Devin (1959) obtained the following expressions for thermal effects[1],

$$\gamma G_1 G_2 \varphi \approx \Phi = 3\gamma / \left\{ 1 - 3(\gamma - 1) i \chi \left[(i/\chi)^{1/2} \coth (i/\chi)^{1/2} - 1 \right] \right\}, \quad (2.44)$$

where

$$\chi = D_{g,p} / \omega R_0^2 .$$

Here, $D_{g,p} = D_{g,v} / \gamma$ is the thermal diffusivity of the gas at constant pressure. Eq. (2.44) is the same as formulas in Pfriem (1940, Eq. (14b)), Devin (1959, Eq. (50)) and Prosperetti et al. (1988, Eq. (41)). Then, Eqs. (2.34) and (2.35) become

$$\mu_{th} = \frac{P_{in,eq}}{4\omega} \operatorname{Im} \Phi ; \quad (2.45)$$

$$\kappa = \frac{1}{3} \operatorname{Re} \Phi . \quad (2.46)$$

2.2.4 Influence of surface tension

Although the formula for damping constant derived by Devin (1959) and Prosperetti et al. (1988) is identical, non-dimensional thermal damping constant (δ_{th}) is usually used in the literature (Medwin, 1977; Medwin and Clay, 1998, Chap. 8.2), which was defined by Devin (1959) as,

$$\delta_{th} = 4\mu_{th}\omega / \rho_l R_0^2 \omega_0^2 = 2\beta_{th}\omega / \omega_0^2 . \quad (2.47)$$

Devin (1959) gave the expression of δ_{th} as follows,

$$\delta_{th} = \operatorname{Im} \Phi / \operatorname{Re} \Phi . \quad (2.48)$$

[1] A concise expression in Prosperetti et al. (1988) is cited here and in the following discussions.

However, we found that above expression do not account for surface tension. If surface tension included, Eq. (2.48) should be

$$\delta_{th} = \frac{\operatorname{Im}\Phi}{\operatorname{Re}\Phi - 2\sigma / R_0 P_{in,eq}}. \tag{2.49}$$

Eq. (2.49) can be directly obtained based on Eqs. (2.13), (2.45) and (2.46) following Prosperetti's framework (Prosperetti, 1977; Prosperetti et al., 1988) or alternatively based on Devin (1959) as shown in Appendix E. Comparing with Eq. (2.48) derived by Devin (1959), a surface tension term ($2\sigma / R_0 P_{in,eq}$) was shown in denominator of Eq. (2.49). The effect of surface tension on damping constants is emphasized here because many published papers did not notice the difference between Eq. (2.48) and Eq. (2.49), which includes review papers (e.g. Medwin, 1977) and textbooks (e.g. Leighton, 1994, Chap. 3.4; Medwin and Clay, 1998, Chap. 8.2). Figure 2.1 compares the predictions of non-dimensional thermal damping constant (δ_{th}) with or without surface tension (i.e. Eq. (2.49) and Eq. (2.48) respectively) for frequencies ω =10^5, 10^6 and 10^7 sec^{-1} respectively. The predictions of δ_{th} without surface tension are lower than those with surface tension. In the following sections, we will use Eq. (2.49) for predictions of δ_{th} if not specified.

We will further consider two specific cases of Eq. (2.49) for fixed frequencies (ω):

Case A. For large bubbles, $2\sigma / R_0 P_{in,eq} \approx 0$ and Eq. (2.49) reduces to Eq. (2.48). Devin (1959, Eq. (55)) found that for $\chi \le 0.08$ in Eq. (2.44), the error of predictions of δ_{th} can be controlled within nearly one percent by

$$\delta_{th} \approx \frac{1-\sqrt{2\chi}}{1+\sqrt{2/\chi}\,/\,3(\gamma-1)} \approx 3(\gamma-1)\sqrt{\chi/2}\,. \qquad (2.50)$$

Case B. For small bubbles, surface tension terms in Eq. (2.49) should be considered. For $\chi \ge 0.5$ in Eq. (2.44), the following simplified formula given by Devin (1959, Eq. (56)) to predict δ_{th} with error within one percent will be impaired,

$$\delta_{th} \approx \frac{\gamma-1}{15\gamma\chi}\,. \qquad (2.51)$$

Instead, we found that the error of the following simplified formula for predicting δ_{th} is within nearly one percent for $\chi \ge 0.5$ in Eq. (2.44),

$$\delta_{th} \approx \frac{\gamma-1}{15\gamma\chi\left(1-2\sigma/3R_0 P_{in,eq}\right)}\,. \qquad (2.52)$$

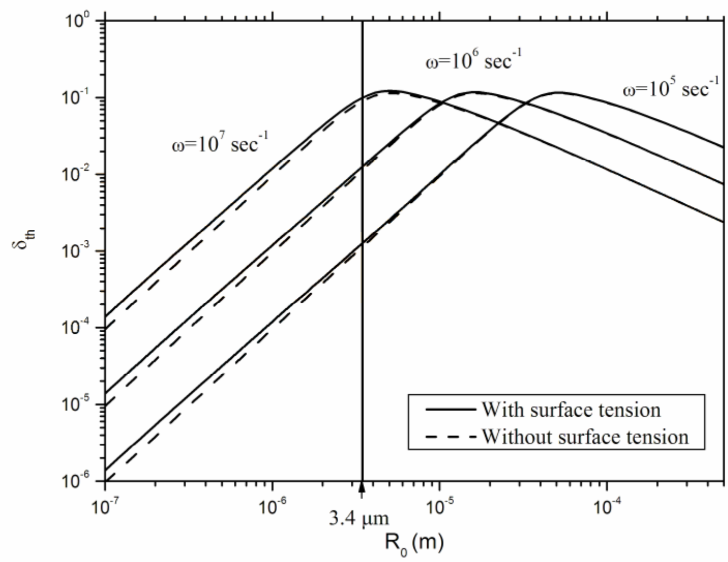

Figure 2.1 Comparison of non-dimensional thermal damping constant predicted with and without surface tension corrections. The marked line (R_0=3.4 μm) corresponds to error between Eq. (2.48) and Eq. (2.49) being 10%.

2.2.5 Re-visit to Prosperetti's analysis

Although Prosperetti (1977) gave the full solution for φ, the influence of approximation of G_1<<1 on the predictions was not shown. Furthermore, the predictions of polytropic exponent for G_1 more than 10^{-5} were not shown in Prosperetti (1977). Therefore, a re-visit to the approximation of G_1<<1 used by Prosperetti in Chap. 2.2.2 will be given in this section. According to Prosperetti (1977, p.24), a nonisothermal layer would become essential only for an extremely high frequency, e.g. in the order of

10^{14} sec^{-1} for water. Though the maximum frequency currently used, say in medical applications, is high, it is still not higher than 10^9 sec^{-1}. Therefore, the framework by Prosperetti (1977) is still followed here[1].

Our re-visit has been performed through comparing those given in Prosperetti (1977) using approximation G_1<<1 (Eqs. (2.36)-(2.39)) with the full solution (Eqs. (2.28)-(2.33)) also derived by Prosperetti (1977). For comparison, air bubbles in water have been used; and values for G_1 between 10^{-9} and 10^{-1} and G_1G_2 between 10^{-10} and 10^1 have been evaluated. In the following discussions, "Prosperetti (1977) (G_1<<1)" refers to the use of Eqs. (2.28), (2.30), (2.32)-(2.33) and (2.36)-(2.39) for predictions; "Prosperetti (1977)" refers to the use of Eqs. (2.28)-(2.33) for predictions.

From the re-visit, the influence of the approximation G_1<<1 on the values of κ and β_{th}, which are determined by the real and imaginary parts of φ respectively (referring to Eqs. (2.10) and (2.11)), are revealed as follows. For κ, only a little difference between "Prosperetti (1977) (G_1<<1)" and "Prosperetti (1977)" is found for G_1=10^{-1}, and no

[1] Eq. (35) and A_3 in the solution of the bubble interior as given in Prosperetti (1977, p.24) have typographical errors and should be

$$\theta_l = A_3(R_0/r)\exp[-(1+i)(\frac{1}{2}G_3)^{1/2}r/R_0]\varepsilon e^{i\omega t}; \qquad A_3 = \frac{1}{2}\Gamma_1\Gamma_2(\lambda_1-\lambda_2)\exp[(1+i)(\frac{1}{2}G_3)^{1/2}]/[(G_1(1+w)\Delta].$$

differences for other regions (Figure 2.2). However, for β_{th}, "Prosperetti (1977)" predicts much lower values for G_1=10^{-1}, 5×10^{-2} and 10^{-2} (Figures 2.3-2.5). Our re-visit indicates that the use of the approximation G_1<<1 in Prosperetti (1977) is no longer valid for $G_1 \geq 10^{-2}$ (corresponding to $\omega=4.07\times10^7$ sec^{-1} for air bubbles), in particular for predicting β_{th}. However, the frequency used for experimental evaluation of the polytropic exponent by Crum (1983) is low enough (22.2 kHz and $G_1 \leq 3.64\times10^{-5}$) that Crum's results have not been impaired by this approximation.

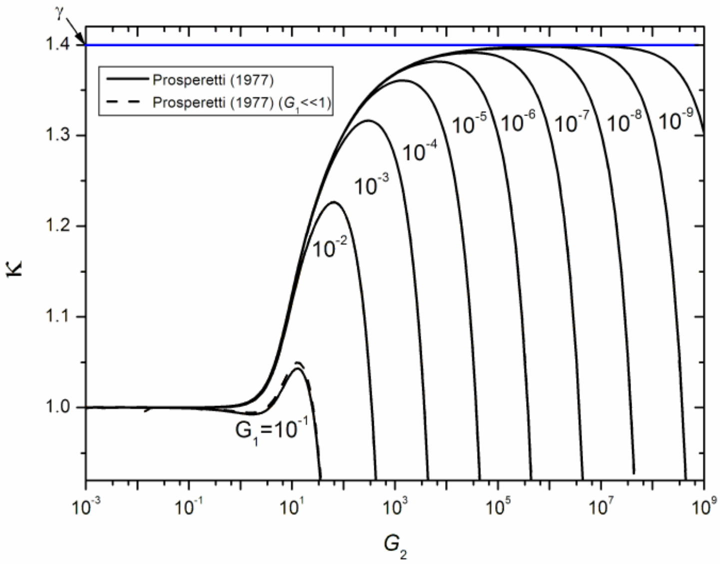

Figure 2.2 Influence of approximation G_1<<1 on the predictions of the polytropic exponent against G_1 and G_2. The labeled value is G_1 varying between 10^{-1} and 10^{-9}.

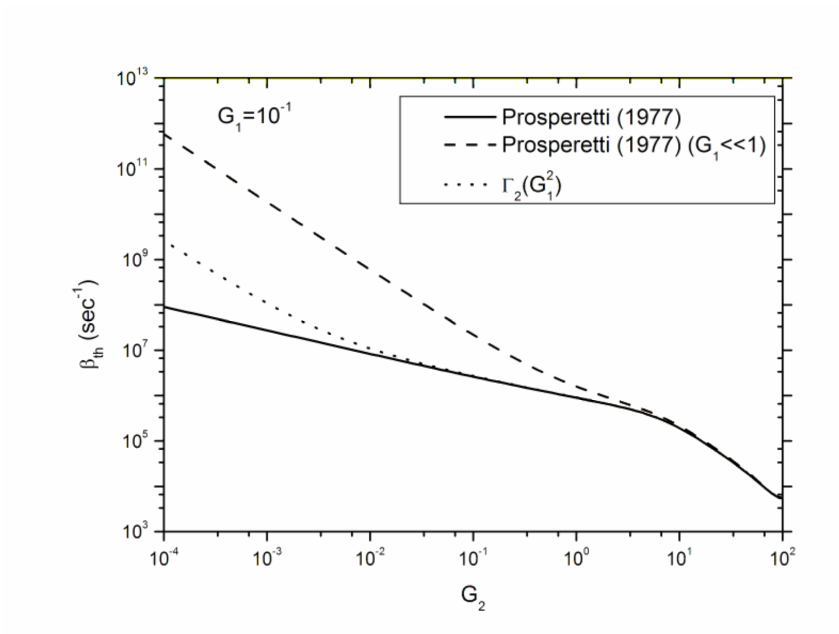

Figure 2.3 Influence of approximation G_1<<1 on the predictions of the thermal damping constant against G_2. $G_1=10^{-1}$.

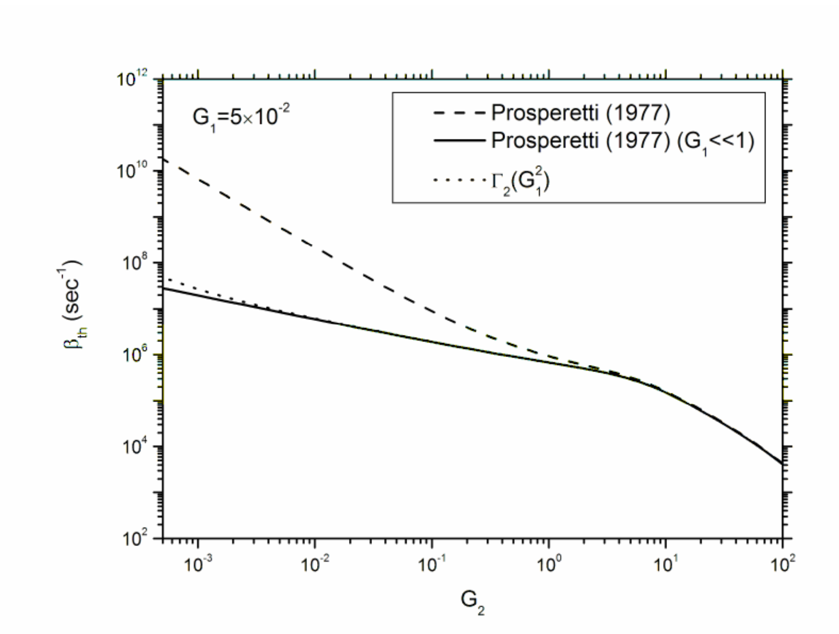

Figure 2.4 Influence of approximation G_1<<1 on the predictions of the thermal damping constant against G_2. $G_1=5\times10^{-2}$.

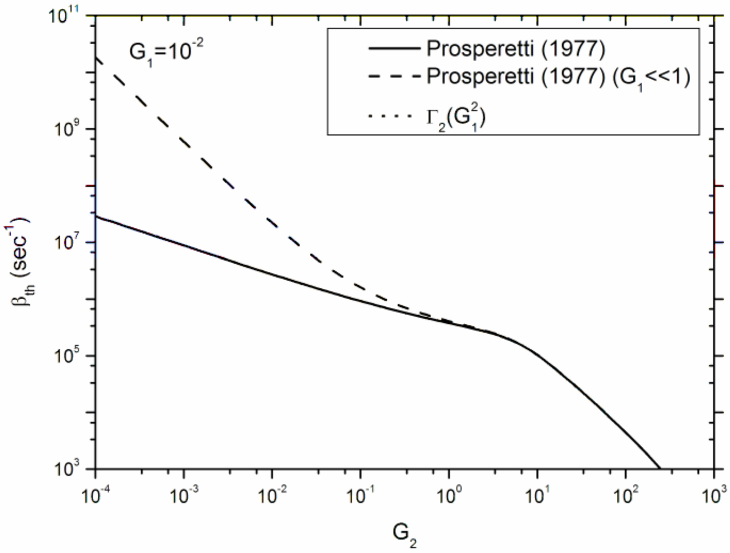

Figure 2.5 Influence of approximation G_1<<1 on the predictions of the thermal damping constant against G_2. $G_1=10^{-2}$.

2.2.6 Simplified formulas for thermal effects

Although the full solution of φ (Eqs. (2.28)-(2.33)) given by Prosperetti (1977) is accurate, it is quite complex and some simplifications are needed. Prosperetti (1977) expanded the Eqs. (2.29) and (2.31) to the first order of G_1 using Taylor series expansion to simplify the formulas, leading to Eqs. (2.36)-(2.39). However, this approach does not work well for high frequencies (i.e. $G_1 \geq 10^{-2}$) as shown in Chap. 2.2.5. In this section, some alternative simplifications are shown and discussed.

Here, we expand the solutions of φ to the third order of G_1 (referring to

Appendix F). After careful examinations, we found that a particular term of order G_1^2 in Γ_2 is paramount for accurate predictions of thermal effects. With inclusion of this term, Γ_2 can be expressed as

$$\Gamma_2 = 2(\gamma-1)G_1\left[1+iG_1/\gamma\right]/\gamma + O\left(G_1^3\right). \tag{2.53}$$

Comparing with Eq. (2.39), an additional term with order of G_1^2 is shown in the imaginary part of Eq. (2.53). In the following discussion, "Γ_2 (G_1^2)" refers to the predictions using Eq. (2.53) for Γ_2 and others are the same as "Prosperetti (1977) (G_1<<1)" (i.e. Eqs. (2.28), (2.30), (2.32)-(2.33), (2.36)-(2.38) and (2.53)). From Figs. 2.3-2.5, one can see that the predictions of thermal damping constant have been significantly improved by "Γ_2 (G_1^2)", even hardly distinguishing the discrepancies between "Γ_2 (G_1^2)" and "Prosperetti (1977)" for the predictions of thermal damping constant for $G_1 \leq 5\times10^{-2}$ (Figures 2.4 and 2.5).

Crum (1983) showed that the Devin's formula (Eq. (2.44)) can be obtained through the reduction of Prosperetti's formulas as follows. For $G_1G_2 << 1$, assumption of uniform pressure inside bubbles used by Devin (1959) is valid. Therefore, using Eqs. (2.30) and (2.37), Crum (1983) obtained

$$\lambda_2 = \beta_2 \coth\beta_2 - 1 \approx \beta_2^2/3 = \frac{1}{3}G_1G_2\left[-1+iG_1(\gamma-1)/\gamma\right] \approx -\frac{1}{3}G_1G_2. \tag{2.54}$$

Substituting Eqs. (2.30), (2.36), (2.38)-(2.39), (2.41) and (2.54) into Eq. (2.43) and neglecting all terms involving G_1, Devin's formula (Eq. (2.44)) can be obtained.

2.2.7 Valid regions of formulas for thermal effects

By the definitions of G_1 (Eq. (2.25)) and G_2 (Eq. (2.26)) (Prosperetti, 1977), one can obtain

$$G_1G_2 = \left(\frac{2\pi R_0}{\lambda_g}\right)^2 = \left(\frac{2\pi R_0}{\lambda_l}\frac{c_l}{c_g}\right)^2, \tag{2.55}$$

where

$$\lambda_g = 2\pi c_g / \omega = (2\pi/\omega)\sqrt{\gamma P_{in,eq}/\rho_g} = (2\pi/\omega)\sqrt{\gamma R_g T_\infty / M_g}\ .$$

Here, c_g and c_l are the speeds of sound in the gas and the liquid respectively; λ_g and λ_l are the wavelengths in the gas and the liquid respectively. Therefore, R_0/λ_g and R_0/λ_l are functions of G_1G_2. In our analysis, uniform pressure assumptions inside and outside gas bubbles are valid if R_0/λ_g<0.1 and R_0/λ_l<0.1 respectively, corresponding to G_1G_2<0.39 and 7.38 respectively for an air bubble oscillating in water. In the following discussion, “Devin (1959)” refers to Eq. (2.44); “Prosperetti (1977)” refers to Eqs. (2.28)-(2.33) as before. According to this criterion, our analysis can be performed for three different regions (Figure 2.6):

Region Ⅰ: G_1G_2<0.39. Both external and internal fields are virtually uniform and the work by Devin (1959) based on the assumption of uniform pressure inside the gas bubble is valid. For this region, there are two sub-cases:

For G_2<1, R_0 is of the order of or smaller than the penetration depth of thermal effects (Prosperetti, 1977). Therefore, the bubble will behave isothermally for almost all G_1.

For $G_2 \geq 1$, R_0 is of the order of or larger than the penetration depth of thermal effects (Prosperetti, 1977) and κ will basically increase with G_2. For $G_1 \leq 10^{-5}$, the value of κ almost approaches the specific heat ratio γ, being virtually independent of G_1 (Prosperetti, 1977). For $G_1 > 10^{-5}$, the maximum κ will gradually decrease with the increase of G_1. Therefore, the maximum κ in the high-frequency regions is much less than γ.

Region Ⅱ: $0.39 \leq G_1 G_2 < 7.38$. In this region, the internal field is no longer uniform because R_0 / λ_g varies between 0.1 and 0.43 and the uniform (pressure) assumption is not applicable. Since the external field can still be treated as uniform one, the solutions for the thermal effects can still be obtained following the analytic approach by Prosperetti (1977).

Region Ⅲ: $G_1 G_2 \geq 7.38$. For this region, both the external and internal fields are non-uniform. Consequently, the shape oscillation of the gas bubble must be considered. The solution will be much more complex.

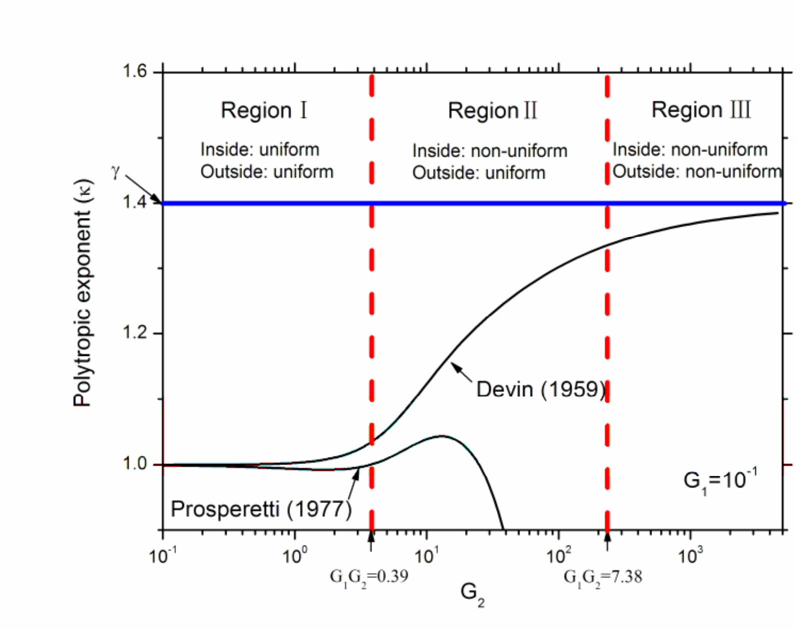

Figure 2.6 Valid regions of formulas for thermal effects.

Valid regions for the formulas derived by Devin (1959) and Prosperetti (1977) are shown above. Since Devin's formulas do not count for G_1 and predict κ between 1 and γ, precautions should be taken when Devin's formulas are used for relatively high frequencies (e.g. $G_1 > 10^{-5}$, see Figure 2.2). The κ predictions of Devin's formulas are not valid for regions II and III. For Prosperetti's formulas, results will not be physical for region III. The κ values predicted in region III by the formulas of Prosperetti (1977) oscillate significantly, with the sign changing alternatively, which has already been shown in Prosperetti (1977, p.20) and in our calculations as well. The valid regions for Devin's or Prosperetti's formulas are emphasized here because such precautions are not seen in many published articles. For example, Prosperetti's formulas were used in Thuraisingham (1997) for calculating the acoustic cross section of a single air bubble in water for the regions of large R_0 / λ_l (referring to Fig.1 in Thuraisingham, 1997), corresponding to $G_1 G_2$ between 1.87×10^{-5} and 1866, partially beyond the valid regions of the Prosperetti's formulas.

2.3 Effect of liquid compressibility

2.3.1 Comments on other published expressions

The expression of acoustic damping constant derived by us in Chap. 2.1 is different with others. The reason for this is explained in this section by re-examination of other published expressions. In this section, for the expression of natural frequency (ω_0), readers are referred to Eq. (2.13). Firstly, we discuss those published papers based on Keller's equation (e.g. Prosperetti, 1984a; Commander and Prosperetti, 1989). If we follow Prosperetti (1984a), dividing Eq. (2.12) by $\left(1 + i\omega R_0 / c_l\right)$, neglecting all terms with order of c^{-2} and using $\ddot{x} = i\omega \dot{x}$ and $\dot{x} = i\omega x$ (Prosperetti, 1984a, p.72), Eq. (2.12) becomes

$$\ddot{x} + 2\left[\frac{2\left(\mu_l + \mu_{th}\right)}{\rho_l R_0^2} + \omega^2 R_0 / 2c_l\right]\dot{x} + \omega_0^2 x = -\alpha_0 \varepsilon e^{i\omega t}. \qquad (2.56)$$

This is just the Eqs. (29)-(30) in Prosperetti (1984a) and Eqs. (32)-(33) in

Commander and Prosperetti (1989). The acoustic damping constant ($\omega^2 R_0 / 2c_l$) shown in Eq. (2.56) is different from ours, i.e. $\omega_0^2 R_0 / 2c_l$ of Eq. (2.14), by noticing that $\omega \neq \omega_0$ for non-resonant oscillations. This also explains why the prediction of acoustic damping constant can be so remarkably improved by using our approach for high frequencies and large bubbles (referring to Figures 2.7-2.10) because for those cases the decreasing ω_0 is further deviating from the increasing ω. Indeed, the approach employed in Prosperetti (1984a) as demonstrated above can be avoided. Instead, the expressions of damping and natural frequency can be determined directly from the coefficients of the harmonic oscillator based on the linearization of Keller's equation, which is a real second order equation. Term $\left(1 + i\omega R_0 / c_l\right)$ can still remain at the right hand side of Eqs. (2.5) and (2.12), without using the relations of $\ddot{x} = i\omega\dot{x}$ and $\dot{x} = i\omega x$. This is exactly the approach employed for the derivations of damping and natural frequency in Chap. 2.1. Furthermore, only for oscillations entering the stationary phase, the transient term disappears from the solution such that $\ddot{x} = i\omega\dot{x}$ and $\dot{x} = i\omega x$ (referring to Chap. 2.1 and Appendix C). Nevertheless, to be strictly speaking, these two relations should not be used for the purpose of determining the expressions of damping and natural frequency. Otherwise, they would have changed the coefficients of this inhomogeneous second-order equation that represent the damping and natural frequency of the harmonic oscillator defined by this equation. Consequently, the resultant expressions defined by Eq. (2.56) will deviate from the true damping and natural frequency.

Now, we turn attention to those expressions based on radiation pressure (e.g. Crandall, 1926),

$$P_{rad} = \rho_l R_0 \ddot{R}\left(1 + i\omega R_0 / c_l\right)^{-1}. \qquad (2.57)$$

Then one can obtain as in Prosperetti (1977),

$$P_{in} - P_0\left[1 + \varepsilon e^{i\omega t}\right] - P_{rad} = \frac{2\sigma}{R} + \frac{4\mu_l}{R}\dot{R}. \qquad (2.58)$$

Substituting Eqs. (2.4) and (2.57) into Eq. (2.58), it becomes

$$\ddot{x} + \left[\frac{4(\mu_l + \mu_{th})}{\rho_l R_0^2}\right]\left(1 + i\omega R_0 / c_l\right)\dot{x}$$
$$+\omega_0^2\left(1 + i\omega R_0 / c_l\right)x = -\left(1 + i\omega R_0 / c_l\right)\alpha_0 \varepsilon e^{i\omega t}. \qquad (2.59)$$

In order to demonstrate how the expressions in published literature were reached, the relations of $\ddot{x} = i\omega\dot{x}$ and $\dot{x} = i\omega x$ are to be employed, which will lead to variable expressions for "natural frequency" and "damping" as shown below.

If both sides of Eq. (2.59) divided by $(1 + i\omega R_0 / c_l)$, it becomes

$$\frac{1 - i\omega R_0 / c_l}{1 + \left(\omega R_0 / c_l\right)^2}\ddot{x} + \left[\frac{4(\mu_l + \mu_{th})}{\rho_l R_0^2}\right]\dot{x} + \omega_0^2 x = -\alpha_0 \varepsilon e^{i\omega t}. \qquad (2.60)$$

The first term in Eq. (2.60) then is rearranged using $\ddot{x} = i\omega\dot{x}$ as,

$$\frac{1 - i\omega R_0 / c_l}{1 + \left(\omega R_0 / c_l\right)^2}\ddot{x} = \frac{1}{1 + \left(\omega R_0 / c_l\right)^2}\ddot{x} + \frac{\omega^2 R_0 / c_l}{1 + \left(\omega R_0 / c_l\right)^2}\dot{x}.$$

Substituting it into Eq. (2.60) leads to

$$\frac{1}{1 + \left(\omega R_0 / c_l\right)^2}\ddot{x} + \left[\frac{4\left(\mu_l + \mu_{th}\right)}{\rho_l R_0^2} + \frac{\omega^2 R_0 / c_l}{1 + \left(\omega R_0 / c_l\right)^2}\right]\dot{x} + \omega_0^2 x = -\alpha_0 \varepsilon e^{i\omega t}. \quad (2.61)$$

If all terms with order of c^{-2} are neglected in Eq. (2.61), this will yield the acoustic damping constant as expressed by Eq. (11) in Smith (1935), Eq. (77) in Devin (1959) and Eq. (27) in Chapman and Plesset (1971).

If the first term in Eq. (2.61) is further treated by using the relations of $\ddot{x} = i\omega\dot{x}$ and $\dot{x} = i\omega x$, i.e.,

$$\frac{1}{1 + \left(\omega R_0 / c_l\right)^2}\ddot{x} = \left[1 - \frac{\left(\omega R_0 / c_l\right)^2}{1 + \left(\omega R_0 / c_l\right)^2}\right]\ddot{x} = \ddot{x} + \frac{\left(\omega R_0 / c_l\right)^2}{1 + \left(\omega R_0 / c_l\right)^2}\omega^2 x.$$

Eq. (2.61) will again become another inhomogeneous 2nd order equation with different coefficients,

$$\ddot{x} + \left[\frac{4\left(\mu_l + \mu_{th}\right)}{\rho_l R_0^2} + \frac{\omega^2 R_0 / c_l}{1 + \left(\omega R_0 / c_l\right)^2}\right]\dot{x} + \left[\omega_0^2 + \frac{\left(\omega R_0 / c_l\right)^2}{1 + \left(\omega R_0 / c_l\right)^2}\omega^2\right]x = -\alpha_0 \varepsilon e^{i\omega t}.$$

(2.62)

This is just the equation used for determining the expressions of damping and natural frequency in Prosperetti (1977).

The demonstration above shows how the use of $\ddot{x} = i\omega\dot{x}$ and $\dot{x} = i\omega x$ changes the coefficients of the equation, resulting in various different expressions for the natural frequency and damping constants of the harmonic oscillator as appeared in the literature such as the contradiction between those two groups of published studies represented by Eqs. (2.61) and (2.62) respectively. It also explains why there is a difference between Prosperetti (1984a) and ours shown in Chap. 2.1.

The expression of acoustic damping constant derived by us is different from those by Smith (1935), Devin (1959), Chapman and Plesset (1971), Prosperetti (1977; 1984a) and Commander and Prosperetti (1989). For natural frequency, our expression is almost identical to theirs except for Prosperetti (1977). Therefore, a comparison with Prosperetti (1977) should be essential. The slight difference of acoustic damping between Eq. (2.61) and Eq. (2.62), i.e. $(\omega R_0 / c_l)^2$, is trivial and excluded from discussions. The assumption of spherical bubble (i.e. uniform pressure outside bubble when R_0 / λ_l<0.1, here λ_l is wavelength in liquids) limits the value of $\omega R_0 / c_l$ (i.e. $2\pi R_0 / \lambda_l$) up to 0.628, referring to Chap. 2.2.7. Therefore, those bubbles within this range are to be considered in the following discussions.

2.3.2 Comparisons

In this section, values predicted by Prosperetti (1977) based on radiation pressure will be compared with ours. Forced oscillations of air bubbles in water will be considered with the constants shown in Appendix A. In following figures and discussions, “Prosperetti (1977)” refers to Eq. (2.62) based on radiation pressure and “Present” refers to Eqs. (2.6)-(2.9) based on Keller’s equation derived by us. For completeness, the viscous and thermal damping mechanisms are also considered in this section. For predictions of thermal effects (μ_{th} and κ), solution (Eqs. (2.28)-(2.33)) by Prosperetti (1977) is used in this section. To focus on liquid compressibility, natural frequency, acoustic and total damping constants are compared.

Figures 2.7-2.11 show the comparisons of the acoustic and the total damping constants and the natural frequencies for ω=10^4, 10^5, 10^6 and 10^7 sec^{-1} respectively. These comparisons demonstrate that the acoustic damping constant predicted by us is quite different from those in Prosperetti (1977). For the total damping constant and the natural frequency, our predictions are much smaller than those by Prosperetti (1977) in particular for the region involving large $\omega R_0 / c_l$.

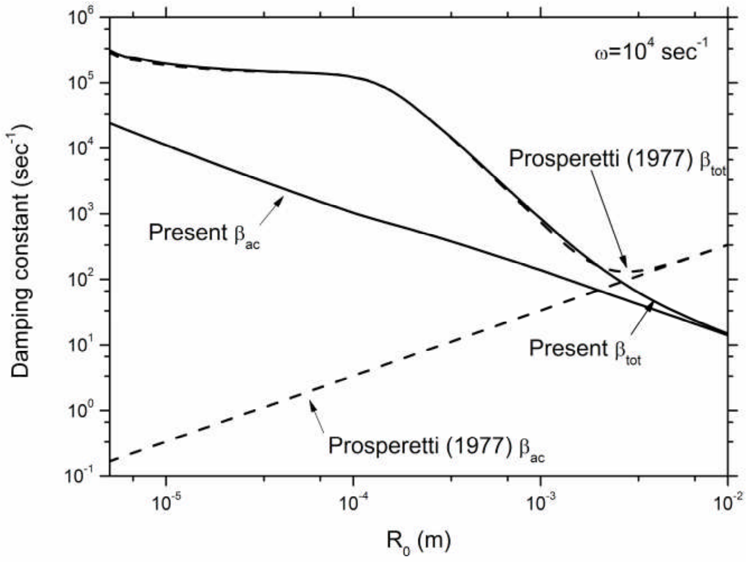

Figure 2.7 Comparison of acoustic and total damping constants between "Prosperetti (1977)" (dash lines) and "Present" (solid lines). ω=10^4 sec^{-1}.

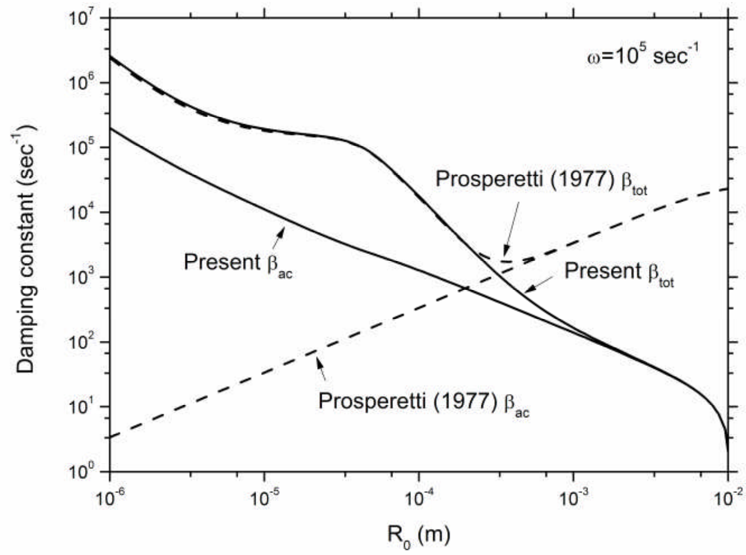

Figure 2.8 Comparison of acoustic and total damping constants between "Prosperetti (1977)" (dash lines) and "Present" (solid lines). ω=10^5 sec^{-1}.

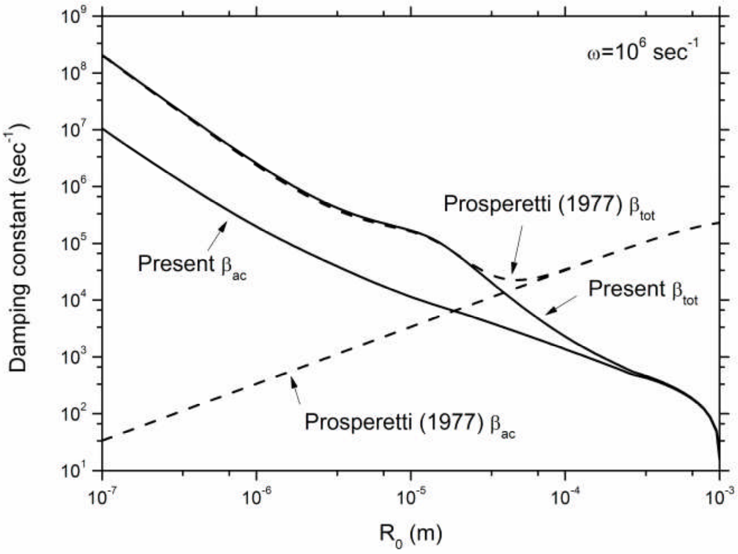

Figure 2.9 Comparison of acoustic and total damping constants between "Prosperetti (1977)" (dash lines) and "Present" (solid lines). ω=10^6 sec^{-1}.

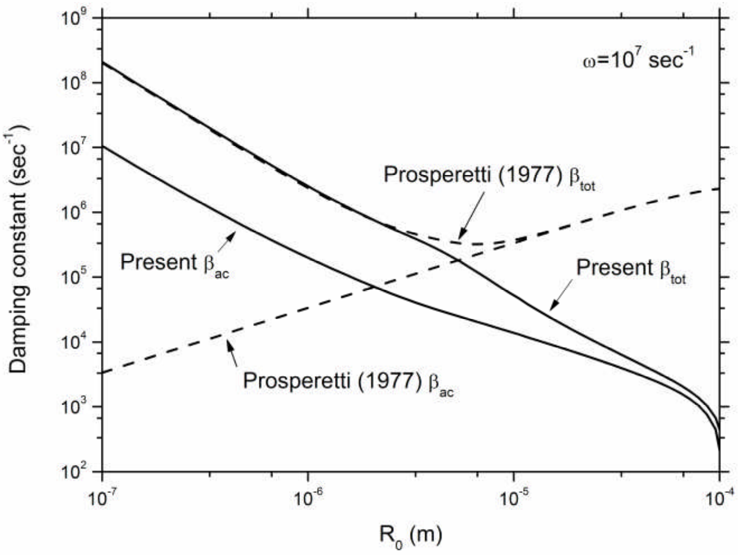

Figure 2.10 Comparison of acoustic and total damping constants between "Prosperetti (1977)" (dash lines) and "Present" (solid lines). ω=10^7 sec^{-1}.

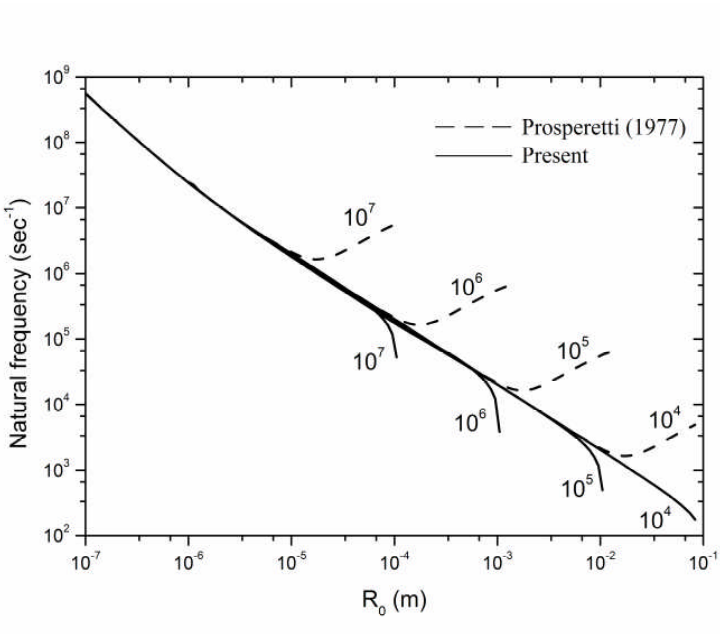

Figure 2.11 Comparison of natural frequency between “Prosperetti (1977)” (dash lines) and “Present” (solid lines). The labeled values are ω (sec^{-1}).

2.4 Examples for air bubbles in water

In this section, contributions from viscous, thermal and acoustic effects to the total damping constant for gas bubbles oscillating in liquids are plotted following Eqs. (2.6)-(2.9) based on Keller’s equation derived by us. For simplicity, forced oscillations of air bubbles in water will be considered.

The viscous, thermal, acoustic and total damping constants are shown for $\omega=10^4$, 10^5, 10^6, and 10^7 sec^{-1} respectively (Figures 2.12-2.15). For fixed frequency, viscous and acoustic damping mechanisms are dominant ones for regions with small and large bubbles respectively while thermal damping is dominant one for the intermediate regions between two regions above. The contribution from thermal damping to the total damping deceases with frequency. Comparing with previous works (e.g. Prosperetti, 1977), values of acoustic damping predicted by us are much smaller and the regions dominated by acoustic damping also shrink.

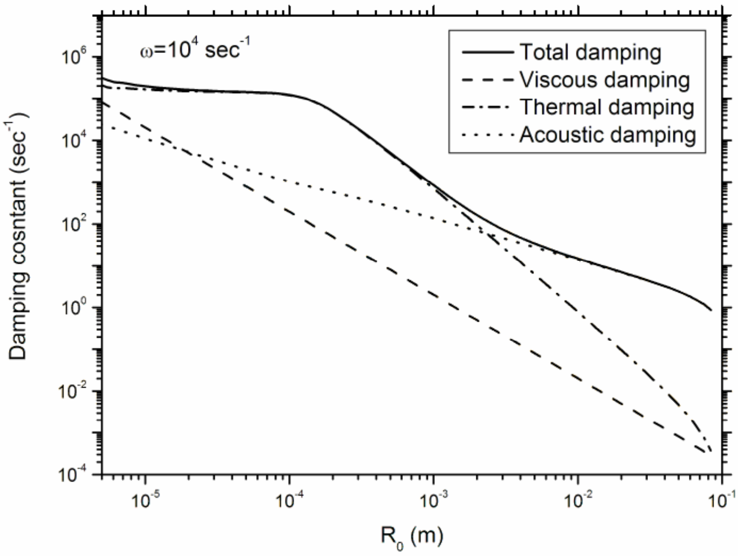

Figure 2.12 Viscous, thermal, acoustic and total damping constant for air bubble oscillations in water. $\omega=10^4$ sec^{-1}.

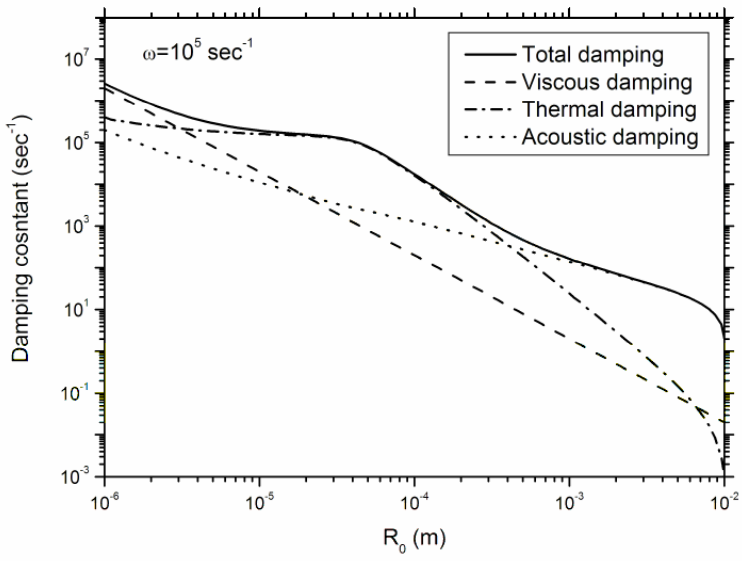

Figure 2.13 Viscous, thermal, acoustic and total damping constant for air bubble oscillations in water. ω=10^5 sec^{-1}.

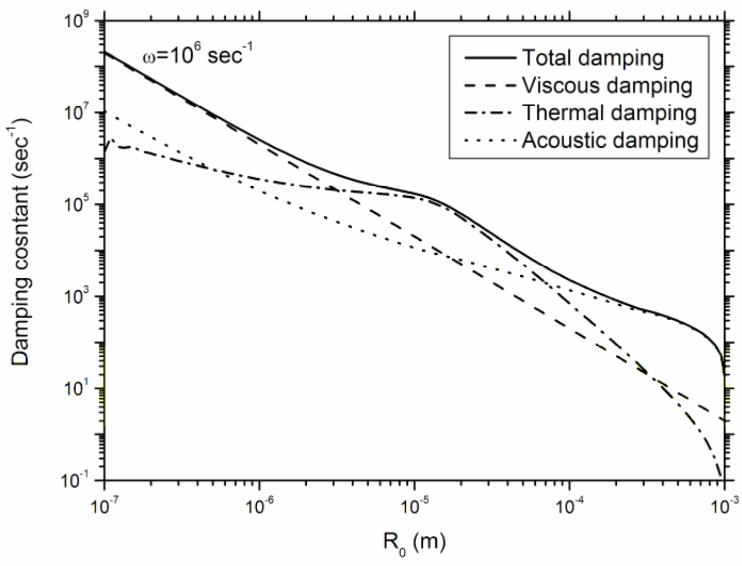

Figure 2.14 Viscous, thermal, acoustic and total damping constant for air bubble oscillations in water. ω=10^6 sec^{-1}.

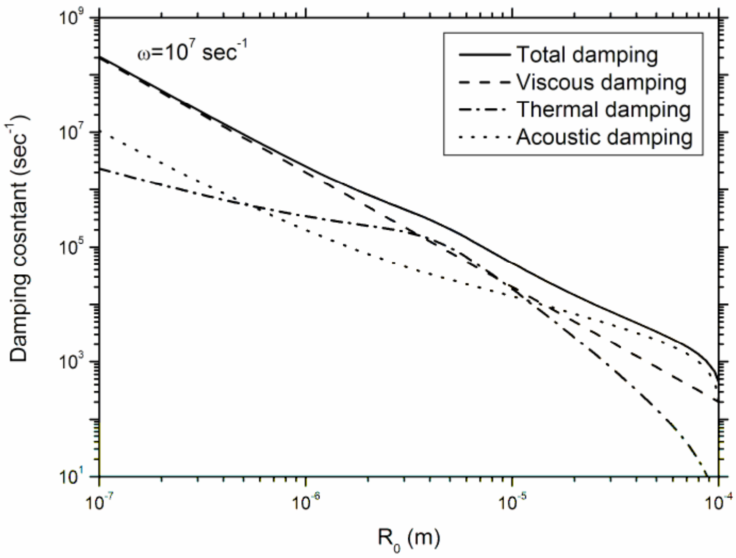

Figure 2.15 Viscous, thermal, acoustic and total damping constant for air bubble oscillations in water. ω=10^7 sec^{-1}.

2.5 Summary

In this chapter, the predictions for damping constants and natural frequency of radial oscillations of gas bubbles in liquids are improved. The concluding remarks are:

a. For acoustic damping constant of forced gas bubble oscillations, a different formula has been proposed by us, which improves the predictions of damping behavior in regions of large bubbles or high frequencies (corresponding to large $\omega R_0 / c_l$).

b. Based on our studies, the approximation of G_1<<1 used by Prosperetti

(1977) to simplify the formulas of thermal effects will be no longer valid for $G_1 \geq 10^{-2}$. Another set of simplified formulas for thermal effects has been proposed by us for the predictions in regions of high frequencies.

c. For $G_1 > 10^{-5}$, the maximum values of the polytropic exponent are less than the specific heat ratio, gradually decreasing with frequency.

d. The valid regions for formulas derived by Devin (1959) and Prosperetti (1977) are categorized based on the ratios of the bubble radii to the wavelengths.

e. A more appropriate expression for non-dimensional thermal damping constant with surface tension included has been proposed by us.

Chapter 3

Rectified mass diffusion of gas bubbles in Newtonian fluids

In this chapter, rectified mass diffusion of gas bubbles in Newtonian fluids will be discussed. Parts of Chaps. 3.3 and 3.4 have been presented at IEEE International Ultrasonic Symposium (2011) held at Orlando, USA (Zhang and Li, 2011). Parts of Chap. 3.5 have been presented at WIMRC 3rd International Cavitation Forum held at Warwick University, Coventry, UK (Li and Zhang, 2011).

3.1 Basic equations

The equation of bubble motion is Keller's equation (Eqs. (2.1)-(2.3)). The mass transfer equation is (Eller and Flynn, 1965),

$$\frac{\partial c}{\partial t} + \mathbf{u} \bullet \nabla c = D \nabla^2 c , \tag{3.1}$$

where c is the concentration of the gas in the liquid; $\mathbf{u}$ is the velocity of the liquid; D is the diffusion constant. The initial and boundary conditions are (Epstain and Plesset, 1950; Eller and Flynn, 1965),

$$c(r,0)=C_i \qquad r>R, \tag{3.2}$$

$$\lim_{r\to\infty} c(r,t)=C_i, \tag{3.3}$$

$$c(R,t)=C_s \qquad t>0, \tag{3.4}$$

where C_i is the initial uniform concentration of the gas in the liquid and also the concentration of the gas in the liquid at infinity; C_s is the concentration of the gas in the liquid at the bubble wall. According to Henry's law, the solubility of the gas in the liquid is proportional to the partial pressure of the gas. Therefore, we have

$$C_0 = k_{\mathbf{H}}^{-1} P_0,$$

$$C_s = k_{\mathbf{H}}^{-1}\left(P_0 + 2\sigma / R\right).$$

Here, C_0 is the saturation concentration of the gas in the liquid; $k_{\mathbf{H}}$ is the Henry's constant. Therefore,

$$C_s = C_0\left(1 + 2\sigma / P_0 R\right). \tag{3.5}$$

3.2 Crum's analysis

As far as analytic work on the subject, Crum and Hansen (1982a) derived the first generalized equations by introducing the polytropic exponent and all three damping mechanisms (i.e. viscous, thermal and acoustic damping mechanisms) which were obtained earlier by Devin (1959) and Eller (1970). Crum and Mao (1996) made further modifications on the formulas in Crum and Hansen (1982a). In this section, formulas derived by Crum and Hansen (1982a) and Crum and Mao (1996) will be cited. We will firstly introduce Crum and Mao (1996)'s work.

Based on Crum and Mao (1996, p.2902), one can obtain

$$\frac{dR_0}{dt} = \frac{Dd}{R_0}\left[\langle R/R_0\rangle + R_0\left(\frac{\left\langle (R/R_0)^4\right\rangle}{\pi t D}\right)^{1/2}\right]$$

$$\times\left(1+\frac{4\sigma}{3P_0R_0}\right)^{-1}\left(\frac{C_i}{C_0}-\frac{\left\langle (R/R_0)^4(P_{in}/P_0)\right\rangle}{\left\langle (R/R_0)^4\right\rangle}\right), \quad (3.6)$$

where

$$d = R_g T_\infty C_0 / P_0 .$$

Here, 〈 〉 denotes time averages.

The instantaneous pressure of the gas in the bubble is given by

$$P_{in} = P_{in,eq}(R_0/R)^{3\kappa} . \quad (3.7)$$

The Rayleigh-Plesset equation is (Crum and Mao, 1996, p.2901)

$$R\ddot{R}+\frac{3}{2}\dot{R}^2+\rho_l^{-1}\left\{P_0+\frac{2\sigma}{R}-P_{in,eq}\left(R_0/R\right)^{3\kappa}+P_A\cos(\omega t)+\rho_l R_0^2\delta_{tot}\frac{\omega_0^2}{\omega}\frac{\dot{R}}{R}\right\}=0.$$

(3.8)[1]

[1] For consistence with Eq. (2.3), $p_s(t) = P_0 - P_A\cos(\omega t)$ in Crum and Mao (1996) is modified as

Here, P_A is the acoustic pressure amplitude; $\delta_{tot} = 2\beta_{tot}\omega / \omega_0^2$ is the non-dimensional total damping constant. The formula for natural frequency is the same as Eq. (2.13), i.e.

$$\omega_0^2 = \frac{1}{\rho_l R_0^2}\left[3\kappa\left(P_0 + \frac{2\sigma}{R_0}\right) - \frac{2\sigma}{R_0}\right].$$

κ and δ_{tot} are given by (Devin, 1959; Eller, 1970)

$$\kappa = \mathrm{Re}\,\Phi / 3,$$

$$\delta_{tot} = \delta_{\mathrm{vis}} + \delta_{th} + \delta_{ac},$$

where

$$\delta_{\mathrm{vis}} = 4\omega\mu / \left[3\kappa\left(P_0 + 2\sigma / R_0\right) - 2\sigma / R_0\right]; \qquad (3.9)$$

$$\delta_{th} = \mathrm{Im}\,\Phi / \mathrm{Re}\,\Phi; \qquad (3.10)$$

$$\delta_{ac} = \rho_l R_0^3 \omega^3 / c_l \left[3\kappa\left(P_0 + 2\sigma / R_0\right) - 2\sigma / R_0\right], \qquad (3.11)$$

which represent non-dimensional viscous, thermal and acoustic damping constants respectively. For the formula of Φ, readers are referred to Eq. (2.44).

Eq. (3.8) can be solved using perturbation method with direct series expansions. Here, an approximate solution of Eq. (3.8) is given such as

$$R / R_0 = 1 + \alpha\left(P_A / P_0\right)\cos\left(\omega t + \delta\right) + \alpha^2 K\left(P_A / P_0\right)^2, \qquad (3.12)$$

where

$p_s(t) = P_0 + P_A \cos(\omega t)$. Therefore, α in Eq. (3.13) is equal to minus of α given by Crum and Mao (1996, p.2902).

$$\alpha = -\frac{P_0}{\rho_l R_0^2}\left[\frac{1}{\left(\omega^2-\omega_0^2\right)^2+\left(\delta_{tot}\omega_0^2\right)^2}\right]^{1/2}; \quad (3.13)$$

$$\delta = \tan^{-1}\left[\frac{\omega_0^2\delta_{tot}}{\left(\omega^2-\omega_0^2\right)}\right]; \quad (3.14)$$

$$K = \frac{\left(3\kappa+1-\beta^2\right)/4+\left(\sigma/2R_0P_0\right)\left(3\kappa+1-2/3\kappa\right)}{1+\left(2\sigma/R_0P_0\right)\left(1-1/3\kappa\right)}; \quad (3.15)$$

$$\beta^2 = \rho\omega^2R_0^2/\left[3\kappa\left(P_0+2\sigma/R_0\right)-2\sigma/R_0\right]. \quad (3.16)^1$$

Then, the time averages $\left\langle R/R_0\right\rangle$, $\left\langle\left(R/R_0\right)^4\right\rangle$ and $\left\langle\left(R/R_0\right)^4\left(P_{in}/P_0\right)\right\rangle$ required in Eq. (3.6) are given by

$$\left\langle R/R_0\right\rangle = 1+K\alpha^2\left(P_A/P_0\right)^2, \quad (3.17)$$

$$\left\langle (R/R_0)^4\right\rangle = 1+\left(3+4K\right)\alpha^2\left(P_A/P_0\right)^2, \quad (3.18)$$

$$\left\langle\left(R/R_0\right)^4\left(P_{in}/P_0\right)\right\rangle = \left[1+\frac{3\left(\kappa-1\right)\left(3\kappa-4\right)}{4}\alpha^2\left(P_A/P_0\right)^2+\left(4-3\kappa\right)K\alpha^2\left(P_A/P_0\right)^2\right]\left(1+\frac{2\sigma}{R_0P_0}\right). \quad (3.19)$$

Substituting Eqs. (3.17)-(3.19) into Eq. (3.6), the bubble growth rate (dR_0/dt) can be obtained. By integration of Eq. (3.6), one can obtain the instantaneous equilibrium bubble radius. If dR_0/dt =0 in Eq. (3.6), the threshold of acoustic pressure amplitude of rectified diffusion (P_T) is

$$P_T^2 = \frac{P_0^2}{\alpha^2}\frac{1+2\sigma/R_0P_0-C_i/C_0}{\left(3+4K\right)C_i/C_0-\left[3\left(\kappa-1\right)\left(3\kappa-4\right)/4+\left(4-3\kappa\right)K\right]\left(1+2\sigma/R_0P_0\right)}.$$

---

[1] Crum and Mao (1996, p.2902) gave the formula of β as

$$\beta = \rho\omega^2R_0^2/\left[3\kappa(P_0+2\sigma/R_0)-2\sigma/R_0\right].$$

Our re-visit to Crum and Mao (1996) found that the missing superscript “2” of β in Crum and Mao (1996) can be treated as typographical error. Therefore, it was corrected in Eq. (3.16) and in the following comparisons in Chap. 3.4.

(3.20)

Now, the formulas derived by Crum and Hansen (1982a) will be cited. The following formulas are used by Crum and Hansen (1982a),

$$\delta_{\text{vis}} = 4\omega\mu / 3\kappa P_0 , \tag{3.21}$$

$$\delta_{ac} = \rho_l R_0^3 \omega^3 / 3\kappa P_0 c_l , \tag{3.22}$$

$$\alpha = -\frac{P_0}{\rho_l R_0^2}\left[\frac{1}{\left(\omega^2-\omega_0^2\right)^2+\left(\omega\omega_0\delta_{tot}\right)^2}\right]^{1/2} , \tag{3.23}$$

$$\beta^2 = \rho\omega^2 R_0^2 / 3\kappa P_0 . \tag{3.24}$$

The other formulas are exactly the same as those in Crum and Mao (1996). Although Crum and Hansen (1982a) considered the effect of surface tension in their physical model, Eqs. (3.21) and (3.22) failed to include effect of surface tension, which has been corrected later in Crum and Mao (1996) i.e. Eqs. (3.9) and (3.11). The difference between Eq. (3.13) and Eq. (3.23) are owing to the incorrect expression of damping term in Eq. (4) of Crum and Hansen (1982a), which has been corrected in Crum and Mao (1996). Comparing Eq. (3.13) with Eq. (3.23), only the term involving δ_{tot} is different, which will be only dominant term near resonance ($\omega \approx \omega_0$). The difference between Eq. (3.16) and Eq. (3.24) will be discussed in next section.

3.3 Improved approach

In this section, we will firstly give some comments on the Crum's analysis shown in Chap. 3.2. Then more physically general formulas for rectified diffusion will be derived. Comments on Crum's analysis are given as follows:

1. Crum's analysis is based on Rayleigh-Plesset equation and the effect of liquid compressibility is considered by using the non-dimensional effective acoustic damping constant (δ_{ac}). For a full account of the effects of liquid compressibility, the equation of bubble motion including liquid compressibility (e.g. Keller's equation Eq. (2.1)) should be directly used.
2. For the formulas of thermal damping (δ_{th}), formulas shown in Devin (1959) were used by Crum and Hansen (1982a) and Crum and Mao (1996). However, as shown in Chap. 2.2.4, Devin's formulas (Eq. (3.10)) should be replaced by Eq. (2.49) with surface tension included. Furthermore, as shown in Chapters 2.2.6 and 2.2.7, formulas of Devin (1959) for thermal effects can be reduced from the solutions by Prosperetti (1977). Therefore, formulas for thermal effects proposed by Prosperetti (1977) i.e. Eqs. (2.28)-(2.35) will be used by us.
3. For acoustic damping, a different formula has been derived by us in

Chap. 2.1 and compared with others (i.e. Devin, 1959) in Chap. 2.3.

4. Our derivations (referring to Appendix G) which is based on Keller's equation with surface tension and liquid compressibility included indicate that Eq. (3.24) given by Crum and Hansen (1982a) is correct while the "correction" Eq. (3.16) by Crum and Mao (1996) is incorrect.

5. With thermal damping included, the instantaneous pressure of the gas in the bubble should be (Prosperetti, 1977, p.18)

$$P_{in} = P_{in,eq}\left(R_0 / R\right)^{3\kappa} - 4\mu_{th}\dot{R}/R\ .$$

Comparing with Eq. (3.7), there is a phase difference between volume and pressure of gas bubbles caused by thermal damping. The influence of μ_{th} on the term $\left\langle \left(R/R_0\right)^4 \left(P_{in}/P_0\right)\right\rangle$ is discussed in Appendix H.

Therefore, a more physically general derivation will be given in this section based on Keller's equation with surface tension and liquid compressibility included. For completeness, more sophisticated formulas (Eqs. (2.28)-(2.35)) for thermal effects proposed by Prosperetti (1977) will be used. In this section, the formulas for non-dimensional total damping constants δ_{tot} ($\delta_{tot} = 2\omega\beta_{tot}/\omega_0^2$) and natural frequency (ω_0) shown in Chap. 2 (i.e. Eqs. (2.6)-(2.9)) will be used. The influences of initial conditions and transient solution of bubble motion equation are discussed in Appendix I. Eller and Flynn (1965) reported that the contributions from second harmonic term to rectified diffusion is of the order $\left(P_A / P_0\right)^4$. The formulas of $\left\langle R / R_0 \right\rangle$, $\left\langle \left(R / R_0\right)^4 \right\rangle$ and $\left\langle \left(R / R_0\right)^4 \left(P_{in} / P_0\right) \right\rangle$ to the order of $\left(P_A / P_0\right)^4$ are shown in Appendix H. For comparing with Crum's analysis, we only consider the terms contributing to rectified diffusion up to the order of $\left(P_A / P_0\right)^2$ here. With second harmonic term neglected, the approximate solution of Eq. (2.1) can be assumed as Eq. (3.12) with (referring to Appendix G)

$$\alpha = -\frac{P_0}{\rho_l R_0^2 M}\left[\frac{1+\left(\omega R_0 / c_l\right)^2}{\left(\omega^2-\omega_0^2\right)^2+\left(\omega_0^2 \delta_{tot}\right)^2}\right]^{1/2}, \tag{3.25}$$

$$\delta = \tan^{-1}\left[\frac{\left(\omega_0^2-\omega^2\right)\omega R_0 / c_l - 2\beta_{tot}\omega}{\left(\omega_0^2-\omega^2\right)+2\beta_{tot}\omega^2 R_0 / c_l}\right], \tag{3.26}$$

$$K = \frac{\left(3\kappa+1-\beta^2\right)/4+\left(\sigma / 2R_0P_0\right)\left(3\kappa+1-2/3\kappa\right)}{1+\left(2\sigma / R_0P_0\right)\left(1-1/3\kappa\right)}, \tag{3.27}$$

$$\beta^2 = \rho\omega^2 R_0^2 / 3\kappa P_0 . \tag{3.28}$$

For the expression of M, readers are referred to Eq. (2.5). Comparing with Eqs. (3.13) and (3.14) derived by Crum and Mao (1996), additional terms involving $\omega R_0 / c_l$ are included in Eqs. (3.25) and (3.26). Those terms reflect the liquid compressibility but were missed in published works. If $\omega R_0 / c_l << 1$ (i.e. regions with low frequencies and small bubble radius), Eqs. (3.25) and (3.26) then reduce to Eqs. (3.13) and (3.14) respectively. Eqs. (3.27) and (3.28) will remain the same as those derived by Crum and Hansen (1982a) (i.e. Eqs. (3.15) and (3.24)). Owing to the fact that formulas of $\langle R/R_0 \rangle$, $\langle (R/R_0)^4 \rangle$ and $\langle (R/R_0)^4 (P_{in}/P_0) \rangle$ together with the threshold of rectified diffusion (Eqs. (3.17)-(3.20)) are all functions of α and K, the predictions of rectified diffusion obtained by using these three groups of formulas derived by Crum and Hansen (1982a), Crum and Mao (1996) and ours will be different.

3.4 Results and discussions

In this section, the predictions of rectified diffusion based on formulas derived by Crum and Hansen (1982a) and Crum and Mao (1996) were compared with those derived by ours in Chap. 3.3 (referred as "Present" in following discussions). Although Crum and Mao (1996) have made corrections to Crum and Hansen (1982a), formulas from Crum and Hansen (1982a) are still prevalent in currently published literature. In the following analysis, we will focus on air bubble in saturated water ($C_i / C_0 = 1$). The constants are the same as those in Crum and Hansen (1982a, p.1588) as shown in Appendix A[1].

[1] Crum (1980) reported that although some precautions have been made against contaminations, the measured equilibrium surface tension coefficient was 68 dyn/m in his experiment. Therefore, σ =68 dyn/m was used in Crum (1980) and Crum and Hansen (1982, p.1588). For predictions of rectified diffusion in Chap. 3.4 and 3.5, σ =68 dyn/m was also used.

Figures 3.1-3.3 showed the threshold of acoustic pressure amplitude of rectified diffusion predicted by Crum and Hansen (1982a), Crum and Mao (1996) and ours (Present) for frequencies 10^7, 5×10^7 and 10^8 sec^{-1}. Our evaluations showed that the difference between three approaches increase with frequencies. For $\omega<5\times10^6$ sec^{-1}, the differences between predictions by three approaches can be safely ignored. Comparing with predictions by Crum and Hansen (1982a), predictions by ours are different in the regions near and above resonance. Comparing with predictions by Crum and Mao (1996), predictions by ours are different in the regions above resonance. As shown in Chap. 3.3, formulas derived by us has fully accounted for the effect of liquid compressibility, surface tension and all the damping mechanisms. For details, readers are referred to Chapters 3.2 and 3.3.

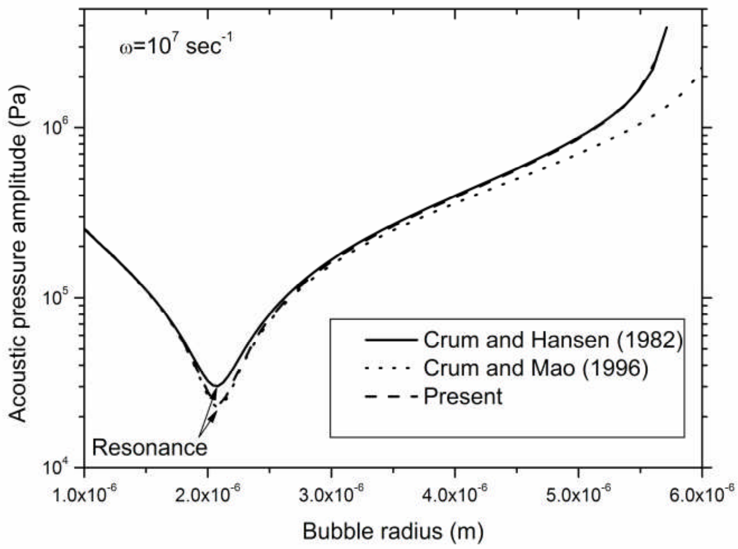

Figure 3.1 Comparisons of threshold of acoustic pressure amplitude of rectified diffusion predicted by Crum and Hansen (1982a), Crum and Mao (1996) and ours (Present). ω=10^7 sec^{-1}.

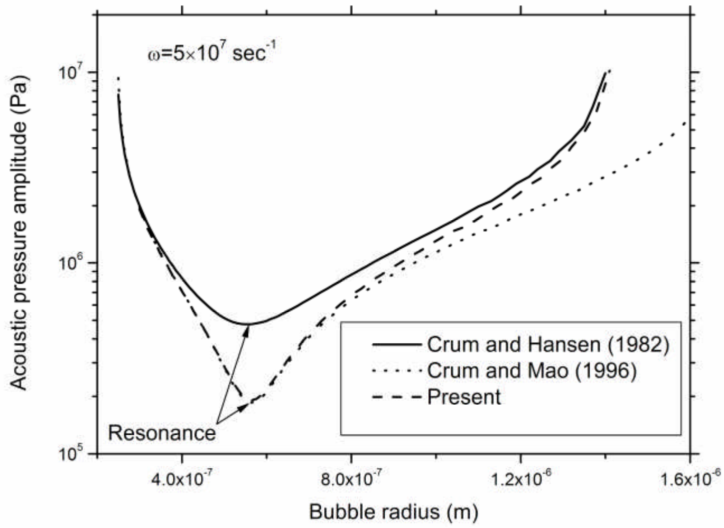

Figure 3.2 Comparisons of threshold of acoustic pressure amplitude of rectified diffusion predicted by Crum and Hansen (1982a), Crum and Mao (1996) and ours (Present). ω=5×10^7 sec^{-1}.

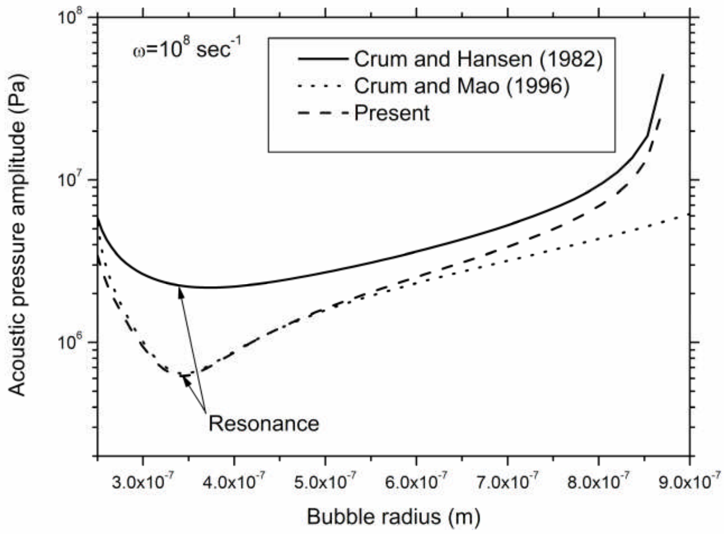

Figure 3.3 Comparisons of threshold of acoustic pressure amplitude of rectified diffusion predicted by Crum and Hansen (1982a), Crum and Mao (1996) and ours (Present). $\omega=10^8$ sec^{-1}.

Nowadays, formulas proposed by Crum and Hansen (1982a) are mostly popular ones in reviews (e.g. Crum, 1984), textbooks (e.g. Leighton, 1994) and research papers. Therefore, we will compare Crum and Hansen (1982a) with ours in the following discussions. Another test case was calculated for behavior of bubble with radius $R_0=2\times10^{-6}$ m under acoustic field with $\omega=10^7$ sec^{-1} and $P_A=3.3\times10^4$ Pa (Figure 3.4). Through comparison with Crum and Hansen (1982a), our predictions showed that the growth rate of equilibrium bubble radius is higher and the final equilibrium radius is larger. As shown in Eq. (3.6), the value of the second term in the bracket decreases with time. Therefore, if time is larger than certain critical value, the bubble growth rate will be reduced as well as shown in Figure 3.4.

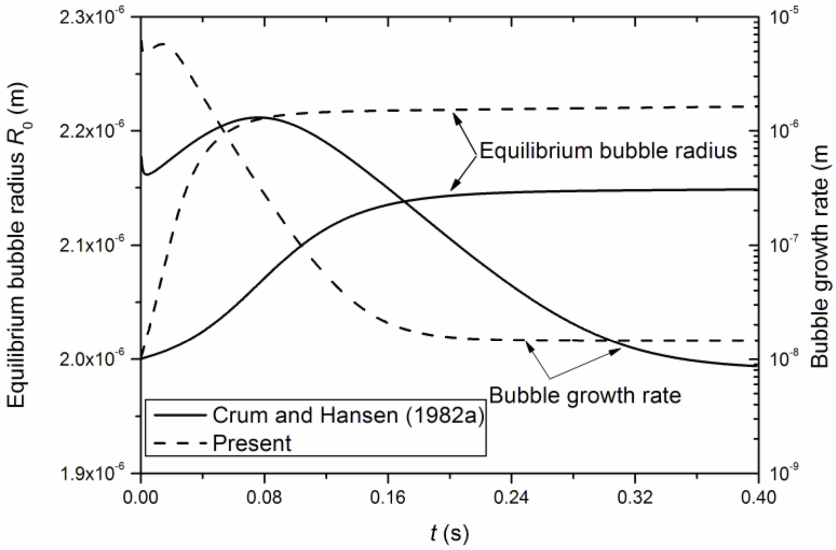

Figure 3.4 Comparison of equilibrium bubble radius and bubble growth rate predicted by Crum and Hansen (1982a) and ours (Present) near resonance. R_0=2×10^{-6} m. ω=10^{7} sec^{-1}. P_A=3.3×10^{4} Pa.

3.5 Dynamic frequency approach

In order to make bubbles grow rapidly, a technique that generates a sound field with its frequency following the decreasing value of the resonant frequency of the growing bubbles (i.e. dynamic-frequency technique) has been proposed by Li (2008). In order to demonstrate the advantage of this technique, comparisons of growing gas bubbles are made between those based on dynamic-frequency and constant-frequency techniques. Formulas in Chap. 3.3 derived by us are used in this section. Demonstrating examples are shown for air bubbles in water with constants given in Appendix A except for σ =68 dyn/m.

For the dynamic-frequency technique, the driving frequency (ω) follows the variation of resonant frequency (ω_r) of the growing bubbles to achieve the fastest growth rate at the status of resonance while the driving frequency (ω) for the constant-frequency technique is equal to resonant frequency (ω_r) of the initial bubble. Here, the resonance frequency is defined as the maximum responsing magnitude of oscillating gas bubbles for given amplitude of sound field (Brennen, 1995, Chap. 4.2). The expression of resonance frequency is (Brennen, 1995, Chap. 4.2),

$$\omega_r = \left(\omega_0^2 - 2\beta_{tot}^2\right)^{1/2}.$$

For many biomedical applications, bubbles are in the range of 1-10 μm, which are chosen for the following demonstrating examples. Fig. 3.5 compares the instantaneous equilibrium bubble radius and its bubble growth rate for the dynamic-frequency and the constant-frequency techniques respectively. The initial equilibrium bubble radius (R_0) is 2 μm, corresponding to the resonant frequency ω_r=1.04×10^7 sec^{-1}. For the constant-frequency approach, the bubble stops growing soon after the resonant frequency (ω_r) of bubbles deviating from the driving frequency

(ω). Whereas, for the dynamic-frequency approach, the bubble grows much quicker since the driving frequency (ω) always follows the reducing resonant frequency (ω_r) of bubbles.

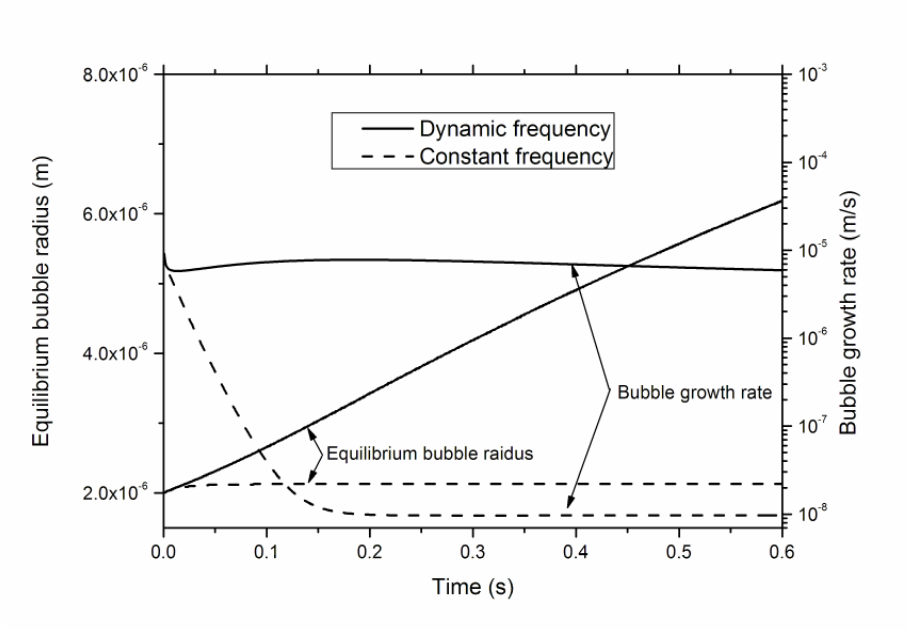

Figure 3.5 Comparison of equilibrium bubble radius and bubble growth rate predicted by the dynamic-frequency and the constant-frequency techniques. The initial R_0 is 2 μm, corresponding to the resonant frequency ω_r=1.04×10^7 sec^{-1}. For the constant-frequency technique, its driving frequency is set as ω_r of initial bubble.

For practical applications, the driving frequency (ω) may vary in steps. Fig. 3.6 shows the effect of step changes in the driving frequency. Even finite steps of frequency change (i=2 or 5, here i is the total number of frequencies used in the dynamic-frequency approach) will accelerate bubble growth considerably.

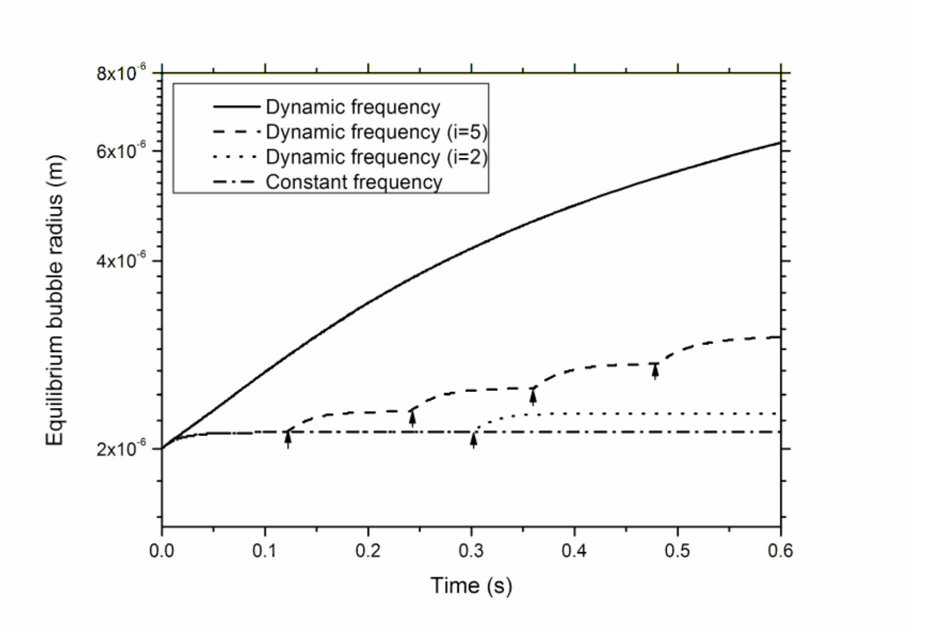

Figure 3.6 Comparison of the dynamic-frequency technique (in step changes) with the constant-frequency technique. Arrows indicate the instance of the step changes in frequency.

Table 3.1 shows the comparisons of the final equilibrium bubble radius for a short duration of 60 ms. Three cases of initial bubble radii (R_0=2, 5 and 10 μm) are tested, representing some typical sizes of bubbles in biomedical applications. Dynamic frequency approach is more effective for small bubbles (e.g. R_0=2 and 5 μm) but less effective for large bubbles (e.g. R_0=10 μm). Whereas for longer duration, say 6s, for the large bubbles (R_0=10 μm) the acceleration effect will manifest itself eventually, giving a final equilibrium bubble radius of 26.84 μm against 12.85 μm (referring to

Table 3.2).

Table 3.1 Comparisons of the final equilibrium bubble radius with time duration of 60 ms.

Method	R_0=2 μm	R_0=5 μm	R_0=10 μm
Constant frequency	2.12 μm	5.42 μm	10.46 μm
Dynamic frequency	2.38 μm	5.59 μm	10.49 μm
Dynamic frequency (i=2)	2.22 μm	5.52 μm	10.48 μm

Table 3.2 Comparisons of the final equilibrium bubble radius with time duration of 6 s.

Method	R_0=2 μm	R_0=5 μm	R_0=10 μm
Constant frequency	2.13 μm	5.94 μm	12.85 μm
Dynamic frequency	22.99 μm	24.27 μm	26.84 μm
Dynamic frequency (i=2)	2.29 μm	7.15 μm	15.23 μm

The dynamic-frequency technique has great potential for accelerating bubble growth in many applications. However, a novel device capable of generating a variable driving frequency is essential for the implementation of this approach.

3.6 Summary

a. A more physically general approach for predictions of rectified mass diffusion of gas bubbles in Newtonian fluids has been proposed and compared with published papers. Results show that predictions of rectified mass diffusion can be improved by using our approach for regions near and above resonance at frequency megahertz and above.

b. To facilitate the growth of gas bubbles through rectified mass diffusion, a dynamic-frequency approach based on near resonance effects has been proposed. The advantages of the dynamic-frequency approach for quick bubble growth has been theoretically demonstrated and compared with constant-frequency approach.

Chapter 4

Rectified mass diffusion of gas bubbles in viscoelastic mediums

In this chapter, rectified mass diffusion of gas bubbles in viscoelastic mediums will be discussed.

4.1 Basic equations for bubble motion

In this section, a model developed by Yang and Church (2005) for the bubble dynamics in soft tissue is introduced. The polytropic model is used to describe the relationship between the internal pressure and the volume of bubble. The generalized Keller equation is (Yang and Church, 2005, Eqs. (12)-(14))

$$\left(1-\frac{\dot{R}}{c_l}\right)R\ddot{R}+\frac{3}{2}\left(1-\frac{\dot{R}}{3c_l}\right)\dot{R}^2=\left(1+\frac{\dot{R}}{c_l}\right)\frac{p_{ext}(t)-p_s(t)}{\rho_l}$$

$$+\frac{R}{\rho_l c_l}\frac{d\left[p_{ext}(t)-p_s(t)\right]}{dt}, \quad (4.1)$$

where

$$p_{ext}(R,t)-p_s(t)=P_{in}-\frac{2\sigma}{R}-P_0-P_0\varepsilon e^{i\omega t}+3\int_R^{\infty}\frac{\tau_{rr}}{r}dr\,. \quad (4.2)$$

Here, τ_{rr} is the stress in the r direction. In order to consider the viscoelastic effect of soft tissue, a linear Voigt model is used by Yang and Church (2005),

$$\tau_{rr}=2\left(G\gamma_{rr}+\mu\dot{\gamma}_{rr}\right). \quad (4.3)$$

Here, G is the shear modulus; γ_{rr} is the strain, which can be expressed as

$$\gamma_{rr}=-\left(2/3r^3\right)\left(R^3-R_0^3\right). \quad (4.4)$$

Combining Eqs. (4.2)-(4.4), Yang and Church (2005) obtained

$$p_{ext}(R,t)-p_s(t)=\left(P_0+\frac{2\sigma}{R_0}\right)\left(R_0/R\right)^{3\kappa}-\frac{2\sigma}{R}-P_0-P_0\varepsilon e^{i\omega t}$$

$$-\left[\frac{4G}{3R^3}\left(R^3-R_0^3\right)+\frac{4\mu\dot{R}}{R}\right]. \quad (4.5)$$

If G =0 in Eqs. (4.1) and (4.5), Keller’s equation (Eqs. (2.1)-(2.3)) is reduced.

4.2 Natural frequency and damping for radial oscillations of gas bubbles in viscoelastic mediums

In this section, Eqs. (4.1) and (4.5) are linearized to obtain the expressions of natural frequency and damping of the radial oscillations of gas bubbles in viscoelastic mediums. Analysis by Yang and Church (2005) is firstly introduced in Chap. 4.2.1. In Chap. 4.2.2, more appropriate expressions of natural frequency and damping are derived by us with comments on Yang and Church (2005). Then expressions of natural frequency and damping derived by us are compared with those derived by Yang and Church (2005) in Chap. 4.2.3.

4.2.1 Yang and Church's analysis

In this section, expressions of natural frequency and damping derived by Yang and Church (2005) are introduced. The Eqs. (4.1) and (4.5) are linearized by using $R = R_0\left(1 + x\right)$ and neglecting all terms involving second or higher order of x (or ε). The term $i\omega P_0 \varepsilon e^{i\omega t} / c_l$, which is like the term in the right hand side of Eq. (2.5), is replaced by radiation pressure wave (Yang and Church, 2005, Eq. (20)),

$$P_{sac} = \frac{\rho_l \ddot{R} R_0}{1 - i\omega R_0 / c_l}. \tag{4.6}$$

Expressions of $\dot{x} = i\omega x$ and $\ddot{x} = i\omega \dot{x}$ are also used during derivations. The resulting equation is

$$\ddot{x} + 2\beta_{tot}\dot{x} + \omega_0^2 x = -\alpha_0 \varepsilon e^{i\omega t} / M \ , \tag{4.7}$$

where

$$\alpha_0 = \frac{P_0}{\rho_l R_0^2} \ ,$$

$$M = 1 + \frac{R_0}{c_l}\frac{4\mu_l}{\rho_l R_0^2} \ .$$

The total damping constant (β_{tot}) is

$$\beta_{tot} = \beta_{vis} + \beta_{th} + \beta_{ac} + \beta_{\mathrm{int}} + \beta_{el} \ ,$$

where

$$\beta_{vis} = \frac{2\mu_l}{\rho_l R_0^2 M} \ , \tag{4.8}$$

$$\beta_{th} = \frac{R_0}{2c_l}\frac{3\kappa P_{in,eq}}{\rho_l R_0^2 M} \ , \tag{4.9}$$

$$\beta_{ac} = \frac{\omega R_0 / c_l}{1 + \left(\omega R_0 / c_l\right)^2}\frac{\omega}{2M} \ , \tag{4.10}$$

$$\beta_{\mathrm{int}} = -\frac{R_0}{2c_l}\frac{2\sigma / R_0}{\rho_l R_0^2 M} \ , \tag{4.11}$$

$$\beta_{el} = \frac{R_0}{c_l}\frac{2G}{\rho_l R_0^2 M} \ , \tag{4.12}$$

representing the viscous, thermal, acoustic, interfacial and elastic damping constants respectively as defined by Yang and Church (2005). The natural frequency is

$$\omega_0^2 = \frac{1}{M}\left[\frac{3\kappa P_{in,eq}}{\rho_l R_0^2} - \frac{2\sigma}{\rho_l R_0^3} + \frac{4G}{\rho_l R_0^2} + \frac{\omega^2}{1+\left(\omega R_0 / c_l\right)^2}\right]. \qquad (4.13)$$

Based on Eqs. (4.8)-(4.13), some demonstrating examples were given by Yang and Church (2005, Figs.1-3).

4.2.2 Comments on Yang and Church's analysis together with our analysis

Comments on the analysis of Yang and Church (2005) shown in Chap. 4.2.1 are given by us as follows:

1. The replacement of $i\omega P_0 \varepsilon e^{i\omega t} / c_l$ using radiation pressure by Yang and Church (2005, Eq. (20)) is not necessary. The term $i\omega P_0 \varepsilon e^{i\omega t} / c_l$ can be kept at right hand side of equation as treated in Chap. 2.1. Radiation pressure should not be further used because liquid compressibility has been fully considered in the equation of bubble motion (Eq. (4.1)).
2. Expressions of $\dot{x} = i\omega x$ and $\ddot{x} = i\omega \dot{x}$ used during derivations of natural frequency and damping by Yang and Church (2005) are also not necessary, which has been fully explained in Chap. 2.3.
3. In fact, Yang and Church (2005) do not account for thermal damping in their derivations by noticing that $P_{in} = P_{in,eq}\left(R_0 / R\right)^{3\kappa}$ in Yang and Church (2005, p.3598) while with thermal damping included it should be $P_{in} = P_{in,eq}\left(R_0 / R\right)^{3\kappa} - 4\mu_{th}\dot{x}$ (referring to Chap. 2.1) based on

Prosperetti (1977). The so-called “thermal damping constant” (Eq. (4.9)) is in fact a part of acoustic damping as shown later. Therefore, comparisons of thermal damping predicted by Prosperetti (1977) shown in Yang and Church (2005, Figs.2-3) are not necessary.

4. According to our analysis shown later, the two new damping terms (interfacial and elastic damping, Eqs. (4.11) and (4.12)) defined by Yang and Church (2005) can both be considered as parts of acoustic damping.

In order to make comparison with Yang and Church (2005) in the following derivations, the thermal damping is also neglected (μ_{th} =0) and Eqs. (4.1) and (4.5) can be directly linearized by following the process shown in Chap. 2.1. The results are

$$\ddot{x} + 2\beta_{tot}\dot{x} + \omega_0^2 x = -\alpha_0 \varepsilon \left(1 + i\omega R_0 / c_l\right) e^{i\omega t} / M \ , \tag{4.14}$$

where

$$\alpha_0 = \frac{P_0}{\rho_l R_0^2} ;$$

$$M = 1 + \frac{R_0}{c_l} \frac{4\mu_l}{\rho_l R_0^2} .$$

The total damping constant (β_{tot}) is

$$\beta_{tot} = \beta_{vis} + \beta_{ac} ,$$

where

$$\beta_{vis} = \frac{2\mu_l}{\rho_l R_0^2 M}; \tag{4.15}$$

$$\beta_{ac} = \frac{R_0}{2c_l}\omega_0^2, \tag{4.16}$$

representing the viscous and acoustic damping respectively. The natural frequency is

$$\omega_0^2 = \frac{3\kappa P_{in,eq} - 2\sigma / R_0 + 4G}{\rho_l R_0^2 M}. \tag{4.17}$$

Comparing with expression of natural frequency in Newtonian fluids (i.e. Eq. (2.9)), an additional term involving the shear modulus (G) appears in Eq. (4.17). If G =0, Eqs. (4.15)-(4.17) reduce to Eqs. (2.6), (2.8) and (2.9) with μ_{th} =0. In order to show the influence of shear modulus on the predictions, the acoustic damping constant (Eq. (4.16)) are split into two terms such as

$$\beta_{ac} = \beta_{ac-0} + \beta_{ac-el},$$

with

$$\beta_{ac-0} = \frac{R_0}{2c_l}\frac{3\kappa P_{in,eq} - 2\sigma / R_0}{\rho_l R_0^2 M}, \tag{4.18}$$

$$\beta_{ac-el} = \frac{R_0}{c_l}\frac{2G}{\rho_l R_0^2 M}. \tag{4.19}$$

4.2.3 Discussions

In this section, predictions based on our analysis (referring to Chap. 4.2.2) are compared with those based on Yang and Church (2005) (referring to

Chap. 4.2.1). The constants (referring to Appendix A) used by Yang and Church (2005) are adopted and adiabatic bubbles (κ =1.4) are considered here.

Figure 4.1 compares the natural frequency predicted by Yang and Church (2005), Prosperetti (1977) (only for water) and ours ("Improved approach") with shear modulus G =0.5, 1 and 1.5 MPa and driving frequency 1 MHz. Fig.1 of Yang and Church (2005) shows the natural frequency for free air bubbles in water and soft tissue (i.e. ε =0 in Eq. (4.7)). However, expressions of $\dot{x} = i\omega x$ and $\ddot{x} = i\omega\dot{x}$ used during derivations of Yang and Church (2005) are only valid for forced bubble oscillations. Therefore, forced gas bubble oscillations are considered in Fig. 4.1. The values predicted by Yang and Church (2005) are quite different with ours. For air bubble oscillations in water (G =0), natural frequency predicted by Yang and Church (2005) (Eq. (4.13)) is also different with the one by Prosperetti (1977) (Eq. (2.62)), which is caused by the improper replacement of $i\omega P_0 \varepsilon e^{i\omega t} / c_l$ with radiation pressure in Yang and Church (2005). Based on our predictions, the natural frequency of air bubbles in soft tissue is larger than the values of air bubbles in water and also gradually increases with the increase of the shear modulus.

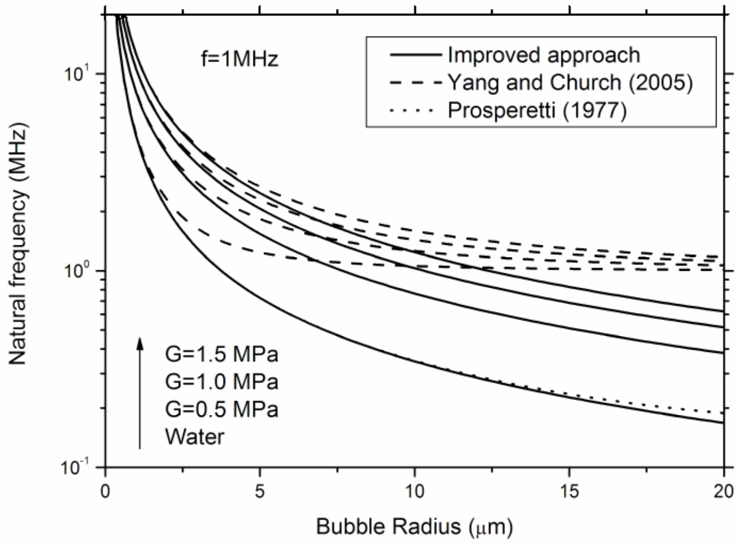

Figure 4.1 Natural frequency predicted by improved approach (Eq. (4.17)), Yang and Church (2005) (Eq. (4.13)) and Prosperetti (1977) (Eq. (2.62), only for water) for forced oscillations of air bubbles in water and soft tissue with shear modulus G =0.5, 1 and 1.5 MPa. Driving frequency is 1 MHz.

For comparisons with Yang and Church (2005, Figs. 2 and 3), the damping constants are plotted for fixed bubble radius (Fig. 4.2) or fixed driving frequency (Fig. 4.3). For Yang and Church (2005), only total damping constant is shown in Figures 4.2 and 4.3. Comparing with Yang and Church (2005), the total damping constant predicted by ours is much smaller especially for regions with high frequencies and large bubbles.

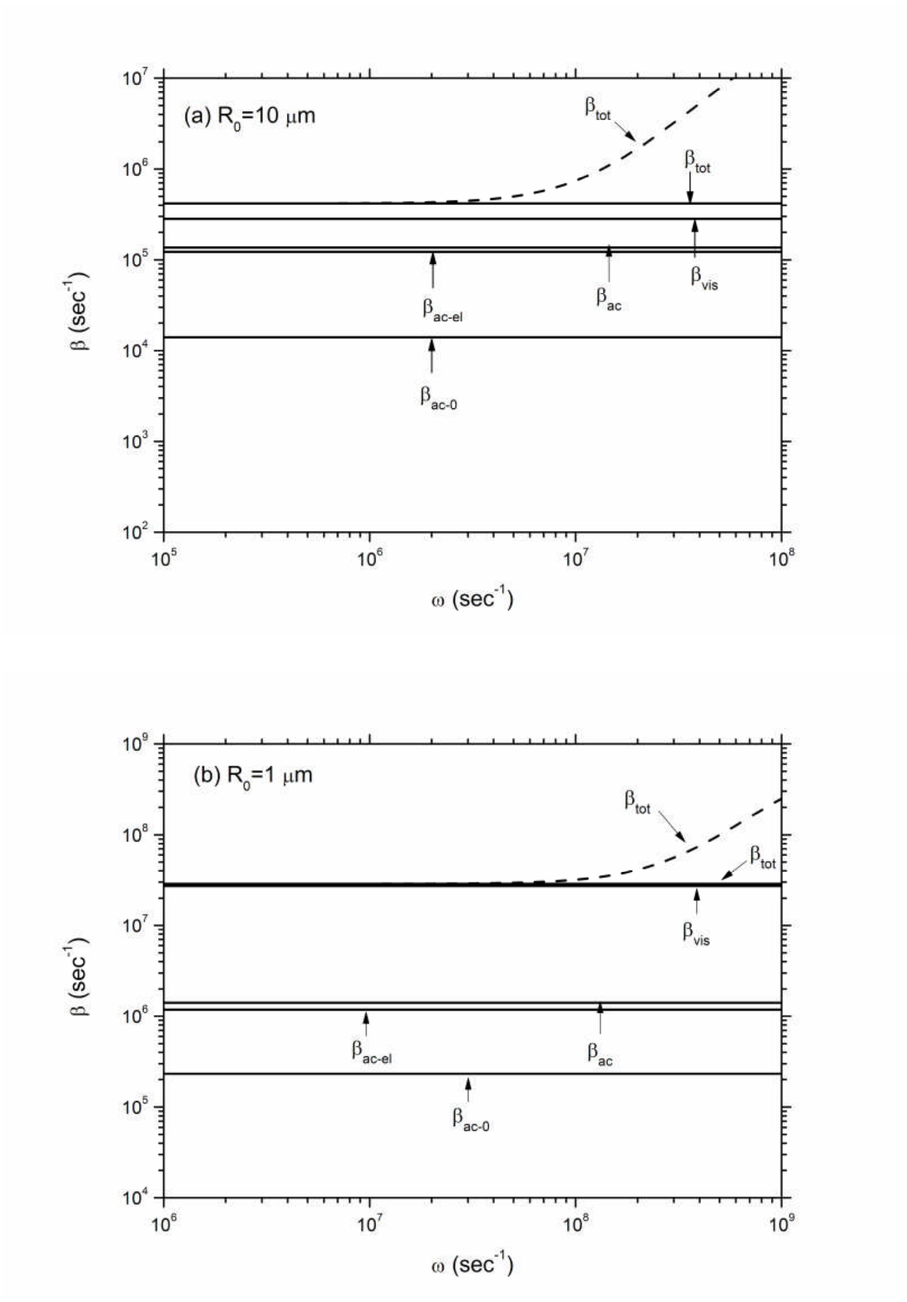

Figure 4.2 Dimensional linear damping constants predicted by Yang and Church (2005) (dash lines) and improved approach (solid lines) against driving frequency for equilibrium bubble radii of (a) 10 μm and (b) 1 μm, surrounded by tissue with G=1.0 MPa and μ_l =0.015 Pa·s.

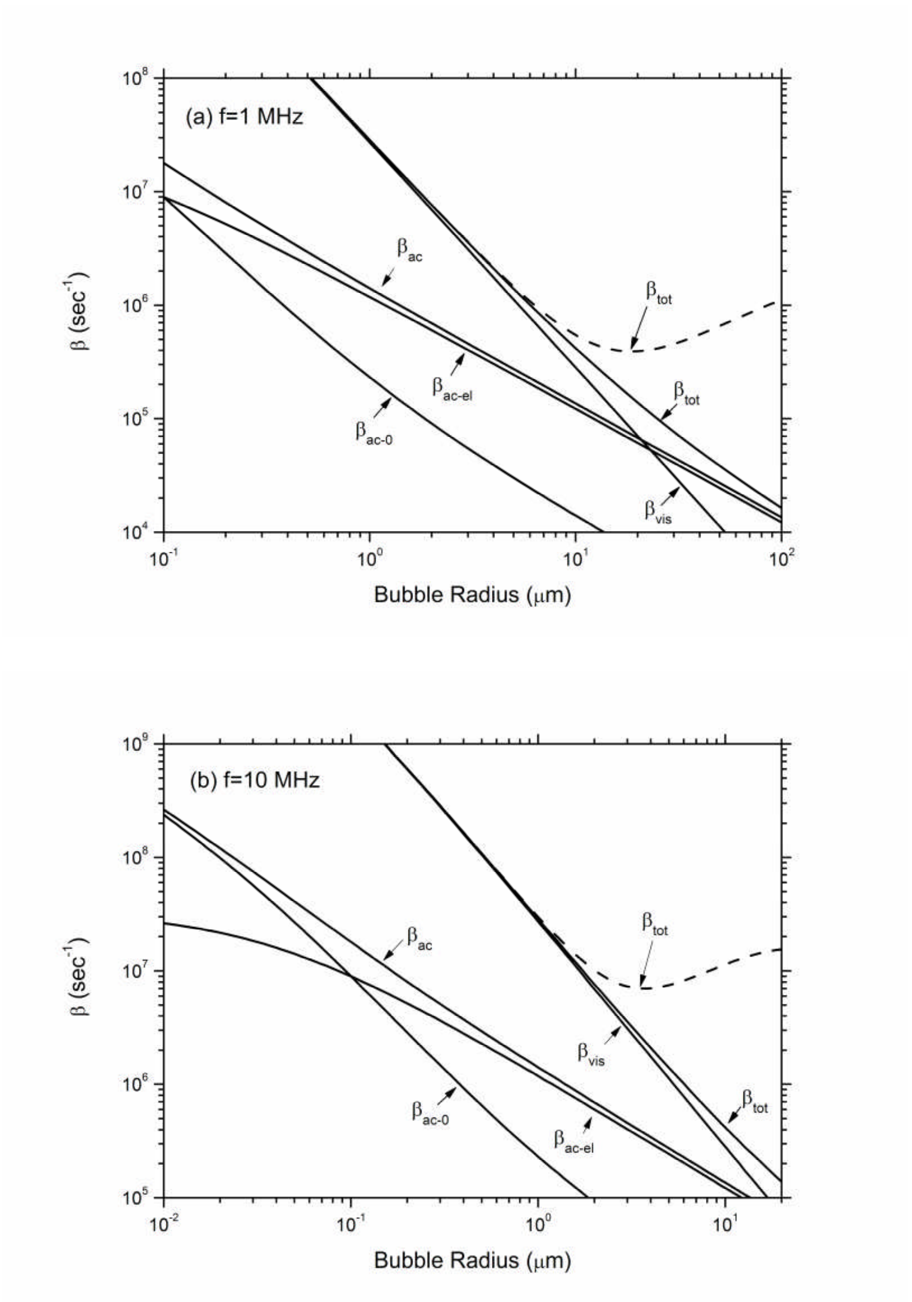

Figure 4.3 Dimensional linear damping constants predicted by Yang and Church (2005) (dash lines) and improved approach (solid lines) against bubble radius for driving frequency of (a) 1 MHz and (b) 10 MHz, surrounded by tissue with *G*=1.0 MPa and μ_l =0.015 Pa·s.

Based on our predictions, for fixed frequency, viscous damping dominates the regions with small bubbles while acoustic damping dominates the regions with large bubbles. Among all the contributions to the acoustic damping, contributions from elastic modulus (β_{ac-el}) is the most important one except for very small bubbles (Fig. 4.3b).

4.3 Rectified mass diffusion in viscoelastic mediums

In this section, the formulas of rectified diffusion in viscoelastic mediums are derived. The governing equations are the equation of bubble motion Eqs. (4.1) and (4.5) and the diffusion equation Eq. (3.1) with initial and boundary conditions Eqs. (3.2)-(3.4). Eller and Flynn (1965) uncoupled the diffusion equation and bubble motion equation based on the fact that gas diffusion across bubble interface is very limited during a single cycle of bubble oscillation. Here, Eller-Flynn's frame work is followed. The solution of bubble motion equations (Eqs. (4.1) and (4.5)) was approximately obtained based on perturbation method with direct series expansions (referring to Appendix G). To the second order of $\left(P_A / P_0\right)$, the approximate solution of Eqs. (4.1) and (4.5) is

$$R / R_0 = 1 + \alpha\left(P_A / P_0\right)\cos\left(\omega t + \delta\right) + \alpha^2 K\left(P_A / P_0\right)^2 , \qquad (4.20)$$

where

$$\alpha = -\frac{P_0}{\rho_l R_0^2 M}\left[\frac{1+\left(\omega R_0 / c_l\right)^2}{\left(\omega^2-\omega_0^2\right)^2+\left(2\omega\beta_{tot}\right)^2}\right]^{1/2}; \qquad (4.21)$$

$$\delta = \tan^{-1}\left[\frac{\left(\omega_0^2-\omega^2\right)\omega R_0 / c_l - 2\beta_{tot}\omega}{\left(\omega_0^2-\omega^2\right)+2\beta_{tot}\omega^2 R_0 / c_l}\right]; \qquad (4.22)$$

$$K = \frac{\left(3\kappa+1-\rho\omega^2 R_0^2 / 3\kappa P_0\right)/4+\left(\sigma / 2R_0P_0\right)\left(3\kappa+1-2/3\kappa\right)+4G/3\kappa P_0}{1+\left(2\sigma / R_0P_0\right)\left(1-1/3\kappa\right)+4G/3\kappa P_0}.$$

(4.23)

For expressions of ω_0, β_{tot} and M, readers are referred to Eqs. (4.14)-(4.17). Second harmonic term is not included in Eq. (4.20) because it contributes to the rectified diffusion with the fourth order of (P_A / P_0) (Eller and Flynn, 1965, p.501; Appendix H) and can be neglected here. The influences of the initial conditions and transient solution are discussed in Appendix I. Based on Eqs. (4.21)-(4.23), one can find that α and K will be influenced by viscoelasticity through shear modulus (G). If G =0, Eqs. (4.21)-(4.23) will reduce to formulas of rectified diffusion in Newtonian liquids Eqs. (3.25)-(3.28) with μ_{th} =0. Because the bubble motion equation and diffusion equation are uncoupled (Eller and Flynn, 1965), the bubble growth rate in viscoelastic mediums can still be expressed as Eq. (3.6) and the time averages $\left\langle R/R_0\right\rangle$, $\left\langle \left(R/R_0\right)^4\right\rangle$ and $\left\langle \left(R/R_0\right)^4\left(P_{in}/P_0\right)\right\rangle$ required in Eq. (3.6) can still be expressed as Eqs. (3.17)-(3.19) with α and K given by Eqs. (4.21) and (4.23).

4.4 Results and discussions

In this section, some demonstrating examples of rectified diffusion of gas bubbles in viscoelastic mediums (e.g. soft tissue) are given. For comparison, air bubble oscillations in water are also shown following formulas in Chap. 3.3. The values of constants used in this section are shown in Appendix A. The driving frequency (f) is 1 MHz. The polytropic exponent (κ) is predicted by Eqs. (2.44) and (2.46) based on Devin (1959) and thermal damping is currently ignored. For air bubbles in water, the following constants are used: σ =72.8 dyn/m; ρ_l =1000 kg/m^3; μ_l =0.001 Pa·s; c_l =1486 m/s. For air bubbles in soft tissue, constants used by Yang and Church (2005) are adopted here: σ =56 dyn/m; ρ_l =1060 kg/m^3; μ_l =0.015 Pa·s; c_l =1540 m/s; G =1 MPa.

Figure 4.4 shows the threshold of the acoustic driving pressure amplitude of rectified diffusion for air bubbles in water and in soft tissue for frequency f =1 MHz and C_i / C_0 =0.96, 1.00 and 1.04. There are two cross points between a given acoustic pressure amplitude and the threshold curve. The corresponding bubble radii of two cross points can be defined as the threshold and maximum bubble radii respectively (Crum and Hansen, 1982b). Bubble will grow to maximum radius through rectified diffusion if its radius is larger than threshold radius. As shown in Figure 4.4, the threshold of acoustic pressure amplitude of rectified diffusion has a minimum value near resonance and increases when bubble is away from resonance. Threshold of acoustic pressure amplitude of rectified diffusion decreases with the increase of the saturation conditions (C_i / C_0). Comparing with air bubbles in water, the shear modulus (G) also contributes to the resonance frequency of bubbles in soft tissue. Therefore, as shown in Figure 4.4, the resonance bubble radius in soft tissue is 10.2 μm, which is larger than 3.2 μm in water. If the acoustic pressure is sufficient high to induce bubble growth through rectified diffusion, large bubbles near 10.2 μm will grow in soft tissue while small bubbles near 3.2 μm will grow in water. For example, with f =1 MHz, C_i / C_0 =1.04 and P_A =1×10^5 Pa, the threshold and maximum bubble radii through rectified diffusion is 9.43 μm and 11.1 μm for air bubbles in soft tissue while is 2.0 μm and 4.5 μm for air bubbles in water respectively.

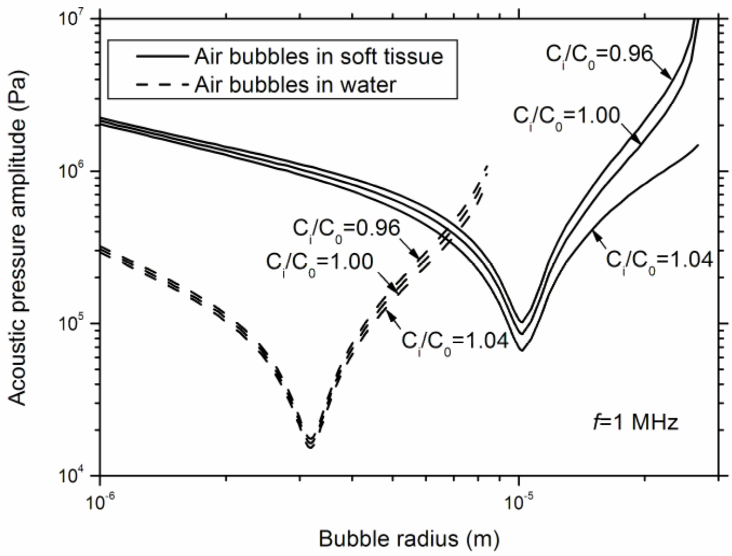

Figure 4.4 Comparison of threshold of acoustic pressure amplitude of rectified diffusion between air bubbles in water and in soft tissue. f =1 MHz. C_i / C_0 =0.96, 1.00 and 1.04.

The bubble growth curves for different saturation conditions are shown in Figure 4.5. The initial bubble radius is assumed to be 10 μm. The bubble radius can reach as large as dozens of micrometers within minutes in soft tissue through rectified diffusion while the growth of bubbles with the same conditions in water is negligible.

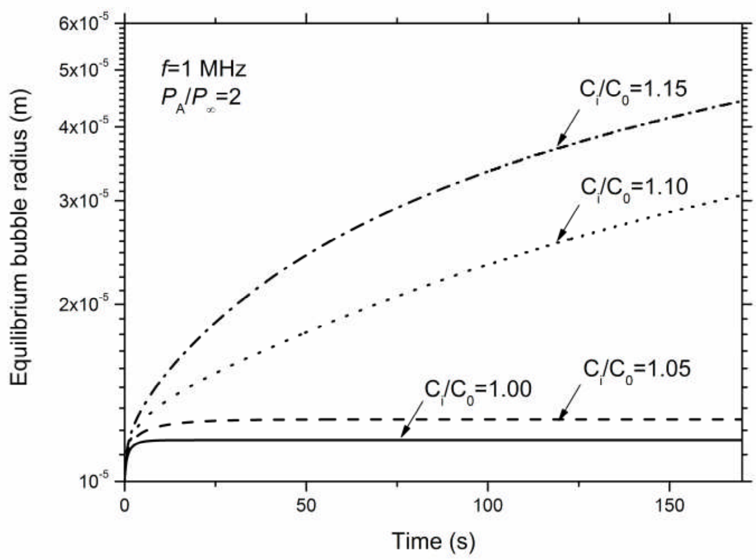

Figure 4.5 Comparison of air bubble growth in soft tissue through rectified diffusion for different saturation conditions. f =1 MHz. C_i / C_0 =1.00 (solid line), 1.05 (dash line), 1.10 (dot line) and 1.15 (dash dot line). P_A / P_0 =2.

4.5 Explanation of cavitation *in vivo*

Ultrasonically induced cavitation *in vivo* has been demonstrated by many researchers (e.g. ter Haar and Daniels, 1981; ter Haar et al., 1982). In ter Haar and Daniels (1981), a guinea-pig hind limb was irradiated using continuous ultrasound and a pulse echo ultrasonic imaging technique was used to visualize both moving and stationary bubbles of diameters down to 10 μm. Crum and Hansen (1982b) proposed that rectified diffusion is the primary mechanism for the bubble growth under irradiation of ultrasound *in vivo* observed by ter Haar and Daniels (1981) and ter Haar et al. (1982). Formulas of Crum (1980) for gas bubble rectified diffusion in Newtonian fluids were used in Crum and Hansen (1982b). Comparing with experimental results of ter Haar and Daniels (1981), Crum and Hansen (1982b) noticed that

1. Based on the predictions, the maximum bubble diameter of the gas bubble growing through rectified diffusion is of order of 20 μm while in the experiment, the diameters of most bubbles is in the range of 10-100 μm (Crum and Hansen, 1982b, p.417). Crum and Hansen (1982b) attributed the above difference to the bubble coalescences and limited resolution of the experimental system.
2. Based on the predictions, the time required for the gas bubble growing to the maximum size is of order of a few seconds while in the experiment, the time is “within the first minute” (Crum and Hansen, 1982b, p.417; ter Haar and Daniels, 1981, p.1147). Crum and Hansen (1982b) attributed the above difference to the “increased damping or local environmental restrictions on the growth rate”.

For the above deviations of predictions, we suggest that the viscoelastic effects of soft tissue which have not been considered in their predictions are possible explanations. In Chap. 4.3, viscoelastic behavior of tissue has been included in the theory of rectified diffusion. In this section, the predictions based on formulas in Chap. 4.3 (named as “soft tissue” in Tab.4.1) are compared with those based on Newtonian fluids previously obtained by Crum and Hansen (1982b). In this section, the constants are chosen as the same as those of experimental conditions of ter Haar and coworkers (ter Haar and Daniels, 1981; ter Haar et al., 1982): continuous ultrasound with frequency f =0.75 MHz and spatial average intensities 80, 150, 300 and 680 mW/cm^2 (corresponding to peak intensities 240, 450, 900 and 2040 mW/cm^2) respectively. Formulas of Crum (1980) were used in Crum and Hansen (1982b) with the constants shown in Appendix A except for σ =60 dyn/cm. Formulas derived by us in Chap. 4.3 are used for the predictions of rectified diffusion in soft tissue with constants shown in Chap. 4.4.

Table 4.1 shows the comparisons of the threshold and maximum bubble radii of rectified diffusion for a wide range of acoustic intensities and dissolved gas concentrations. Both the threshold and maximum bubble radii in soft tissue are much larger than those predicted by Crum and Hansen (1982b). Under oversaturation status, the maximum bubble diameter through rectified diffusion is as large as dozens of micrometers.

Table 4.1 Threshold and maximum bubble radii of rectified diffusion for a wide range of acoustic intensities and dissolved gas concentrations for bubbles in Newtonian fluids and soft tissue respectively. The data for "Newtonian fluids" were directly adapted from Table.1 of Crum and Hansen (1982b).

Concentration	Peak intensity (mW/cm^2)	Newtonian fluids		Soft tissue	
		Threshold bubble radius (μm)	Maximum bubble radius (μm)	Threshold bubble radius (μm)	Maximum bubble radius (μm)
1.00	240	2.4	5.9	---	---
1.00	450	2.0	6.5	13.2	14.1
1.00	900	1.5	7.3	12.6	14.7
1.00	2040	1.1	8.4	11.8	15.4
1.05	240	2.3	6.2	12.9	14.5
1.05	450	1.8	6.9	12.4	15.1
1.05	900	1.4	7.8	11.8	16.0
1.05	2040	1.0	9.1	10.9	18.0
1.10	240	2.2	6.8	10.5	---

1.10	450	1.8	7.8	10.2	---
1.10	900	1.4	9.4	9.7	---
1.10	2040	0.9	11.2	8.9	---
1.15	240	2.0	---	7.3	---
1.15	450	1.6	---	7.2	---
1.15	900	1.2	---	7.1	---
1.15	2040	0.9	---	6.7	---

Figure 4.6 shows the effect of saturation conditions on the bubble growth through rectified diffusion. Comparing with the Figure 2 of Crum and Hansen (1982b), the time required for the gas bubble growing to the maximum size is of order of minute based on our predictions. As shown in Fig. 4.6, the bubble growth rate through rectified diffusion is strongly dependent on the saturation conditions. Therefore, with the viscoelastic effects of tissue included, both the predictions of bubble diameters and time required to reach the maximum bubble sizes are much closer to the experimental observations by ter Haar and Daniels (1981).

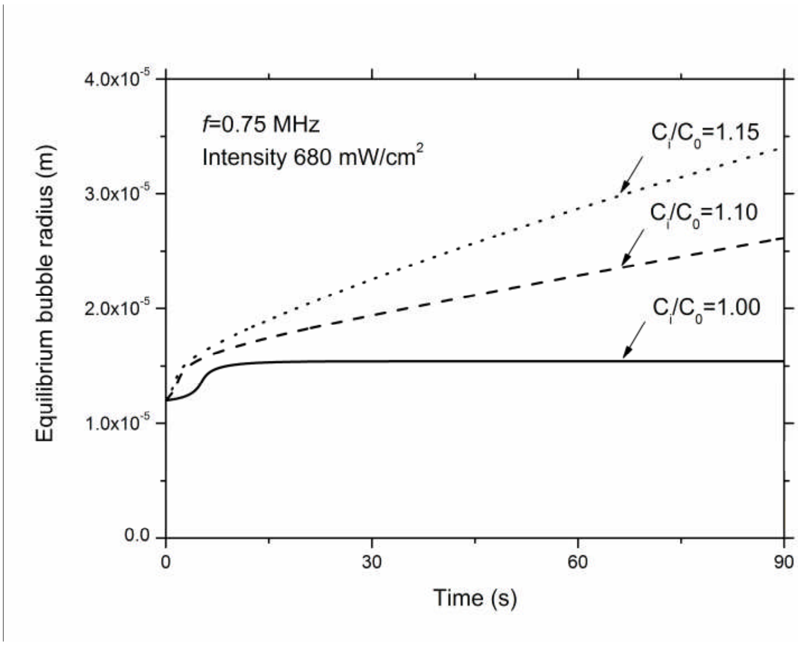

Figure 4.6 Comparison of air bubble growth in soft tissue through rectified diffusion for different saturation conditions. f =0.75 MHz. C_i / C_0 =1.00 (solid line), 1.10 (dash line) and 1.15 (dot line). The spatial average intensity is 680 mW/cm^2. Initial R_0 is 12 μm.

4.6 Summary

a. More physically general formulas for natural frequency and damping of gas bubbles in soft tissue have been proposed. Comparing with Yang and Church (2005), total damping constant and natural frequency predicted by us is much smaller especially for regions involving high frequencies and large bubbles.

b. Formulas for rectified mass diffusion of gas bubble in soft tissue are derived. Comparing with rectified diffusion of gas bubbles in Newtonian fluids, shear modulus of soft tissue also contributes to the rectified diffusion of gas bubbles in soft tissue. We further proposed that viscoelasticity of the soft tissue is a paramount factor to explain the differences between the previous predictions and experiments.

Chapter 5

Rectified mass diffusion of gas bubbles in Newtonian fluids under acoustic field of dual frequency

In this chapter, theoretical expressions of rectified diffusion of gas bubbles in Newtonian fluids under acoustic field of dual frequency are derived and some demonstrating examples are given.

5.1 Formulas of rectified mass diffusion

In this section, formulas of rectified diffusion under acoustic field with dual frequency are derived. Thermal damping is currently neglected. The equation of bubble motion is Keller's equation (Eqs. (2.1) and (2.2)) with Eq. (2.3) replaced by

$$P_s(t) = P_0 + P_{A_1}\cos(\omega_1 t) + P_{A_2}\cos(\omega_2 t). \qquad (5.1)$$

Here, P_{A_1} and P_{A_2} are the amplitudes of external sound field of frequency ω_1 and ω_2 respectively (assuming that $\omega_1 < \omega_2$ for convenience). Solution of equation of bubble motion under acoustic field of dual frequency was given in Appendix J. The solution is

$$\frac{R}{R_0} = 1 + B_2\left(P_{A_1}/P_0\right)^2 + \left[A_{11}\cos(\omega_1 t + \delta_{11}) + A_{12}\cos(\omega_2 t + \delta_{12})\right]\left(P_{A_1}/P_0\right). \qquad (5.2)$$

For simplicity, terms involving (P_{A_2}/P_0) in Eq. (5.2) have been replace by terms involving (P_{A_1}/P_0) noticing that $P_{A_2}/P_0 = (P_{A_2}/P_{A_1})(P_{A_1}/P_0)$. For the expressions of A_{11}, δ_{11}, A_{12}, δ_{12} and B_2, readers are referred to Eqs. (J8)-(J11) and (J16). Although B_2 has no contribution to bubble size measurement in the previous work (Newhouse and Shankar, 1984), B_2 contributes to the rectified diffusion as shown later and its expression has been derived by us. Second-harmonic term and terms involving sum or difference of two frequencies (referring to Eq. (J14)) have no contribution to rectified diffusion to the second order of P_{A_1}/P_0 as shown in Appendix K.

Then, the time averages up to the second order of (P_{A_1}/P_0) can be given as follows (referring to Appendix K)

$$\langle R/R_0\rangle = 1 + B_2\left(P_{A_1}/P_0\right)^2, \qquad (5.3)$$

$$\left\langle (R/R_0)^4\right\rangle = 1 + \left[4B_2 + 3\left(A_{11}^2 + A_{12}^2\right)\right]\left(P_{A_1}/P_0\right)^2, \qquad (5.4)$$

$$\left\langle \left(R/R_0\right)^4 \left(P_{in}/P_0\right)\right\rangle = \left(1+\frac{2\sigma}{P_0R_0}\right)\langle \left(R/R_0\right)^{4-3\kappa}\rangle =$$

$$\left(1+\frac{2\sigma}{P_0R_0}\right)\left\{1+\left[\left(4-3\kappa\right)B_2+\frac{\left(4-3\kappa\right)\left(3-3\kappa\right)}{4}\left(A_{11}^2+A_{12}^2\right)\right]\left(P_{A_1}/P_0\right)^2\right\}.$$

(5.5)

Substituting Eqs. (5.3)-(5.5) into Eq. (3.6), the expression for bubble growth rate can be obtained. With $dR_0/dt=0$ in Eq. (3.6), threshold of acoustic pressure amplitude of rectified diffusion can be obtained. For simplicity, we assume that the acoustic pressure amplitudes of two frequencies are equal (i.e. $P_{A_1}=P_{A_2}=\tilde{P}$). Therefore, the expression of threshold acoustic pressure amplitude of rectified diffusion ($\tilde{P}_T$) will be

$$\tilde{P}_T^2 = \frac{\left(1+\frac{2\sigma}{R_0P_0}\right)-C_i/C_0}{\frac{A_{11}^2+A_{12}^2}{P_0^2}\left[\frac{3C_i}{C_0}-\frac{3\left(4-3\kappa\right)\left(1-\kappa\right)}{4}\left(1+\frac{2\sigma}{R_0P_0}\right)\right]+\frac{B_2}{P_0^2}\left[\frac{4C_i}{C_0}-\left(1+\frac{2\sigma}{R_0P_0}\right)\left(4-3\kappa\right)\right]}.$$

(5.6)

For a given bubble, if the acoustic pressure amplitude is above $\tilde{P}_T$, the bubble will grow. Otherwise, the bubble will dissolve. Now, we will consider one specific case. If the acoustic sources only contains a single frequency (i.e. $P_{A_2}=0$ and $A_{12}=0$), one can find that solution of the equation of bubble motion (Eq. (5.2)) and expression of threshold of acoustic pressure amplitude (Eq. (5.6)) will reduce to those of rectified diffusion under acoustic field of single frequency as shown in Chap. 3.3.

5.2 Results and discussions

In this section, some demonstrating examples based on formulas in Chap. 5.1 are given and compared with those based on single-frequency technique. Air bubbles in water are considered. For the constants used in the calculations, readers are referred to Appendix A. The two driving frequencies are ω_1=5×10^5 sec^{-1} and ω_2=1.5×10^6 sec^{-1}. For convenience, we firstly assume that the amplitudes of the two frequencies in dual-frequency technique are both equal to $\tilde{P}$. For single-frequency technique, the polytropic exponent (κ) is predicted based on Prosperetti (1977). For dual-frequency technique, κ is approximated using the value predicted under the average of two driving frequencies.

Figure 5.1 shows the comparisons of the threshold of acoustic pressure amplitude of rectified diffusion predicted based on single-frequency and dual-frequency techniques. If $\tilde{P}$ is above threshold acoustic pressure amplitude, the bubble will grow otherwise it will dissolve. Resonance bubble radius (R_{r1} and R_{r2}) for two frequencies used in tests can be calculated based on Eq. (2.9) and are shown in Figure 5.1. R_1 and R_2 are the cross points between the threshold curve based on single-frequency technique (ω_2) and $\tilde{P}$. R_3 and R_4 are the cross points between the threshold curve based on single-frequency technique (ω_1) and $\tilde{P}$.

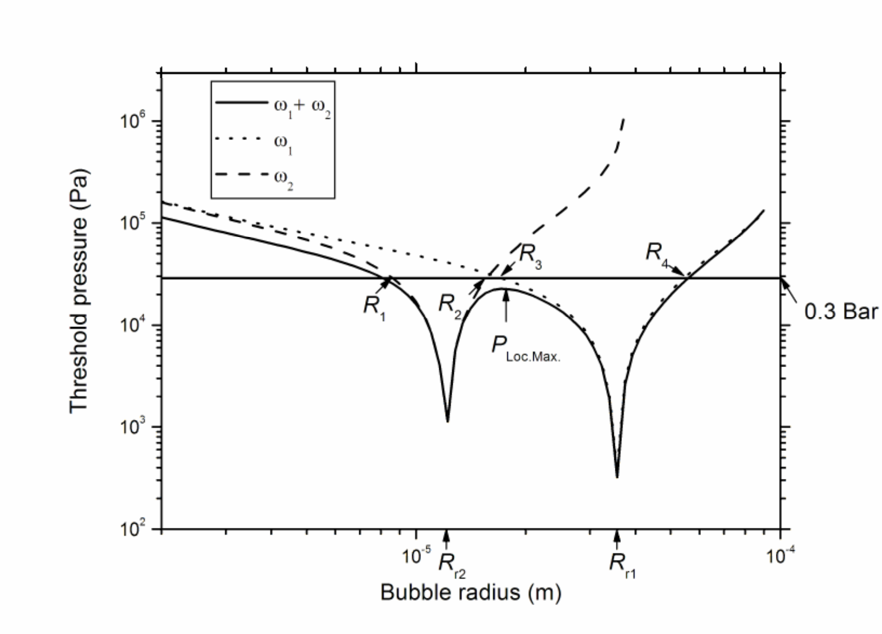

Figure 5.1 Threshold of acoustic pressure amplitude of rectified diffusion predicted based on single- and dual-frequency techniques. R_{r1} and R_{r2} are the resonance radius of frequency ω_1 and ω_2 respectively. $C_i / C_0 = 1$. ω_1=5×10^5 sec^{-1}. ω_2=1.5×10^6 sec^{-1}. For others, readers are referred to texts.

Comparing with single-frequency technique, the threshold of rectified diffusion under dual-frequency technique in the regions between two resonance bubble radius [R_{r1}, R_{r2}] is much smaller and there exists a local maximum threshold pressure ($P_{Loc.Max.}$). If the given amplitude of acoustic field is above the $P_{Loc.Max.}$ (e.g. $\tilde{P}$=0.3 bar in Figure 5.1), the two cross points between threshold curve and $\tilde{P}$ for dual-frequency technique can be assumed as the points marked as R_1 and R_4 with negligible differences. Therefore, the bubble behavior can be divided into five regions as shown in Figure 5.1 and Table 5.1. Comparing with single-frequency technique, a much wider range of bubbles can grow under dual-frequency technique. For example, bubbles with radii within regions [R_1, R_4] can grow based on dual-frequency technique while only bubbles within regions [R_1, R_2] for single-frequency technique with ω_2 or [R_3, R_4] for single-frequency technique with ω_1 can grow. Because threshold of acoustic pressure amplitude of rectified diffusion based on dual-frequency technique is lower than one based on single-frequency technique in the regions [R_{r1}, R_{r2}], one can find that the bubble growth rate under dual-frequency technique will be also higher than those under single-frequency technique in the above regions based on Eq. (3.6).

Comparing with single-frequency technique, the bubble growth rate through rectified diffusion is much higher and the final equilibrium bubble radius is much larger under dual-frequency technique (Figure 5.2). As shown in Figure 5.1, for the given amplitude, bubbles with radius R_0 =10 μm under single frequency ω_1 will dissolve, which is not shown in Figure 5.2. Some examples for over saturation (i.e. $C_i / C_0 = 1.05$) are also shown in Figure 5.2.

Table 5.1 Bubble response and final equilibrium bubble radius under single-frequency and dual-frequency techniques. Others refer to Fig. 5.1.

R_0	ω_1		ω_2		$\omega_1+\omega_2$	
	Response	Final R_0	Response	Final R_0	Response	Final R_0
$R_0 < R_1$	Dissolution	_	Dissolution	_	Dissolution	_
$[R_1, R_2]$	Dissolution	_	Growth	R_2	Growth	R_4
$[R_2, R_3]$	Dissolution	_	Dissolution	_	Growth	R_4
$[R_3, R_4]$	Growth	R_4	Dissolution	_	Growth	R_4
$R_0 > R_4$	Dissolution	_	Dissolution	_	Dissolution	_

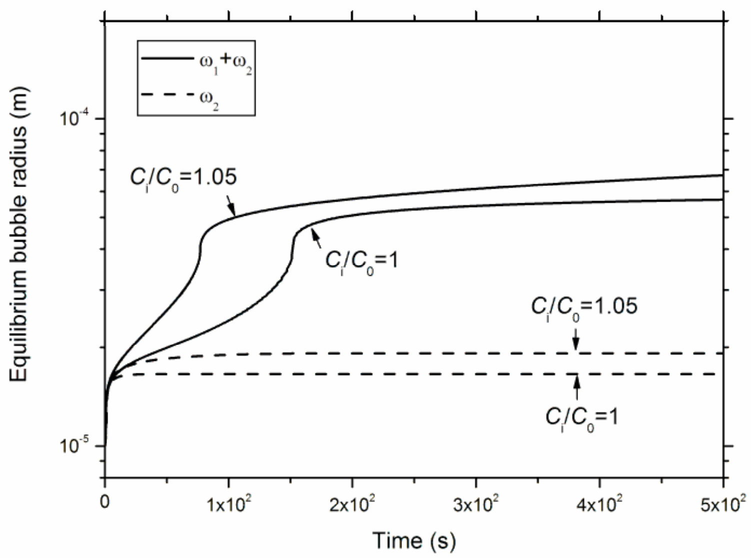

Figure 5.2 Predicted equilibrium bubble radius against time with different saturation status. Initial bubble radius is 10 μm. $\tilde{P}$=0.3 bar. $C_i / C_0 = 1$ and 1.05. ω_1=5×10^5 sec^{-1}. ω_2=1.5×10^6 sec^{-1}.

Figure 5.3 shows the cases of the two frequencies in dual-frequency technique with unequal amplitudes. The bubble growth rate will increase with the increase of amplitude of acoustic field (Figure 5.3). Because the change of acoustic amplitude is limited in Figure 5.3, no distinguishable change of the final equilibrium bubble radius is observed, although the final equilibrium bubble radius through rectified diffusion is dependent on the amplitudes of acoustic field (Figure 5.1).

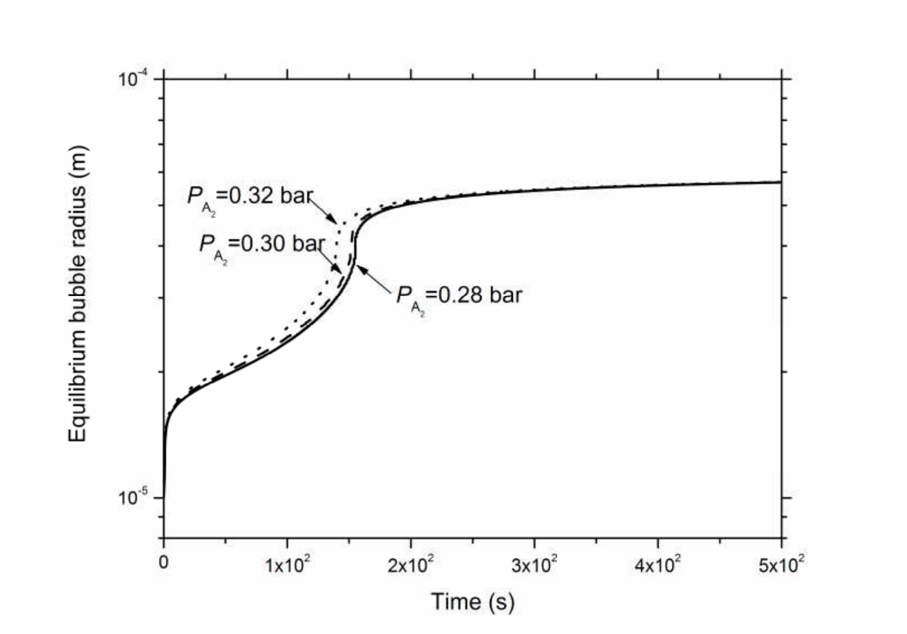

Figure 5.3 Predicted equilibrium bubble radius against time based on dual-frequency technique. Initial bubble radius is 10 μm. P_1=0.3 bar. P_2=0.28 (solid line), 0.3 (dash line) and 0.32 (dot line) bar. $C_i / C_0 = 1$. ω_1=5×10^5 sec^{-1}. ω_2=1.5×10^6 sec^{-1}.

The dual frequency can be generated by a combination of two transducers as used by Newhouse and Shankar (1984) or a single dual-frequency ultrasonic transducer (Saitoh et al., 1995; Wang and Chan, 2003; Huang et al., 2005).

5.3 Summary

a. Formulas of rectified mass diffusion under acoustic field of dual frequency are derived. The formulas for bubble growth rate and threshold of acoustic pressure amplitude of rectified diffusion are obtained.

b. Comparing with single-frequency technique, a wide range of bubbles can rapidly grow into larger bubbles through rectified diffusion by using dual-frequency technique.

Chapter 6

Conclusions

6.1 Achievements

1. Formulas of damping and natural frequency of radial oscillations of gas bubbles in the Newtonian liquids have been derived for more general cases and compared with those in the literature.

   a) For thermal effects, a re-visit to Prosperetti's work (Prosperetti, 1977) has been done. An approximation used by Prosperetti (1977) to simplify the solutions has been re-evaluated and the simplified formulas proposed by Prosperetti (1977) have been improved for high-frequency regions. Surface tension has been considered for non-dimensional thermal damping constant based on Devin (1959). The valid regions of formulas involving the thermal effects have also been defined and discussed.

   b) For effects of liquid compressibility, an improved formula for acoustic damping constant has been derived and compared with previous ones. The predictions of acoustic damping constant can be improved by using our formulas for regions of large bubbles and high frequencies (i.e. large $\omega R_0 / c_l$).

2. Formulas for rectified mass diffusion of gas bubbles in Newtonian fluids have been studied more precisely and compared with those in the literature. Predictions for frequencies at megahertz and above can be improved compared with previous work. The advantages of the dynamic-frequency approach have been demonstrated in comparison with the constant-frequency approach.
3. More physically general formulas of damping and natural frequency of radial oscillations of gas bubbles in the viscoelastic mediums have been derived and compared with those in the literature. Formulas for rectified mass diffusion of gas bubbles in viscoelastic mediums have been derived and applied for explanations of experimental data obtained *in vivo*. The maximum sizes of bubbles growing through rectified diffusion together with the required time can be predicted more precisely by including the viscoelastic effects.
4. Formulas for rectified mass diffusion of gas bubbles in Newtonian fluids under acoustic field with dual frequency have been derived. The advantages of this dual-frequency technique have been demonstrated

by comparing with those under single-frequency excitation. A much wider range of bubbles can grow through rectified diffusion under dual-frequency acoustic excitation.

6.2 Future work

1. The primary assumptions used in this thesis is that bubbles are spherical, which has been well accepted and validated by the previous researchers for studying the problems presented in this thesis. For example, for rectified diffusion, Crum (1980) has demonstrated a fairly well agreement between the experiments and predictions except for addition of surface active agent. Although there is little reason for the unexpected large discrepancy between experiments and predictions based on our formulas, a sound experimental verification will be an excellent propellant for our work in this field. Some experimental work is being considered in the future, e.g. rectified mass diffusion near and above resonance at megahertz and above, rectified mass diffusion in viscoelastic mediums and rectified mass diffusion under dual-frequency acoustic excitation. Development of a numerical code for solving the full set of equations related with heat and mass transfer across bubble interfaces under acoustic excitation

with large amplitude is also being considered to validate results shown in this thesis and gain deeper physical understandings of this problem.

2. Due to the presence of other bubbles (e.g. in a cloud of bubbles), the bubble is non-spherical and bubble-bubble interaction should be considered. For those circumstances, more sophisticated models should be used for simulations. A direct numerical simulation code based on front tracking method developed by Tryggvason and coworkers (Tryggvason et al., 2007; Tryggvason et al., 2011) is a good choice to solve those problems. During this PhD program, some works on this subject have been delivered (Zhang and Li, 2010b; 2010c). Zhang and Li (2010b) proposed a virtual grid based front tracking method for the simulations of bubble cloud with high void fraction. By allowing for the oscillations of non-spherical bubbles, Zhang and Li (2010c) studied the response of cloud of bubbles to pressure waves with finite amplitude. More works are being performed toward the understanding of bubble cloud dynamics.

Appendix A: Constants used for calculations

If not specified, the following values for air and water are used for calculations in Chapters 2-5: ambient pressure P_0 =1.01×10^5 Pa; density of the liquid ρ_l =1000 kg/m^3; speed of sound in the liquid c_l =1486 m/s; viscosity of the liquid μ_l =0.001 Pa·s; surface tension coefficient σ =72.8 dyn/m; diffusion constant D =2.4×10^{-9} m^2/s; thermal diffusivity of the gas at constant pressure $D_{g,p}$ = 2.08×10^{-5} m^2/s; thermal diffusivity of the gas at constant volume $D_{g,v}$ = 2.90×10^{-5} m^2/s; thermal diffusivity of the liquid D_l = 1.42×10^{-7} m^2/s; thermal conductivity of the gas k_g = 2.54×10^3 erg/cm/sec/K; thermal conductivity of the liquid k_l = 5.9×10^4 erg/cm/sec/K; specific heat ratio γ =1.40; universal gas constant R_g =8.314 J/mol/K; molecular weight of the gas in the bubble M_g =28.88 g; absolute temperature T =293.15 K; coefficient $d = R_g T C_0 / P_\infty$ =0.02 (C_0 is saturation concentration of the gas in the liquid in moles per unit volume).

In Chapter 4, if not specified, the following values from Yang and Church (2005, p.3599) are used to mimic the properties of soft tissue: density of the liquid ρ_l =1060 kg/m^3; speed of the sound in the liquid c_l =1540 m/s; surface tension coefficient σ =56 dyn/m; viscosity of the liquid μ_l =0.015 Pa·s; shear modulus G =0.5, 1.0 and 1.5 MPa.

Appendix B: Damping and natural frequency determined based on equations of bubble motion in one-parameter family

In this appendix, the expressions of damping and natural frequency for the radial oscillations of gas bubbles in liquids are determined based on equations of bubble motion in the one-parameter family. Prosperetti and Lezzi (1986) proposed the following one-parameter family equation (i.e. “general Keller-Herring equation”) if written in terms of pressure

$$\left[1-\left(\lambda+1\right)\frac{\dot{R}}{c_l}\right]R\ddot{R}+\frac{3}{2}\left[1-\left(3\lambda+1\right)\frac{\dot{R}}{3c_l}\right]\dot{R}^2 =$$

$$\left[1+\left(1-\lambda\right)\frac{\dot{R}}{c_l}\right]\frac{p_{ext}\left(R,t\right)-p_s\left(t\right)}{\rho_l}+\frac{R}{\rho_l c_l}\frac{d\left[p_{ext}\left(R,t\right)-p_s\left(t\right)\right]}{dt}. \quad \text{(B1)}$$

Here, λ is an arbitrary parameter which is of smaller order of $1/Ma$ (Ma is the bubble wall Mach number) (Prosperetti and Lezzi, 1986, p.466&Eq. (4.3)); $p_{ext}(R,t)$ and $p_s(t)$ are given by Eqs. (2.2) and (2.3) respectively. If $\lambda = 0$, Eq. (B1) reduces to Keller's equation (Eq. (2.1)). If $\lambda = 1$, Eq. (B1) reduces to the equation derived by Herring (1941) such as

$$\left[1-2\frac{\dot{R}}{c_l}\right]R\ddot{R}+\frac{3}{2}\left[1-\frac{4\dot{R}}{3c_l}\right]\dot{R}^2=\frac{p_{ext}(R,t)-p_s(t)}{\rho_l}+\frac{R}{\rho_l c_l}\frac{d\left[p_{ext}(R,t)-p_s(t)\right]}{dt}. \tag{B2}$$

The derivation procedure of damping and natural frequency for linear gas bubble oscillations in liquids based on Eq. (B1) is identical with one shown in Chap. 2.1. Here, we will only keeps terms up to the second order of ε (or x). Substituting $R = R_0(1+x)$ into terms involving λ in Eq. (B1), we obtain the following expressions term by term

$$\left[1-(\lambda+1)\frac{\dot{R}}{c_l}\right]R\ddot{R}=\left[1-(\lambda+1)\frac{R_0\dot{x}}{c_l}\right]R_0^2(1+x)\ddot{x}\approx R_0^2(1+x)\ddot{x}-(\lambda+1)\frac{R_0^3\dot{x}\ddot{x}}{c_l}, \tag{B3}$$

$$\frac{3}{2}\left[1-(3\lambda+1)\frac{\dot{R}}{3c_l}\right]\dot{R}^2\approx\frac{3}{2}R_0^2\dot{x}^2, \tag{B4}$$

$$\begin{aligned}
&\left[1+(1-\lambda)\frac{\dot{R}}{c_l}\right]\frac{p_{ext}(R,t)-p_s(t)}{\rho_l}=\frac{1}{\rho_l}\left[1+(1-\lambda)\frac{\dot{R}}{c_l}\right]\left[\left(P_0+\frac{2\sigma}{R_0}\right)(R_0/R)^{3\kappa}-\frac{2\sigma}{R}\right.\\
&\left.-\frac{4(\mu_l+\mu_{th})}{R}\dot{R}-P_0\left(1+\varepsilon e^{i\omega t}\right)\right]\approx\frac{1}{\rho_l}\left[1+(1-\lambda)\frac{R_0\dot{x}}{c_l}\right]\left\{\left[(-3\kappa)\left(P_0+\frac{2\sigma}{R_0}\right)+\frac{2\sigma}{R_0}\right]x\right.\\
&\left.-4(\mu_l+\mu_{th})\dot{x}-P_0\varepsilon e^{i\omega t}+\left[\frac{3\kappa(3\kappa+1)}{2}\left(P_0+\frac{2\sigma}{R_0}\right)-\frac{2\sigma}{R_0}\right]x^2+4(\mu_l+\mu_{th})x\dot{x}\right\}\\
&\approx\frac{1}{\rho_l}\left\{\left[(-3\kappa)\left(P_0+\frac{2\sigma}{R_0}\right)+\frac{2\sigma}{R_0}\right]x-4(\mu_l+\mu_{th})\dot{x}-P_0\varepsilon e^{i\omega t}\right.\\
&\left.+\left[\frac{3\kappa(3\kappa+1)}{2}\left(P_0+\frac{2\sigma}{R_0}\right)-\frac{2\sigma}{R_0}\right]x^2+4(\mu_l+\mu_{th})x\dot{x}\right\}
\end{aligned}$$

$$+\frac{1}{\rho_l}(1-\lambda)\frac{R_0\dot{x}}{c_l}\left\{\left[(-3\kappa)\left(P_0+\frac{2\sigma}{R_0}\right)+\frac{2\sigma}{R_0}\right]x-4(\mu_l+\mu_{th})\dot{x}-P_0\varepsilon e^{i\omega t}\right\}. \quad \text{(B5)}$$

Based on Eqs. (B3)-(B5), we can clearly see that terms involving the λ in Eq. (B1) is of the second or higher order of ε (or x). Therefore, for linear oscillations, the equations of bubble motion falling into the one-parameter family will give the same expressions for damping and natural frequency (i.e. Eqs. (2.6)-(2.9)).

Appendix C: A complete solution of harmonic oscillator

Eq. (2.5) can be rewritten as

$$\ddot{x} + 2\beta_{tot}\dot{x} + \omega_0^2 x = A_0 \varepsilon e^{i\omega t}, \qquad \text{(C1)}$$

with

$$A_0 = -\alpha_0 \left(1 + i\omega R_0 / c_l\right) / M .$$

A complete solution of Eq. (C1) includes two parts: solution of the corresponding homogenous equation (i.e. Eq. (C1) with $\varepsilon = 0$); solution of the inhomogenous equation. The complete solution of Eq. (C1) is

$$x(t) = e^{-\beta_{tot} t}\left[c_1 e^{\left(-i\sqrt{\omega_0^2 - \beta_{tot}^2}\right)t} + c_2 e^{\left(i\sqrt{\omega_0^2 - \beta_{tot}^2}\right)t}\right] + A_1 \varepsilon e^{i(\omega t + \delta_1)}, \qquad \text{(C2)}$$

where

$$A_1 = \frac{A_0}{\left(\omega_0^2 - \omega^2\right) + 2i\beta_{tot}\omega};$$

$$\delta_1 = \tan^{-1}\left\{\frac{\left(\omega R_0 / c_l\right)\left(\omega_0^2 - \omega^2\right) - 2\beta_{tot}\omega}{\left(\omega_0^2 - \omega^2\right) + 2\beta_{tot}\omega^2 R_0 / c_l}\right\}.$$

Here, c_1 and c_2 are two constants to be determined by the initial conditions. Based on Eq. (C2), one can see that the solution of the corresponding homogenous equation (i.e. the first term in Eq. (C2)) can be safely ignored because it decays exponentially with time owing to the term $e^{-\beta_{tot}t}$.

Appendix D: A brief note on linearization of Gilmore's equation

Gilmore (1952) obtained following equation of bubble motion

$$\left(1-\frac{\dot{R}}{C_l}\right)R\ddot{R}+\frac{3}{2}\left(1-\frac{\dot{R}}{3C_l}\right)\dot{R}^2=\left(1+\frac{\dot{R}}{C_l}\right)H+\left(1-\frac{\dot{R}}{C_l}\right)\frac{R}{C_l}\frac{dH}{dt}. \qquad \text{(D1)}$$

Here, C_l is the speed of sound at bubble wall; H is the enthalpy at the bubble wall. Eq. (D1) was linearized by Shima (1970) and a brief note on the linearization of Eq. (D1) is given in this appendix. Following Prosperetti and Lezzi (1986, Eqs. (3.1) and (3.2)), if the pressure in the liquid p does not deviate too strongly from the undisturbed pressure, one can express the enthalpy h and the speed of sound c as,

$$h=\frac{p-p_0}{\rho_l}\left(1-\frac{p-p_0}{2\rho_l c_l^2}+\dots\right), \qquad \text{(D2)}$$

$$c^{-2}=c_l^{-2}\left(1-\frac{B}{A}\frac{p-p_0}{\rho_l c_l^2}+\dots\right). \qquad \text{(D3)}$$

Here, (B/A) is the standard nonlinearity parameter of acoustics. Based on Prosperetti and Lezzi (1986, p.461), C_l and H can be determined if pressures (p) in Eqs. (D2) and (D3) are set as instantaneous pressure at the bubble wall (i.e. $p = P_{in} - \frac{2\sigma}{R} - \frac{4\mu_l}{R}\dot{R} - P_0\varepsilon e^{i\omega t}$). For the linear oscillations, the terms in the bracket of the order of c_l^{-2} in Eqs. (D2) and (D3) can be safely neglected. Similarly, terms of the order of C_l^{-2} in Eq. (D1) can also be safely ignored for linear oscillations. Then, one can obtain

$$C_l \approx c_l ,$$

$$H \approx \frac{1}{\rho_l}\left(P_{in} - \frac{2\sigma}{R} - \frac{4\mu_l}{R}\dot{R} - P_0\varepsilon e^{i\omega t} - P_0 \right).$$

Therefore, Gilmore's equation (Eq. (D1)) has reduced to Keller's equation (Eqs. (2.1)-(2.3)). Then, Shima's result (Eq. (2.17)) can be reduced from Eq. (2.12) by setting μ_{th} =0 and ε =0.

Appendix E: Derivations of non-dimensional thermal damping constant with surface tension

In this appendix, expression of non-dimensional thermal damping constant (δ_{th}) with surface tension is derived based on frame work of Devin (1959). For convenience, we firstly employ the notations used by Devin (1959) and then those used by Prosperetti (1977).

With inclusion of surface tension, Eq. (4) of Devin (1959) becomes

$$P_2' = P' \exp\left(i\omega t\right) + P_{in,eq} . \qquad \text{(E1)}$$

Here, P_2' is the instantaneous pressure inside bubble; P' is the complex amplitude of the deviation of pressure from the equilibrium value. Because Devin (1959) assumed that the pressure inside bubble is uniform, P_2' and P' are only functions of time (t) rather than radial coordinate (r). Based on the first law of thermodynamics, the deviation of absolute temperature ($\theta_1(r,t)$) from the equilibrium absolute temperature (T_0) was determined by Devin (1959, Eqs. (38) and (40)). The expression of $\theta_1(r,t)$ is not influenced by the surface tension. For a spherical shell with radius r and thickness dr, the equilibrium volume of the shell v_0 can be written as (Devin, 1959, Eq. (41)),

$$v_0 = 4\pi r^2 dr\,. \tag{E2}$$

Based on the law of ideal gas, we obtain[1]

$$P_2'\left(v + v_0\right) = P_{in,eq} v_0 T / T_0\,. \tag{E3}$$

Here, v is the deviation of volume of the shell from the equilibrium volume. Following Eqs. (43)-(47) of Devin (1959) and differentiating both sides of Eq. (E3) and integrating dv from $r=0$ to $r=R_0$, one can obtain the deviation of the total bubble volume (V') from equilibrium total bubble volume (V_0) as follows,

$$V' = \frac{3V_0 P' \exp\left(i\omega t\right)}{P_{in,eq}\Phi}\,. \tag{E4}$$

Here, Φ is given by Eq. (2.44). P_0 in Eq. (47) of Devin (1959) was replaced by $P_{in,eq}$ in Eq. (E4) to include surface tension. Now, we will convert the Devin's notations into Prosperetti's notations (referring to Chap.2.1) such as

$$P_{in} = P_2'\,, \tag{E5}$$

---

[1] Eq. (42) of Devin (1959) ($P_2' v = P_{in,eq} v_0 T / T_0$) contains typo-error, which has been corrected in Eq. (E3).

$$P_0 p(r,t) = P' \exp(i\omega t) . \qquad \text{(E6)}[1]$$

Then, based on Eq. (49) of Devin (1959) and Eq. (2.4), we obtain

$$b_{th}\dot{V}' + k'V' = -P' \exp(i\omega t) = -P_0 p(R_0,t) \approx 4\mu_{th}\dot{x} + 3\kappa P_{in,eq} x . \qquad \text{(E7)}$$

Here, b_{th} is the thermal dissipation coefficient. The meaning of k' will be explained later. Noticing that $V' = (4\pi R_0^3/3)\left[(1+x)^3 - 1\right] \approx 3V_0 x$, we have,

$$b_{th} = 4\mu_{th} / 3V_0 , \qquad \text{(E8)}$$

$$k' = \kappa P_{in,eq} / V_0 . \qquad \text{(E9)}$$

In Eq. (E7), stiffness k used by Devin (1959, Eq. 49) was replaced by k' because the two expressions are not identical with inclusion of surface tension. Devin (1959, p.1661) derived the expression of stiffness k with inclusion of surface tension as follows,

$$k = (\kappa P_{in,eq} / V_0)(1 - 2\sigma / 3\kappa R_0 P_{in,eq}) . \qquad \text{(E10)}$$

k reduces to k' only if surface tension in Eq. (E10) is neglected. Devin (1959) defined δ_{th} as,

$$\delta_{th} = \omega b_{th} / k . \qquad \text{(E11)}$$

Substituting Eq. (E4) into Eq. (E7), one can obtain b_{th} and k' as functions of Φ. Further using Eqs. (E8)-(E11), one can obtain Eq. (2.46) for κ and Eq. (2.49) for δ_{th}.

---

[1] Pay attention that $P' \exp(i\omega t)$ derived based on assumption of uniform pressure inside gas bubble used by Devin (1959) is only a function of time while $P_0 p(r,t)$ derived based on assumption of non-uniform pressure inside gas bubble used by Prosperetti (1977) is a function of both time and radial coordinate.

Appendix F: Derivations of a set of simplified formulas for thermal effects

In this appendix, Taylor Series expansion is applied to Eqs. (2.29) and (2.31) up to the third order of G_1. $\left[(i-G_1)^2+4iG_1/\gamma\right]^{1/2}$ in Eqs. (2.29) and (2.31) can be expanded as

$$\left[(i-G_1)^2+4iG_1/\gamma\right]^{1/2}=i+(2/\gamma-1)G_1+(2i/\gamma)(1/\gamma-1)G_1^2$$
$$+(2/\gamma)(2/\gamma-1)(1-1/\gamma)G_1^3+O\left(G_1^4\right). \quad \text{(F1)}$$

Substituting Eq. (F1) into Eqs. (2.29) and (2.31), we obtain

$$\Gamma_1=2G_1/\gamma+(2/\gamma)(2/\gamma-1)(1-1/\gamma)G_1^3$$
$$+i\left[2+(2/\gamma)(1/\gamma-1)G_1^2\right]+O\left(G_1^4\right), \quad \text{(F2)}$$

$$\Gamma_2=2(1-1/\gamma)G_1-(2/\gamma)(2/\gamma-1)(1-1/\gamma)G_1^3$$
$$-i\left[(2/\gamma)(1/\gamma-1)G_1^2\right]+O\left(G_1^4\right), \quad \text{(F3)}$$

$$\beta_1=(\gamma G_2/2)^{1/2}\left\{2(1/\gamma-1)G_1+(2/\gamma)(2/\gamma-1)(1-1/\gamma)G_1^3\right.$$
$$\left.+i\left[2+(2/\gamma)(1/\gamma-1)G_1^2\right]+O\left(G_1^4\right)\right\}^{1/2}, \quad \text{(F4)}$$

$$\beta_2=(G_1G_2)^{1/2}\left\{-1-(2/\gamma-1)(1-1/\gamma)G_1^2+(1-1/\gamma)G_1 i+O\left(G_1^3\right)\right\}^{1/2}. \quad \text{(F5)}$$

If neglecting G_1^2 and G_1^3 terms in the bracket of Eqs. (F4) and (F5), we obtain

$$\beta_1 = \left(\gamma G_2 / 2\right)^{1/2} \left\{2\left(1/\gamma - 1\right)G_1 + 2i + O\left(G_1^2\right)\right\}^{1/2}, \qquad \text{(F6)}$$

$$\beta_2 = \left(G_1 G_2\right)^{1/2} \left\{-1 + \left(1 - 1/\gamma\right)G_1 i + O\left(G_1^2\right)\right\}^{1/2}. \qquad \text{(F7)}$$

Prosperetti (1977) further applied Taylor series expansion to Eqs. (F6) and (F7), which lead to Eqs. (2.36) and (2.37) for the expressions of β_1 and β_2.

By checking the real and imaginary parts of Eqs. (F2)-(F5) and comparing with Prosperetti's analysis (Eqs. (2.36)-(2.39)), we identify that term $-i\left[\left(2/\gamma\right)\left(1/\gamma - 1\right)G_1^2\right]$ in Γ_2 is a paramount one because this term is of the leading order in the imaginary part of Γ_2 and cannot be neglected for high-frequency regions.

Appendix G: Solution of equation of bubble motion based on perturbation method

In this appendix, the equation of bubble motion is solved based on perturbation method with direct series expansions. The generalized Keller's equation (Eqs. (4.1) and (4.5)) will be firstly solved. The solution of Keller's equation (Eqs. (2.1)-(2.3)) can be conveniently obtained by setting shear modulus G =0 in the solution of the generalized Keller's equation. Although thermal damping is not considered in Eqs. (4.1) and (4.5), it can be conveniently included through the use of effective thermal viscosity (μ_{th}) in the following derivations. The solution of Eqs. (4.1) and (4.5) is assumed as

$$\frac{R}{R_0} = 1 + \varepsilon x_1 + \varepsilon^2 x_2 + \ldots . \qquad \text{(G1)}$$

Here, $\varepsilon = P_A / P_0$. In this appendix, we only consider the terms up to the second order of ε. In Eq. (G1), only stable solution of Eqs. (4.1) and (4.5) is considered. The influence of initial conditions and the solution of homogenous equation (transient solution) are discussed in Appendix I. Substituting Eq. (G1) into Eqs. (4.1) and (4.5), we obtain,

$$
\begin{aligned}
& R_0^2\left[\varepsilon\ddot{x}_1+\varepsilon^2\left(\ddot{x}_2+x_1\ddot{x}_1-\frac{R_0}{c_l}\dot{x}_1\ddot{x}_1\right)\right]+\frac{3}{2}R_0^2\varepsilon^2\dot{x}_1^2 \\
& -\rho_l^{-1}\varepsilon\left[-3\kappa\left(P_0+\frac{2\sigma}{R_0}\right)x_1+\frac{2\sigma}{R_0}x_1-P_0\cos(\omega t)-4\mu_l\dot{x}_1-4Gx_1\right] \\
& -\rho_l^{-1}\frac{R_0}{c_l}\varepsilon\left[(-3\kappa)\left(P_0+\frac{2\sigma}{R_0}\right)\dot{x}_1+\frac{2\sigma}{R_0}\dot{x}_1+P_0\omega\sin(\omega t)-4\mu_l\ddot{x}_1-4G\dot{x}_1\right] \\
& -\rho_l^{-1}\varepsilon^2\left\{\left[-3\kappa\left(P_0+\frac{2\sigma}{R_0}\right)+\frac{2\sigma}{R_0}\right]x_2+\left[\frac{3\kappa(3\kappa+1)}{2}\left(P_0+\frac{2\sigma}{R_0}\right)-\frac{2\sigma}{R_0}\right]x_1^2\right. \\
& \left.-4\mu_l(\dot{x}_2-x_1\dot{x}_1)-4G\left(x_2-2x_1^2\right)\right\} \\
& -\rho_l^{-1}\frac{R_0}{c_l}\varepsilon^2\left\{(-3\kappa)\left(P_0+\frac{2\sigma}{R_0}\right)\dot{x}_2+3\kappa(3\kappa+1)\left(P_0+\frac{2\sigma}{R_0}\right)x_1\dot{x}_1+\frac{2\sigma}{R_0}(\dot{x}_2-2x_1\dot{x}_1)\right. \\
& -4\mu_l\left(-\dot{x}_1^2+\ddot{x}_2-x_1\ddot{x}_1\right)-4G\left[\dot{x}_2-2x_1\dot{x}_1\right] \\
& +x_1\left[(-3\kappa)\left(P_0+\frac{2\sigma}{R_0}\right)\dot{x}_1+\frac{2\sigma}{R_0}\dot{x}_1+P_0\omega\sin(\omega t)-4\mu_l\ddot{x}_1\right] \\
& \left.+\dot{x}_1\left[-3\kappa x_1\left(P_0+\frac{2\sigma}{R_0}\right)+\frac{2\sigma}{R_0}x_1-P_0\cos(\omega t)-4\mu_l\dot{x}_1\right]\right\}=0.
\end{aligned}
\tag{G2}
$$

Based on Eq. (G2), the sum of all terms with order ε (or ε^2) in Eq. (G2) should be equal to zero. To the first order of ε, the result is

$$\ddot{x}_1+2\beta_{tot}\dot{x}_1+\omega_0^2x_1=-\frac{P_0}{M\rho_lR_0^2}\left[\cos(\omega t)-(\omega R_0/c_l)\sin(\omega t)\right], \tag{G3}$$

where

$$\beta_{tot}=\frac{2\mu_l}{M\rho_lR_0^2}+\frac{R_0}{2c_l}\omega_0^2; \tag{G4}$$

$$\omega_0^2 = \frac{1}{M\rho_l R_0^2}\left[3\kappa\left(P_0+\frac{2\sigma}{R_0}\right)-\frac{2\sigma}{R_0}+4G\right]; \tag{G5}$$

$$M = 1+\frac{4\mu_l}{\rho_l R_0^2}\frac{R_0}{c_l}. \tag{G6}$$

The solution of Eq. (G3) can be written as

$$x_1 = A_1\cos\left(\omega t+\delta_1\right), \tag{G7}$$

where

$$A_1 = -\frac{P_0}{M\rho_l R_0^2}\left[\frac{1+\left(\omega R_0/c_l\right)^2}{\left(\omega_0^2-\omega^2\right)^2+4\beta_{tot}^2\omega^2}\right]^{1/2}; \tag{G8}$$

$$\delta_1 = \tan^{-1}\left\{\frac{\left(\omega R_0/c_l\right)\left(\omega_0^2-\omega^2\right)-2\beta_{tot}\omega}{\left(\omega_0^2-\omega^2\right)+2\beta_{tot}\omega^2 R_0/c_l}\right\}. \tag{G9}$$

To the second order of ε in Eq. (G2), we obtain,

$$\ddot{x}_2+2\beta_{tot}\dot{x}_2+\omega_0^2 x_2 = -\frac{\Phi_0}{M}, \tag{G10}$$

where

$$\begin{aligned}\Phi_0 = {} & x_1\ddot{x}_1-\frac{R_0}{c_l}\dot{x}_1\ddot{x}_1+\frac{3}{2}\dot{x}_1^2-\frac{1}{\rho_l R_0^2}\left[\frac{3\kappa(3\kappa+1)}{2}\left(P_0+\frac{2\sigma}{R_0}\right)-\frac{2\sigma}{R_0}\right]x_1^2-\frac{4\mu_l}{\rho_l R_0^2}x_1\dot{x}_1\\ & -\frac{R_0}{c_l}\frac{1}{\rho_l R_0^2}\left[3\kappa\left(3\kappa+1\right)\left(P_0+\frac{2\sigma}{R_0}\right)x_1\dot{x}_1-\frac{4\sigma}{R_0}x_1\dot{x}_1-4\mu_l\left(-\dot{x}_1^2-x_1\ddot{x}_1\right)\right]\\ & -\frac{R_0}{c_l}\frac{1}{\rho_l R_0^2}x_1\left[\left(-3\kappa\right)\left(P_0+\frac{2\sigma}{R_0}\right)\dot{x}_1+\frac{2\sigma}{R_0}\dot{x}_1+P_0\omega\sin\left(\omega t\right)-4\mu_l\ddot{x}_1\right]\\ & -\frac{R_0}{c_l}\frac{1}{\rho_l R_0^2}\dot{x}_1\left[-3\kappa\left(P_0+\frac{2\sigma}{R_0}\right)x_1+\frac{2\sigma}{R_0}x_1-P_0\cos\left(\omega t\right)-4\mu_l\dot{x}_1\right]\\ & +\frac{8G}{\rho_l R_0^2}\left[-x_1^2-\frac{R_0}{c_l}x_1\dot{x}_1\right].\end{aligned} \tag{G11}$$

The solution of Eq. (G10) can be written as

$$x_2 = A_2 \cos\left(2\omega t + \delta_2\right) + B_2 . \tag{G12}$$

Here, B_2 is a constant. To the second order of ε, the oscillating term $A_2 \cos\left(2\omega t + \delta_2\right)$ in Eq. (G12) has no contribution to the time averages $\langle R / R_0 \rangle$, $\langle (R / R_0)^4 \rangle$ and $\langle (R / R_0)^4 (P_{in} / P_0) \rangle$ in the equation of bubble growth rate of rectified diffusion i.e. Eq. (3.6) (Eller and Flynn, 1965, p.501). For details, readers are referred to Appendix H. Therefore, we only need to determine B_2 in Eq. (G12). One can find that terms involving $\dot{x}_1 \ddot{x}_1$ and $x_1 \dot{x}_1$ can be written as harmonic terms (i.e. $\sin\left(2\omega t\right)$) and

$$x_1 \omega \sin\left(\omega t\right) - \dot{x}_1 \cos\left(\omega t\right) = \omega A_1 \sin\left(2\omega t + \delta_1\right).$$

Those terms have no contribution to the time averages in Eq. (3.6) to the second order of ε. Then, the sum of constant terms (Φ_0') in Φ_0 can be written as

$$\Phi_0' = -\frac{1}{2} A_1^2 \omega^2 + \frac{3}{4} A_1^2 \omega^2 - \frac{1}{4} \frac{A_1^2}{\rho_l R_0^2} \left[3\kappa\left(3\kappa + 1\right)\left(P_0 + \frac{2\sigma}{R_0} \right) - \frac{4\sigma}{R_0} \right] - \frac{4 G A_1^2}{\rho_l R_0^2} . \tag{G13}$$

Therefore, we obtain

$$B_2 = -\Phi_0' / \omega_0^2 M . \tag{G14}$$

Although the expression of A_2 in Eq. (G12) is not used in this thesis, it can be determined by Eq. (G11). Now the solution of the generalized Keller's equation (Eqs. (4.1) and (4.5)) required in Chap. 4.3 is given. To the first order of ε (i.e. Eq. (G3)), the above solution reduces to the solution of the linear oscillations in Chap. 4.2.2.

If we set G=0 in Eqs. (G8), (G9) and (G14) and include the thermal damping term by replacing μ_l with $(\mu_l + \mu_{th})$, the solution of Keller's equation (Eqs. (2.1)-(2.3)) to the second order of ε required in Chap. 3.3 will be obtained. To the first order of ε, the above solution reduces to the solution of linear oscillations in Chap. 2.1.

In the other sections, the notations used by Crum and Hansen (1982a) will be used with the following conversions

$$\alpha = A_1 ; \quad \delta = \delta_1 ; \quad P_A / P_0 = \varepsilon ; \quad \alpha^2 K = B_2 .$$

Thus, Eq. (G1) can be written as

$$R / R_0 = 1 + \alpha \left(P_A / P_0 \right) \cos\left(\omega t + \delta \right) + \alpha^2 K \left(P_A / P_0 \right)^2 .$$

Appendix H: Formulas of rectified mass diffusion to the fourth order

In this appendix, we will derive the time averages $\langle R/R_0 \rangle$, $\langle (R/R_0)^4 \rangle$ and $\langle (R/R_0)^4 (P_{in}/P_0) \rangle$ required in the formulas of rectified diffusion (Eq. (3.6)) to the fourth order of (P_A/P_0). The complete solution of the equation of bubble motion is (referring to Appendix G)

$$R/R_0 = 1 + \alpha (P_A/P_0)\cos(\omega t + \delta) + \alpha^2 K (P_A/P_0)^2 + A_2 (P_A/P_0)^2 \cos(2\omega t + \delta_2). \quad \text{(H1)}$$

Comparing with Eq. (3.12), Eq. (H1) contains the second harmonic term. We perform the time averages within time duration T_b, such as

$$T_b = nT_0 = n(2\pi/\omega).$$

Here, T_0 is the period of the applied sound field and *n* is a small integer. The general Binomial theorem is

$$(x+y)^m = \sum_{k=0}^{\infty} C_m^k x^{m-k} y^k ,$$

where

$$C_m^k = \frac{m(m-1)...(m-k+1)}{k!}.$$

Here, m is a real number; k is an integer; x and y are two terms in the Binomial. Following relationships of trigonometric function will be used during the derivations,

$$d\left[\sin(\theta)\right]/dt = \cos(\theta);$$

$$d\left[\cos(\theta)\right]/dt = -\sin(\theta);$$

$$\sin(2\theta) = 2\sin(\theta)\cos(\theta);$$

$$\cos^2(\theta) = \frac{1}{2}\left[1+\cos(2\theta)\right];$$

$$\cos(3\theta) = 4\cos^3(\theta) - 3\cos(\theta);$$

$$\cos(\theta_1)\cos(\theta_2) = \frac{1}{2}\left[\cos(\theta_1+\theta_2)+\cos(\theta_1-\theta_2)\right];$$

$$\sin(\theta_1)\cos(\theta_2) = \frac{1}{2}\left[\sin(\theta_1+\theta_2)+\sin(\theta_1-\theta_2)\right].$$

Here, θ_1 and θ_2 are two arbitrary variables. To the fourth order of (P_A/P_0), one can find following expressions,

$$\left\langle R/R_0 \right\rangle = \frac{1}{T_b}\int_t^{t+T_b} \frac{R}{R_0} dt = 1+\alpha^2 K\left(P_A/P_0\right)^2, \quad \text{(H2)}$$

$$\begin{aligned}
\left\langle \left(R/R_0\right)^m \right\rangle &= \frac{1}{T_b}\int_t^{t+T_b} \left(\frac{R}{R_0}\right)^m dt \\
&\approx 1 + C_m^1 \alpha^2 K\left(P_A/P_0\right)^2 + C_m^2 \alpha^4 K^2 \left(P_A/P_0\right)^4 \\
&+ C_m^2 \left[1 + C_{m-2}^1 \alpha^2 K\left(P_A/P_0\right)^2\right] \frac{1}{2}\alpha^2 \left(P_A/P_0\right)^2 \\
&+ \frac{3}{8} C_m^4 \alpha^4 \left(P_A/P_0\right)^4 + C_m^1 C_{m-1}^2 \frac{1}{4}\alpha^2 A_2 \left(P_A/P_0\right)^4 \cos\left(2\delta-\delta_2\right) + \frac{1}{2} C_m^2 A_2^2 \left(P_A/P_0\right)^4.
\end{aligned}$$

(H3)

If $m=4$ in Eq. (H3), we obtain

$$\left\langle (R/R_0)^4 \right\rangle = 1 + [3+4K]\alpha^2 (P_A/P_0)^2$$

$$+\left[6\alpha^4 K^2 + 6\alpha^4 K + \frac{3}{8}\alpha^4 + 3\alpha^2 A_2 \cos(2\delta - \delta_2) + 3A_2^2\right](P_A/P_0)^4 . \quad \text{(H4)}$$

If $m = 4 - 3\kappa$ in Eq. (H3), we obtain

$$\left\langle (R/R_0)^{4-3\kappa} \right\rangle = 1 + \left[(4-3\kappa)K + \frac{(4-3\kappa)(3-3\kappa)}{4}\right]\alpha^2 (P_A/P_0)^2$$

$$+\left[\frac{(4-3\kappa)(3-3\kappa)}{2}\alpha^4 K^2 + \frac{(4-3\kappa)(3-3\kappa)(2-3\kappa)}{4}\alpha^4 K\right.$$

$$+\frac{3}{8}\frac{(4-3\kappa)(3-3\kappa)(2-3\kappa)(1-3\kappa)}{4!}\alpha^4$$

$$\left.+\frac{(4-3\kappa)(3-3\kappa)(2-3\kappa)}{8}\alpha^2 A_2 \cos(2\delta - \delta_2) + \frac{(4-3\kappa)(3-3\kappa)}{4}A_2^2\right](P_A/P_0)^4 .$$

(H5)

Although thermal damping was considered in Crum and Hansen (1982a), it was not included in the expression of instantaneous pressure inside bubbles (P_{in}) (Crum and Hansen, 1982a, Eq. (3)). If thermal damping is considered using an effective thermal viscosity, the instantaneous pressure inside bubbles is (Prosperetti, 1977, p.18)

$$P_{in} = (P_0 + 2\sigma/R_0)(R_0/R)^{3\kappa} - 4\mu_{th}\dot{R}/R .$$

Therefore,

$$\left\langle (R/R_0)^4 (P_{in}/P_0) \right\rangle = (1 + 2\sigma/P_0R_0)\left\langle (R/R_0)^{4-3\kappa} \right\rangle$$

$$+(4\mu_{th}\omega/P_0)(P_A/P_0)\left\langle (R/R_0)^3 \left[\alpha \sin(\omega t + \delta) + 2A_2 (P_A/P_0)\sin(2\omega t + \delta_2)\right]\right\rangle .$$

We notice that

$$\left\langle (R/R_0)^3\left[\alpha\sin(\omega t+\delta)+2A_2(P_A/P_0)\sin(2\omega t+\delta_2)\right]\right\rangle$$

$$=\frac{1}{T_b}\int_t^{t+T_b}\left\{\left[1+\alpha(P_A/P_0)\cos(\omega t+\delta)+\alpha^2K(P_A/P_0)^2\right]^3\right.$$

$$+3\left[1+\alpha(P_A/P_0)\cos(\omega t+\delta)+\alpha^2K(P_A/P_0)^2\right]^2A_2(P_A/P_0)^2\cos(2\omega t+\delta_2)$$

$$\left.+3\left[1+\alpha(P_A/P_0)\cos(\omega t+\delta)+\alpha^2K(P_A/P_0)^2\right]\left[A_2(P_A/P_0)^2\cos(2\omega t+\delta_2)\right]^2\right\}dt$$

$$\approx\frac{1}{T_b}\int_t^{t+T_b}\left\{1+3\alpha^2K(P_A/P_0)^2+3\alpha^4K^2(P_A/P_0)^4\right.$$

$$+3\left[1+2\alpha^2K(P_A/P_0)^2\right]\alpha(P_A/P_0)\cos(\omega t+\delta)$$

$$+3\left[1+\alpha^2K(P_A/P_0)^2\right]\left[\alpha(P_A/P_0)\cos(\omega t+\delta)\right]^2+\left[\alpha(P_A/P_0)\cos(\omega t+\delta)\right]^3$$

$$+3\left[1+2\alpha^2K(P_A/P_0)^2+2\alpha(P_A/P_0)\cos(\omega t+\delta)+\alpha^2(P_A/P_0)^2\cos^2(\omega t+\delta)\right]$$

$$\left.\bullet A_2(P_A/P_0)^2\cos(2\omega t+\delta_2)+3\left[A_2(P_A/P_0)^2\cos(2\omega t+\delta_2)\right]^2\right\}$$

$$\left[\alpha\sin(\omega t+\delta)+2A_2(P_A/P_0)\sin(2\omega t+\delta_2)\right]dt=0.$$

Therefore, thermal damping has no contribution to the $\left\langle (R/R_0)^4(P_{in}/P_0)\right\rangle$. Then, we obtain

$$\left\langle (R/R_0)^4(P_{in}/P_0)\right\rangle=(1+2\sigma/P_0R_0)\left\langle (R/R_0)^{4-3\kappa}\right\rangle$$

$$=(1+2\sigma/P_0R_0)\left\{1+\left[(4-3\kappa)K+\frac{(4-3\kappa)(3-3\kappa)}{4}\right]\alpha^2(P_A/P_0)^2\right.$$

$$+\left[\frac{(4-3\kappa)(3-3\kappa)}{2}\alpha^4K^2+\frac{(4-3\kappa)(3-3\kappa)(2-3\kappa)}{4}\alpha^4K\right.$$

$$+\frac{3}{8}\frac{(4-3\kappa)(3-3\kappa)(2-3\kappa)(1-3\kappa)}{4!}\alpha^4$$

$$\left.\left.+\frac{(4-3\kappa)(3-3\kappa)(2-3\kappa)}{8}\alpha^2A_2\cos(2\delta-\delta_2)+\frac{(4-3\kappa)(3-3\kappa)}{4}A_2^2\right](P_A/P_0)^4\right\}.$$

(H6)

Based on Eqs. (H2), (H4) and (H6), we have proofed that the harmonic term $A_2\cos(2\omega t+\delta_2)$ contributes to the time averages required by rectified diffusion with the fourth or higher orders of $\left(P_A / P_0\right)$. To the second order of $\left(P_A / P_0\right)$, Eqs. (H2), (H4) and (H6) reduce to those derived by Crum and Hansen (1982a), i.e. Eqs. (3.17)-(3.19).

Appendix I: Influences of initial conditions and solution of homogenous equation on rectified mass diffusion

In this appendix, the influences of initial conditions and solution of homogenous equation are discussed. Assuming that the initial conditions of the equation of bubble motion without damping are $R(0)=R_0$ and $\dot{R}(0)=0$, Eller and Flynn (1965, Eq. 52, p.501) obtained the solution as

$$R/R_0 = 1-\alpha\left[\sin(\omega t)-(\omega/\omega_0)\sin(\omega_0 t)\right](P_A/P_\infty)+\alpha^2 K(P_A/P_\infty)^2. \quad \text{(I1)}$$

The second term in bracket is the solution of homogenous equation (transient solution). Strictly speaking, Eq. (I1) only satisfies the boundary conditions to the first order of (P_A/P_0). To the second order of (P_A/P_0), $\dot{R}(0)=0$ but $R(0)=R_0\left[1+\alpha^2 K(P_A/P_0)^2\right]\neq R_0$.

According to Appendix C, the solution of inhomogeneous equation is the sum of the stable solution and the transient solution. If the damping mechanisms are considered in the bubble motion, the transient solution will decay exponentially with the time owing to the term $e^{-\beta_{tot} t}$ (referring to Appendix C). Noticing that the problems discussed in this thesis (e.g. rectified diffusion) are a rather slow process, we can safely ignore the transient solution. Then the solution is (Eller, 1969, Eq. 7)

$$R / R_0 = 1 + \alpha \left(P_A / P_0 \right) \cos\left(\omega t \right) + \alpha^2 K \left(P_A / P_0 \right)^2 . \qquad \text{(I2)}$$

Comparing with Eq. (3.12), only a phase δ is missing in Eq. (I2) but it will not influence the discussions in this thesis (e.g. rectified diffusion phenomenon). Based Eq. (I2), one can obtain the initial conditions as

$$\dot{R}\left(0\right) = 0 , \qquad \text{(I3)}$$

$$R\left(0\right) = R_0 \left[1 + \alpha \left(P_A / P_0 \right) + \alpha^2 K \left(P_A / P_0 \right)^2 \right]. \qquad \text{(I4)}$$

For convenience, Eller (1969, p.1248, Eq.6) employed the Eqs. (I3) and (I4) as the initial conditions in order that Eq. (I2) is strictly the solution of the equation of bubble motion with corresponding initial boundary conditions. Eq. (I2) was also used by Crum (1980, Eq.6) and was further modified by Crum and Hansen (1982a, Eq.7) with the phase δ included

$$R / R_0 = 1 + \alpha \left(P_A / P_0 \right) \cos\left(\omega t + \delta \right) + \alpha^2 K \left(P_A / P_0 \right)^2 . \qquad \text{(I5)}$$

For convenience, Eq. (I5) is employed in our work and the following initial conditions are used

$$R\left(0\right) = R_0 \left[1 + \alpha \left(P_A / P_0 \right) \cos\delta + \alpha^2 K \left(P_A / P_0 \right)^2 \right], \qquad \text{(I6)}$$

$$\dot{R}(0) = -\alpha\left(P_A / P_0\right)\omega \sin\delta \,. \tag{I7}$$

Appendix J: Solution of equation of bubble motion under acoustic field of dual frequency

In this appendix, the equation of bubble motion under acoustic field with dual frequency is solved based on perturbation method with direct series expansions. We assume that the solution of Keller's equation under dual-frequency acoustic field (Eqs. (2.1), (2.2) and (5.1)) is

$$\frac{R}{R_0} = 1 + \varepsilon x_1 + \varepsilon^2 x_2 + \dots \quad \text{(J1)}$$

Here, $\varepsilon = P_{A_1} / P_0$. In Eq. (J1), only stable solution is considered and the influences of initial conditions and the solution of homogenous equation (transient solution) have been discussed in Appendix C and I. Here, we only consider the terms up to the second order of ε. We denote $\varepsilon_{21} = P_{A_2} / P_{A_1}$. Thermal damping is currently ignored. Substituting Eq. (J1) into Eqs. (2.1), (2.2) and (5.1), we obtain,

$$R_0^2\left[\varepsilon\ddot{x}_1+\varepsilon^2\left(\ddot{x}_2+x_1\ddot{x}_1-\frac{R_0}{c_l}\dot{x}_1\ddot{x}_1\right)\right]+\frac{3}{2}R_0^2\varepsilon^2\dot{x}_1^2$$

$$-\rho_l^{-1}\varepsilon\left[-3\kappa\left(P_0+\frac{2\sigma}{R_0}\right)x_1+\frac{2\sigma}{R_0}x_1-P_0\cos(\omega_1 t)-P_0\varepsilon_{21}\cos(\omega_2 t)-4\mu_l\dot{x}_1\right]$$

$$-\rho_l^{-1}\frac{R_0}{c_l}\varepsilon\left[(-3\kappa)\left(P_0+\frac{2\sigma}{R_0}\right)\dot{x}_1+\frac{2\sigma}{R_0}\dot{x}_1+P_0\omega_1\sin(\omega_1 t)+P_0\varepsilon_{21}\omega_2\sin(\omega_2 t)-4\mu_l\ddot{x}_1\right]$$

$$-\rho_l^{-1}\varepsilon^2\left\{\left[-3\kappa\left(P_0+\frac{2\sigma}{R_0}\right)+\frac{2\sigma}{R_0}\right]x_2+\left[\frac{3\kappa(3\kappa+1)}{2}\left(P_0+\frac{2\sigma}{R_0}\right)-\frac{2\sigma}{R_0}\right]x_1^2\right.$$

$$\left.-4\mu_l(\dot{x}_2-x_1\dot{x}_1)\right\}$$

$$-\rho_l^{-1}\frac{R_0}{c_l}\varepsilon^2\left\{(-3\kappa)\left(P_0+\frac{2\sigma}{R_0}\right)\dot{x}_2+3\kappa(3\kappa+1)\left(P_0+\frac{2\sigma}{R_0}\right)x_1\dot{x}_1+\frac{2\sigma}{R_0}(\dot{x}_2-2x_1\dot{x}_1)\right.$$

$$-4\mu_l\left(-\dot{x}_1^2+\ddot{x}_2-x_1\ddot{x}_1\right)$$

$$+x_1\left[(-3\kappa)\left(P_0+\frac{2\sigma}{R_0}\right)\dot{x}_1+\frac{2\sigma}{R_0}\dot{x}_1+P_0\omega_1\sin(\omega_1 t)+P_0\varepsilon_{21}\omega_2\sin(\omega_2 t)-4\mu_l\ddot{x}_1\right]$$

$$\left.+\dot{x}_1\left[-3\kappa x_1\left(P_0+\frac{2\sigma}{R_0}\right)+\frac{2\sigma}{R_0}x_1-P_0\cos(\omega_1 t)-P_0\varepsilon_{21}\cos(\omega_2 t)-4\mu_l\dot{x}_1\right]\right\}=0.$$

(J2)

Based on Eq. (J2), the sum of all terms with order ε (or ε^2) in Eq. (J2) should be equal to zero respectively. To the first order of ε, the result is

$$\ddot{x}_1+2\beta_{tot}\dot{x}_1+\omega_0^2x_1=-\frac{P_0}{M\rho_l R_0^2}\left\{\left[\cos(\omega_1 t)-(\omega_1 R_0/c_l)\sin(\omega_1 t)\right]\right.$$

$$\left.+\varepsilon_{21}\left[\cos(\omega_2 t)-(\omega_2 R_0/c_l)\sin(\omega_2 t)\right]\right\} \quad \text{(J3)}$$

where

$$\beta_{tot}=\frac{2\mu_l}{M\rho_l R_0^2}+\frac{R_0}{2c_l}\omega_0^2; \quad \text{(J4)}$$

$$\omega_0^2=\frac{1}{M\rho_l R_0^2}\left[3\kappa\left(P_0+\frac{2\sigma}{R_0}\right)-\frac{2\sigma}{R_0}\right]; \quad \text{(J5)}$$

$$M=1+\frac{4\mu_l}{\rho_l R_0^2}\frac{R_0}{c_l}. \quad \text{(J6)}$$

The solution of Eq. (J3) can be written as

$$x_1 = A_{11}\cos\left(\omega_1 t+\delta_{11}\right)+A_{12}\cos\left(\omega_2 t+\delta_{12}\right), \tag{J7}$$

where

$$A_{11} = -\frac{P_0}{M\rho_l R_0^2}\left[\frac{1+\left(\omega_1 R_0 / c_l\right)^2}{\left(\omega_0^2-\omega_1^2\right)^2+4\beta_{tot}^2\omega_1^2}\right]^{1/2}; \tag{J8}$$

$$\delta_{11} = \tan^{-1}\left\{\frac{\left(\omega_1 R_0 / c_l\right)\left(\omega_0^2-\omega_1^2\right)-2\beta_{tot}\omega_1}{\left(\omega_0^2-\omega_1^2\right)+2\beta_{tot}\omega_1^2 R_0 / c_l}\right\}; \tag{J9}$$

$$A_{12} = -\frac{P_0\varepsilon_{21}}{M\rho_l R_0^2}\left[\frac{1+\left(\omega_2 R_0 / c_l\right)^2}{\left(\omega_0^2-\omega_2^2\right)^2+4\beta_{tot}^2\omega_2^2}\right]^{1/2}; \tag{J10}$$

$$\delta_{12} = \tan^{-1}\left\{\frac{\left(\omega_2 R_0 / c_l\right)\left(\omega_0^2-\omega_2^2\right)-2\beta_{tot}\omega_2}{\left(\omega_0^2-\omega_2^2\right)+2\beta_{tot}\omega_2^2 R_0 / c_l}\right\}. \tag{J11}$$

To the second order of ε in Eq. (J2), we obtain,

$$\ddot{x}_2+2\beta_{tot}\dot{x}_2+\omega_0^2 x_2 = -\frac{\Phi_0}{M}, \tag{J12}$$

where

$$\begin{aligned}\Phi_0 = {} & x_1\ddot{x}_1-\frac{R_0}{c_l}\dot{x}_1\ddot{x}_1+\frac{3}{2}\dot{x}_1^2-\frac{1}{\rho_l R_0^2}\left[\frac{3\kappa\left(3\kappa+1\right)}{2}\left(P_0+\frac{2\sigma}{R_0}\right)-\frac{2\sigma}{R_0}\right]x_1^2-\frac{4\mu_l}{\rho_l R_0^2}x_1\dot{x}_1\\ & -\frac{R_0}{c_l}\frac{1}{\rho_l R_0^2}\left[3\kappa\left(3\kappa+1\right)\left(P_0+\frac{2\sigma}{R_0}\right)x_1\dot{x}_1-\frac{4\sigma}{R_0}x_1\dot{x}_1-4\mu_l\left(-\dot{x}_1^2-x_1\ddot{x}_1\right)\right]\\ & -\frac{R_0}{c_l}\frac{x_1}{\rho_l R_0^2}\left[\left(-3\kappa\right)\left(P_0+\frac{2\sigma}{R_0}\right)\dot{x}_1+\frac{2\sigma}{R_0}\dot{x}_1+P_0\omega_1\sin\left(\omega_1 t\right)+P_0\varepsilon_{21}\omega_2\sin\left(\omega_2 t\right)-4\mu_l\ddot{x}_1\right]\\ & -\frac{R_0}{c_l}\frac{1}{\rho_l R_0^2}\dot{x}_1\left[-3\kappa\left(P_0+\frac{2\sigma}{R_0}\right)x_1+\frac{2\sigma}{R_0}x_1-P_0\cos\left(\omega_1 t\right)-P_0\varepsilon_{21}\cos\left(\omega_2 t\right)-4\mu_l\dot{x}_1\right].\end{aligned} \tag{J13}$$

The solution of Eq. (J12) can be written as

$$x_2 = B_2+A_{21}\cos\left(2\omega_1 t+\delta_{21}\right)+A_{22}\cos\left(2\omega_2 t+\delta_{22}\right)$$

$$+A_{23}\cos\left[\left(\omega_1+\omega_2\right)t+\delta_{23}\right]+A_{24}\cos\left[\left(\omega_1-\omega_2\right)t+\delta_{24}\right]. \quad \text{(J14)}$$

Here, B_2 is a constant. In Eq. (J14), the second and third terms are second harmonics. The frequencies of the fourth and fifth terms in Eq. (J14) are the sum or difference of the two driving frequencies. To the second order of ε, the second harmonic terms and the sum and difference terms in Eq. (J14) have no contribution to rectified diffusion (referring to Appendix K). Therefore, we only need to determine B_2 in Eq. (J14). The sum of constant terms (Φ_0') in Φ_0 can be written as

$$\Phi_0'=\frac{1}{4}\left(A_{11}^2\omega_1^2+A_{12}^2\omega_2^2\right)-\frac{1}{4}\frac{A_{11}^2+A_{12}^2}{\rho_l R_0^2}\left[3\kappa\left(3\kappa+1\right)\left(P_0+\frac{2\sigma}{R_0}\right)-\frac{4\sigma}{R_0}\right]. \quad \text{(J15)}$$

Then we obtain

$$B_2=-\Phi_0'/\omega_0^2 M\ . \quad \text{(J16)}$$

Now the solution of Keller's equation (Eqs. (2.1), (2.2) and (5.1)) under acoustic field with dual frequency has been obtained. For convenience, we set $M\approx 1$ in the following discussions. As shown in Chap. 2.1, Keller's equation reduces to Rayleigh-Plesset equation without liquid compressibility. If the effects of liquid compressibility (i.e. the terms involving c_l) are ignored, Eqs. (J8)-(J11) reduce to

$$\varepsilon A_{11}=-\frac{P_{A_1}}{\rho_l R_0^2\omega_0^2}\left[\left(1-\omega_1^2/\omega_0^2\right)^2+\left(4\mu_l\omega_1/\rho_l R_0^2\omega_0^2\right)^2\right]^{-1/2}, \quad \text{(J17)}$$

$$\delta_{11}=\tan^{-1}\left\{\frac{-2\beta_{tot}\omega_1}{\omega_0^2-\omega_1^2}\right\}, \quad \text{(J18)}$$

$$\varepsilon A_{12} = -\frac{P_{A_2}}{\rho_l R_0^2 \omega_0^2} \left[\left(1 - \omega_2^2 / \omega_0^2\right)^2 + \left(4\mu_l \omega_2 / \rho_l R_0^2 \omega_0^2\right)^2 \right]^{-1/2}, \qquad \text{(J19)}$$

$$\delta_{12} = \tan^{-1}\left\{ \frac{-2\beta_{tot}\omega_2}{\omega_0^2 - \omega_2^2} \right\}. \qquad \text{(J20)}$$

Eqs. (J17) and (J19) are exactly the same as those derived by Newhouse and Shankar (1984, Eq. (10)) based on Rayleigh-Plesset equation.

Appendix K: Time averages required in formulas of rectified mass diffusion under acoustic field of dual frequency

As shown in Appendix J, the solution of bubble motion under acoustic field with dual frequency is

$$R/R_0 = 1 + B_2\left(P_{A_1}/P_0\right)^2 + \left[A_{11}\cos\left(\omega_1 t + \delta_{11}\right) + A_{12}\cos\left(\omega_2 t + \delta_{12}\right)\right]\left(P_{A_1}/P_0\right)$$
$$\left\{A_{21}\cos\left(2\omega_1 t + \delta_{21}\right) + A_{22}\cos\left(2\omega_2 t + \delta_{22}\right) + A_{23}\cos\left[\left(\omega_1 + \omega_2\right)t + \delta_{23}\right]\right.$$
$$\left. + A_{24}\cos\left[\left(\omega_1 - \omega_2\right)t + \delta_{24}\right]\right\}\left(P_{A_1}/P_0\right)^2.$$

(K1)

This solution has included the second harmonics and terms with sum or difference of dual frequency. For convenience, we set $\omega_2 = n_0\omega_1$ (Here, n_0 is an integer). Therefore, the time averages are performed within time duration $T_b = nT_1$ (Here, $T_1 = 2\pi/\omega_1$ and n is an integer). The period of the sum of frequencies $\left(\omega_1 + \omega_2\right)$ is $T_+ = T_b/\left[n(n_0+1)\right]$. The period of the sum of frequencies $(\omega_2-\omega_1)$ is $T_-=T_b/[n(n_0-1)]$. Here, we only give the expressions of time averages to the second order of (P_{A_1}/P_0):

$$\langle R/R_0\rangle=\frac{1}{T_b}\int_t^{t+T_b}\frac{R}{R_0}dt=1+B_2\left(P_{A_1}/P_0\right)^2, \tag{K2}$$

$$\begin{aligned}
&\left\langle (R/R_0)^m\right\rangle=\frac{1}{T_b}\int_t^{t+T_b}(\frac{R}{R_0})^m dt\\
&\approx\frac{1}{T_b}\int_t^{t+T_b}\left\{\left[1+B_2\left(P_{A_1}/P_0\right)^2+A_{11}\cos\left(\omega_1 t+\delta_{11}\right)\left(P_{A_1}/P_0\right)\right.\right.\\
&\left.\left.+A_{12}\cos\left(\omega_2 t+\delta_{12}\right)\left(P_{A_1}/P_0\right)\right]^m\right\}dt\\
&+\frac{1}{T_b}\int_t^{t+T_b}C_m^1\left\{A_{21}\cos\left(2\omega_1 t+\delta_{21}\right)+A_{22}\cos\left(2\omega_2 t+\delta_{22}\right)+A_{23}\cos\left[\left(\omega_1+\omega_2\right)t+\delta_{23}\right]\right.\\
&\left.+A_{24}\cos\left[\left(\omega_1-\omega_2\right)t+\delta_{24}\right]\right\}\left(P_{A_1}/P_0\right)^2 dt\\
&\approx\frac{1}{T_b}\int_t^{t+T_b}\left\{\left[1+B_2\left(P_{A_1}/P_0\right)^2\right]^m\right.\\
&+C_m^1\left[1+B_2\left(P_{A_1}/P_0\right)^2\right]^{m-1}\left[A_{11}\cos\left(\omega_1 t+\delta_{11}\right)+A_{12}\cos\left(\omega_2 t+\delta_{12}\right)\right]\left(P_{A_1}/P_0\right)\\
&\left.+C_m^2\left[1+B_2\left(P_{A_1}/P_0\right)^2\right]^{m-2}\left[A_{11}\cos\left(\omega_1 t+\delta_{11}\right)+A_{12}\cos\left(\omega_2 t+\delta_{12}\right)\right]^2\left(P_{A_1}/P_0\right)^2\right\}dt\\
&\approx 1+C_m^1 B_2\left(P_{A_1}/P_0\right)^2+\frac{1}{2}C_m^2\left(A_{11}^2+A_{12}^2\right)\left(P_{A_1}/P_0\right)^2.
\end{aligned} \tag{K3}$$

If m=4 in Eq. (K3), we obtain

$$\left\langle (R/R_0)^4\right\rangle=1+\left[4B_2+3\left(A_{11}^2+A_{12}^2\right)\right]\left(P_{A_1}/P_0\right)^2. \tag{K4}$$

If $m=4-3\kappa$ in Eq. (K3), we obtain

$$\begin{aligned}
&\left\langle (R/R_0)^4(P_{in}/P_0)\right\rangle=\left(1+\frac{2\sigma}{P_0R_0}\right)\left\langle (R/R_0)^{4-3\kappa}\right\rangle=\\
&\left(1+\frac{2\sigma}{P_0R_0}\right)\left\{1+\left[\left(4-3\kappa\right)B_2+\frac{\left(4-3\kappa\right)\left(3-3\kappa\right)}{4}\left(A_{11}^2+A_{12}^2\right)\right]\left(P_{A_1}/P_0\right)^2\right\}.
\end{aligned} \tag{K5}$$

Based on Eqs. (K2), (K4) and (K5), the contributions of second harmonics and sum or difference terms to rectified diffusion is with the fourth or higher orders of (P_{A_1} / P_0).

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