Results in Physics 25 (2021) 104193

Contents lists available at ScienceDirect

Results in Physics

journal homepage: www.elsevier.com/locate/rinp

A fourth-order approximation Rayleigh-Plesset equation written in volume variation for an adiabatic-gas bubble in an ultrasonic field: Derivation and numerical solution

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ARTICLE INFO

Keywords:
Nonlinear acoustics
Bubble dynamics
Mathematical modeling
Nonlinear ordinary differential equation
Numerical solution

ABSTRACT

The derivation of a nonlinear ordinary differential equation for modeling the nonlinear oscillations of a gas bubble placed in an ultrasonic field is performed in terms of bubble-volume variations up to the fourth-order approximation. The equation, written within the Rayleigh-Plesset framework, is solved through numerical approximations. Results from simulations are compared to data obtained from the classic second-order approximation equation derived in the 1960–70's, usually used in this framework, and from the third-order approximation equation derived in the 1990's. This comparison shows that the fourth-order approximation allows us to observe the nonlinear behavior of the bubble at high finite amplitude, which differs from the other approximations when the nonlinearity of the phenomenon is higher, i.e., when the driving acoustic frequency is close to the bubble resonance.

1. Introduction

Among the applications of ultrasound, sonochemistry and other acoustic cavitation based techniques and industrial processes (surface cleaning, waste water treatment…) have been the topics of many theoretical and applied researches [1–4]. These applications rely on the nonlinear behavior of oscillating gas bubbles under the effect of high-finite amplitude ultrasound. It is thus fundamental to increase our knowledge about this nonlinear behavior and its effects (Bjerknes forces, acoustic streaming, liberation of radicals) to improve the efficiency and reliability of these techniques. In this framework, we propose here the derivation of a bubble differential equation that includes the nonlinear terms up to the fourth order and responds to the Rayleigh-Plesset assumptions.

The Rayleigh-Plesset approximation describes the behavior of oscillating gas bubbles in an acoustic field assuming hypothesis that simplify the derivation of the related differential equations. Most of the differential equations that have been derived in this context consider the bubble radius as dependent variable (radius frame) [5–8]. These equations are able to describe the nonlinear oscillations and collapse of the bubble. An alternative valid for moderate amplitudes exists in the literature. Firstly derived by Zabolotskaya and Soluyan in the 1960–70's [9,10], and improved in the 1990's [11], these models assume the bubble volume variation as the dependent variable to describe the behavior of the bubble. In these approximations, the volume frame, some added restrictions are considered [9–12]. The main restrictive hypothesis is that the volume variation must be much lower than the initial volume of the bubble. Many of these restrictions are got rid in Ref. [13], in which a bubble volume variation equation is derived without those approximations, which also includes an extension of the dissipation term, but, as far as we know, to date no solution has been given to that complex differential equation.

In this framework, here we aim to derive an intermediate model that would improve the second and third-order approximations [9–12], and therefore the bubble response submitted to a pressure field. In this sense, we add the fourth-order term in the adiabatic gas law in the development of the volume-frame Rayleigh-Plesset equation in Section 2.1. Once this equation is obtained, we use a numerical method to track the nonlinear behavior of the bubble impinged by an ultrasonic wave in Section 2.2. In Section 3, we study the effect of this new term by considering several ultrasonic fields of different amplitudes. Section 4 gives the conclusions of this work.

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https://doi.org/10.1016/j.rinp.2021.104193
Received 22 September 2020; Accepted 9 April 2021
Available online 20 April 2021

2. Material and methods

2.1. Derivation of the bubble equation

The derivation of the ordinary differential equation, Eq. (19), is described in this section. In addition to the approximations made for this derivation (see the development below), in the Rayleigh-Plesset framework used for the description of the bubble oscillations due to an acoustic field we assume some physical restrictions: the bubble has a spherical shape and its pulsation are spherically symmetric; the bubble radius is small compared to the wavelength of the acoustic field; the bubble has a constant gas content, without vapor; the spatial conditions within the bubble are uniform; the thermal damping is neglected; the nonlinearity of the problem is only due to the bubble behavior; the bubble does not radiate sound itself; the surface tension at the gas–liquid boundary is not taken into account. Moreover, in this work we also neglect the buoyancy (no body force considered), Bjerknes, and viscous drag forces.

Thus, we now consider a bubble with gas content (no vapor) in a liquid. The following adiabatic law is assumed for the definition of the gas pressure inside the bubble, P_b:

$$P_b V_b^{\gamma} = P_{b0} V_{b0}^{\gamma},\qquad P_b = P_{b0}(V_{b0}/V_b)^{\gamma} = P_{b0}(R_{b0}/R_b)^{3\gamma}, \tag{1}$$

in which γ is the ratio of specific heats of the gas, $P_{b0} = \rho_b c_b^2/\gamma$ is the gas pressure inside the bubble at rest (equilibrium initial natural state of repose), where ρ_b and c_b are the density and sound speed of the gas at the equilibrium state, R_{b0} and $V_{b0} = 4\pi R_{b0}^3/3$ are the radius and volume of the bubble at rest (equilibrium initial natural state of repose), $R_b(t) = R_{b0} + r(t)$ and $V_b(t) = V_{b0} + v(t) = 4\pi R_b^3(t)/3$ are the radius and volume of the bubble at time $t, r(t)$ and $v(t)$ are the radius and volume variations of the bubble (from their initial value in repose, due to an external excitation such as an acoustic field), which are real-value functions of t that are sufficiently smooth to be expanded in Taylor series up to an indefinite order of derivation. The current gas pressure inside the bubble, at time t, can be written $P_b(t) = P_{b0} + p(t)$, where $p(t)$ is the "excess" pressure inside the bubble from its initial value in repose due to an external excitation, i.e., the acoustic pressure. We apply the exponent Taylor series expansion up to the fourth order, neglecting the terms from the fifth order, to approximate the adiabatic gas law written in terms of volume, Eq. (1), provided that $v \ll V_{b0}$ (bubble oscillations assumed to be of moderate amplitude):

$$\frac{P_b}{P_{b0}} = \left(\frac{V_{b0}}{V_b}\right)^{\gamma} = \left(\frac{V_b}{V_{b0}}\right)^{-\gamma} = \left(\frac{V_{b0}+v}{V_{b0}}\right)^{-\gamma} = \left(1+\frac{v}{V_{b0}}\right)^{-\gamma}$$
$$\approx 1 - \gamma\frac{v}{V_{b0}} + \frac{(-\gamma)(-\gamma-1)}{2}\left(\frac{v}{V_{b0}}\right)^2 + \frac{(-\gamma)(-\gamma-1)(-\gamma-2)}{6}\left(\frac{v}{V_{b0}}\right)^3 + \frac{(-\gamma)(-\gamma-1)(-\gamma-2)(-\gamma-3)}{24}\left(\frac{v}{V_{b0}}\right)^4, \tag{2}$$

which leads to, assuming the error of approximation of order $O(v/V_{b0})^5$ ($\approx \longrightarrow =$),

$$P_b - P_{b0} = -P_{b0}\gamma\frac{v}{V_{b0}} + P_{b0}\frac{\gamma^2+\gamma}{2}\left(\frac{v}{V_{b0}}\right)^2 - P_{b0}\frac{\gamma^3+3\gamma^2+2\gamma}{6}\left(\frac{v}{V_{b0}}\right)^3 + P_{b0}\frac{\gamma^4+6\gamma^3+11\gamma^2+6\gamma}{24}\left(\frac{v}{V_{b0}}\right)^4. \tag{3}$$

We now multiply the last expression by $4\pi R_{b0}/\rho_l$, where ρ_l is the density of the liquid at the equilibrium state, and use the identity $V_{b0} = 4\pi R_{b0}^3/3$, i.e., $4\pi R_{b0}/V_{b0} = 3/R_{b0}^2, 4\pi R_{b0}/V_{b0}^2 = 3/R_{b0}^2 V_{b0}, 4\pi R_{b0}/V_{b0}^3 = 3/R_{b0}^2 V_{b0}^2$, and $4\pi R_{b0}/V_{b0}^4 = 3/R_{b0}^2 V_{b0}^3$, to obtain

$$\frac{4\pi R_{b0}}{\rho_l}(P_b - P_{b0}) = -\frac{3\gamma P_{b0}}{\rho_l R_{b0}^2}v + \frac{3P_{b0}(\gamma^2+\gamma)}{2\rho_l R_{b0}^2 V_{b0}}v^2 - \frac{3P_{b0}(\gamma^3+3\gamma^2+2\gamma)}{6\rho_l R_{b0}^2 V_{b0}^2}v^3 + \frac{3P_{b0}(\gamma^4+6\gamma^3+11\gamma^2+6\gamma)}{24\rho_l R_{b0}^2 V_{b0}^3}v^4. \tag{4}$$

Using the resonance frequency of the bubble, $\omega_b = \sqrt{3\gamma P_{b0}/\rho_l R_{b0}^2}$ [14], and factorizing γ from the three nonlinear terms, we obtain

$$\frac{4\pi R_{b0}}{\rho_l}(P_b - P_{b0}) = -\omega_b^2 v + \frac{\omega_b^2(\gamma+1)}{2V_{b0}}v^2 - \frac{\omega_b^2(\gamma^2+3\gamma+2)}{6V_{b0}^2}v^3 + \frac{\omega_b^2(\gamma^3+6\gamma^2+11\gamma+6)}{24V_{b0}^3}v^4. \tag{5}$$

The nonlinear terms are due to the nonlinearity of the adiabatic state equation of gas in the bubble. We now set the corresponding nonlinear parameters, the known second and third-order parameters,

$$a_2 = \frac{\omega_b^2(\gamma+1)}{2V_{b0}}, \quad a_3 = \frac{\omega_b^2(\gamma^2+3\gamma+2)}{6V_{b0}^2}, \tag{6}$$

and the new defined fourth-order parameter,

$$a_4 = \frac{\omega_b^2(\gamma^3+6\gamma^2+11\gamma+6)}{24V_{b0}^3}. \tag{7}$$

Their units are Hz^2/m^3 for a_2, Hz^2/m^6 for a_3, and Hz^2/m^9 for a_4. Eq. (5) can finally be written in the following fourth-degree expression,

$$\frac{4\pi R_{b0}}{\rho_l}(P_b - P_{b0}) = -\omega_b^2 v + a_2 v^2 - a_3 v^3 + a_4 v^4. \tag{8}$$

The Rayleigh-Plesset equation written in radius variable for the description of bubble pulsations is [5,10,12,6],

$$R_b R_b'' + \frac{3}{2}R_b'^2 + 4\nu_l \frac{R_b'}{R_b} = \frac{(P_w - P_\infty)}{\rho}, \tag{9}$$

where ν_l is the cinematic viscosity of the liquid, ρ is the density of the medium (liquid and gas), P_w is the pressure at the wall of the bubble, and P_∞ is the pressure in the infinite limit of the space domain surrounding the bubble. Note that the $'$ and $''$ symbols mean first and second-order time derivatives, respectively, in the entire document. Now, assuming that $P_w = P_b$ (homogeneous pressure inside the bubble from the center up to the wall), that $P_\infty = P_l$, the pressure in the liquid, and that $\rho = \rho_l$ (the gas fraction in the liquid is extremely low), the right-hand side of Eq. (9) can be written $(P_b - P_l)/\rho_l$. Moreover, $P_l = P_{l0} + p_a$, i.e., the sum of the pressure in the liquid in repose and the acoustic pressure. Since the equilibrium of the bubble wall at rest yields $P_{l0} = P_{b0}$, we obtain $(P_b - P_l)/\rho_l = (P_b - P_{b0} - p_a)/\rho_l$. Thus, Eq. (9), once multiplied by $4\pi R_{b0}$, is written in the following way:

$$4\pi R_{b0}\left(R_b R_b'' + \frac{3}{2}R_b'^2 + 4\nu_l \frac{R_b'}{R_b}\right) = \frac{4\pi R_{b0}}{\rho_l}(P_b - P_{b0}) - \frac{4\pi R_{b0}}{\rho_l}p_a. \tag{10}$$

The expression (8) of the first term of the right-hand side of the last equation is introduced to write:

$$4\pi R_{b0}\left(R_b R_b'' + \frac{3}{2}R_b'^2 + 4\nu_l \frac{R_b'}{R_b}\right) = -\omega_b^2 v + a_2 v^2 - a_3 v^3 + a_4 v^4 - \frac{4\pi R_{b0}}{\rho_l}p_a. \tag{11}$$

The left-hand side of this equation must be now changed into a v variable expression. To this purpose we use the relations $V_b = 4\pi R_b^3/3$, $V_b' = 4\pi R_b^2 R_b'$, $V_b'' = 4\pi R_b(2R_b'^2 + R_b R_b'')$, which allow us to rewrite the left-hand side of the previous relation in terms of V_b:

$$R_b R_b'' + \frac{3}{2}R_b'^2 + 4\nu_l \frac{R_b'}{R_b} = \frac{1}{4\pi}\left(\frac{4\pi}{3V_b}\right)^{1/3}\left(V_b'' - \frac{V_b'^2}{6V_b}\right) + \frac{4\nu_l}{3}\frac{V_b'}{V_b}. \tag{12}$$

Since $V_b = V_{b0} + v$, we have $V_b' = v'$ and $V_b'' = v''$. On the other hand, we can write $V_b^{-1/3} = V_{b0}^{-1/3}(1 + v/V_{b0})^{-1/3}$. We approximate the second term through the linear exponent Taylor series expansion, providing that $v \ll V_{b0}$, which implies $V_b = V_{b0} + v \approx V_{b0}$, neglecting the terms from the second order, $V_b^{-1/3} \approx V_{b0}^{-1/3}(1 - (1/3)(v/V_{b0}))$, which is by assuming the error of approximation of order $O(v/V_{b0})^2$ ($\approx \longrightarrow =$), $V_b^{-1/3} = 1/V_{b0}^{1/3} - 1/(3V_{b0}^{4/3})v$. Eq. (12) can now be written,

$$R_b R_b'' + \frac{3}{2}R_b'^2 + 4\nu_l \frac{R_b'}{R_b} = \frac{1}{4\pi}\left(\frac{4\pi}{3}\right)^{1/3}\left(\frac{1}{V_{b0}^{1/3}} - \frac{1}{3V_{b0}^{4/3}}v\right)\left(v'' - \frac{v'^2}{6(V_{b0}+v)}\right) + \frac{4\nu_l}{3}\frac{v'}{V_{b0}+v}. \tag{13}$$

As seen in the above paragraph, we have $v \ll V_{b0}$ and $V_b = V_{b0} + v \approx V_{b0}$, which leads us to write:

$$R_b R_b'' + \frac{3}{2}R_b'^2 + 4\nu_l \frac{R_b'}{R_b} \approx \frac{1}{4\pi}\left(\frac{4\pi}{3}\right)^{1/3}\left(\frac{v''}{V_{b0}^{1/3}} - \frac{v'^2}{6V_{b0}^{4/3}} - \frac{vv''}{3V_{b0}^{4/3}} + \frac{vv'^2}{18V_{b0}^{7/3}}\right) + \frac{4\nu_l}{3}\frac{v'}{V_{b0}}. \tag{14}$$

Assuming the error of the approximation $V_b = V_{b0} + v \approx V_{b0}$ ($\approx \longrightarrow =$), and since $V_{b0} = 4\pi R_{b0}^3/3$, the above expression yields,

$$R_b R_b'' + \frac{3}{2}R_b'^2 + 4\nu_l \frac{R_b'}{R_b} = \frac{1}{4\pi R_{b0}}\left(v'' - \frac{1}{6V_{b0}}\left(v'^2 + 2vv''\right) + \frac{vv'^2}{18V_{b0}^2}\right) + \frac{\nu_l}{\pi R_{b0}^3}v'. \tag{15}$$

Thus, the left-hand side of Eq. (11) can be written,

$$4\pi R_{b0}\left(R_b R_b'' + \frac{3}{2}R_b'^2 + 4\nu_l \frac{R_b'}{R_b}\right) = v'' - \frac{1}{6V_{b0}}\left(v'^2 + 2vv''\right) + \frac{vv'^2}{18V_{b0}^2} + \frac{4\nu_l}{R_{b0}^2}v', \tag{16}$$

and we neglect the third-order term to obtain, after assuming this last approximation ($\approx \longrightarrow =$),

$$4\pi R_{b0}\left(R_b R_b'' + \frac{3}{2}R_b'^2 + 4\nu_l \frac{R_b'}{R_b}\right) = v'' - \frac{1}{6V_{b0}}\left(v'^2 + 2vv''\right) + \frac{4\nu_l}{R_{b0}^2}v'. \tag{17}$$

Eq. (11) can finally be written exclusively in terms of variable v as

$$v'' - \frac{1}{6V_{b0}}\left(v'^2 + 2vv''\right) + \frac{4\nu_l}{R_{b0}^2}v' = -\omega_b^2 v + a_2 v^2 - a_3 v^3 + a_4 v^4 - \frac{4\pi R_{b0}}{\rho_l}p_a, \tag{18}$$

in which the first two nonlinear terms $v'^2 + 2vv''$ are attributable to the dynamic nonlinearity of the bubble, and for which the corresponding well-known nonlinear parameter is $b = 1/6V_{b0}$. After setting the viscous damping coefficient $\delta = 4\nu_l/\omega_b R_{b0}^2$ and the well-known constant $\eta = 4\pi R_{b0}/\rho_l$, and by arranging the equation with the dynamic and damping terms on the left-hand side and the nonlinear and forcing terms on the right-hand side, we finally obtain the following new Rayleigh-Plesset differential equation of fourth order in variable v, which governs the nonlinear behavior of a bubble in an acoustic field in terms of bubble volume variation:

$$v'' + \delta\omega_b v' + \omega_b^2 v = b\left(2vv'' + v'^2\right) + a_2 v^2 - a_3 v^3 + a_4 v^4 - \eta p_a. \tag{19}$$

2.2. Numerical solution of the bubble equation

Eq. (19) is a nonlinear inhomogeneous second-order ordinary differential equation. $v(t)$ is the dependent variable, t is the time (independent variable). $p_a(t)$ is a known function that represents the acoustic pressure source exciting the bubble. We solve the differential system formed by Eq. (19) and the Cauchy conditions, which state the non-perturbation of the bubble at the start of the experiment, in which T is the lifetime of the experiment,

$$\begin{cases} v'' + \delta\omega_b v' + \omega_b^2 v = b\left(2vv'' + v'^2\right) \\ +a_2 v^2 - a_3 v^3 + a_4 v^4 - \eta p_a, \quad 0 < t < T, \\ v(0) = 0, \\ v'(0) = 0. \end{cases} \tag{20}$$

The second-order ordinary differential equation is reduced to a first-order system of two coupled ordinary differential equations by setting the new variable $u = v'$, which yields

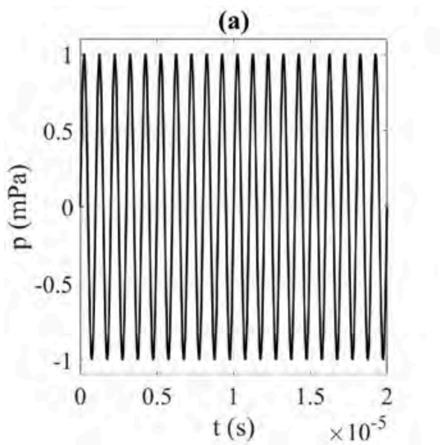

Fig. 1. 4RPv Model. $f = f_b$. Infinitesimal amplitude. Pressure source signal (a), bubble volume variation (b).

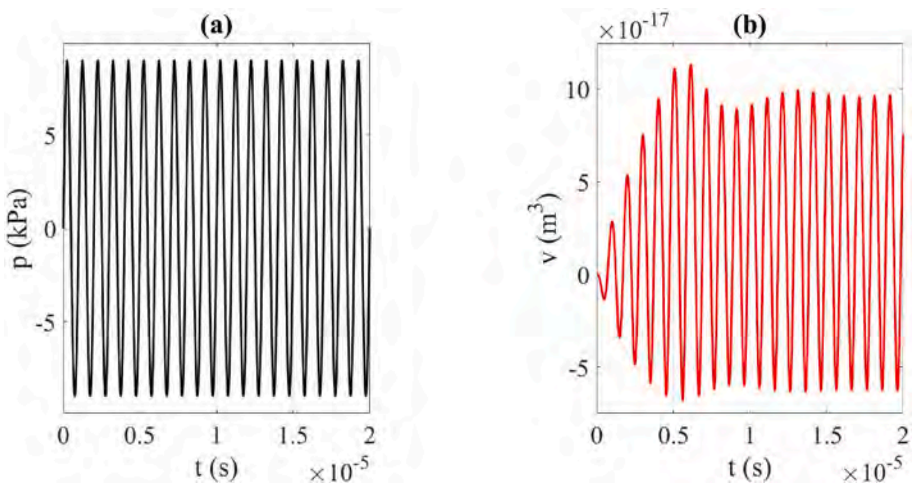

Fig. 2. 4RPv Model. $f = f_b$. High finite amplitude. Pressure source signal (a), bubble volume variation (b).

$$\begin{cases} v' = u, 0 < t < T, \\ u' + \delta\omega_b u + \omega_b^2 v = b\left(2vu' + u^2\right) + a_2 v^2 - a_3 v^3 + a_4 v^4 - \eta p_a, \quad 0 < t < T, \\ v(0) = 0, \\ u(0) = 0. \end{cases} \tag{21}$$

This differential system is written in the vector form,

$$\begin{cases} \overline{v}'\left(t\right) = \overline{f}\left(t, \overline{v}\right), \quad 0 < t < T, \\ \overline{v}\left(0\right) = \overline{0}, \end{cases} \tag{22}$$

in which

$$\begin{aligned} &\overline{v}(t) = (v(t), u(t))^t, \\ &\overline{f}\left(t, \overline{v}\right) = (f_v(t, v, u), f_u(t, v, u))^t, \\ &f_v(t, v, u) = u, \\ &f_u\left(t, v, u\right) = \left(-\delta\omega_b u - \omega_b^2 v + bu^2 + a_2 v^2 - a_3 v^3 + a_4 v^4 - \eta p_a\right)/(1 - 2bv), \\ &\overline{0} = (0, 0)^t. \end{aligned} \tag{23}$$

Before solving the first-order Cauchy problem, System (22), variables are set without dimension, to limit their variation range: $\widetilde{t} = \omega t, \widetilde{v} = v/V_{b0}, \widetilde{u} = u/\omega V_{b0}, \widetilde{p_a} = p_a/p_0$, where ω is the frequency of the acoustic wave and p_0 is its amplitude. The resulting system is then solved by applying the vector Runge–Kutta method of fourth-order approximation [15]. To this end, the continuous dimensionless time domain $0 \leqslant \widetilde{t} \leqslant \omega T$ is discretized using the step τ. The set $\{\widetilde{t}_n\}_{n=1}^{N}$ is then obtained, with $N = (\omega T - 0)/\tau + 1$. The approximation of the solution of System (22) is evaluated through the iterative process at each point $\widetilde{t}_n$ of that set, starting from the initial data $\overline{v}_1 = \overline{0}$. Note that the approximate value of the variable at point $\widetilde{t}_n$ is denoted by sub-index n.

3. Results and discussion

The equation derived in Section 2.1 and solved in Section 2.2 is tested (Matlab® R2017a environment) in this section by comparing the results obtained to the classic Rayleigh-Plesset equations which takes only nonlinear terms up to the second and third-order into account. To this purpose we consider an air bubble of radius $R_{b0} = 3.3663\,\mu m$ in water, of resonance $f_b = \omega_b/2\pi = 1\,MHz$. An ultrasonic perturbation (a continuous sine ultrasonic pressure wave of amplitude p_0 and frequency $\omega = 2\pi f$, $p_a(t) = p_0 sin(\omega t)$) propagating in the liquid impinges the bubble. The parameters of the biphasic medium are $\rho_l = 1000\,kg/m^3$, $\nu_l = 1.43 \times 10^{-6}\,m^2/s$, $\rho_b = 1.29\,kg/m^3$, $c_b = 340\,m/s$, $\gamma = 1.4$, $P_{b0} = 106.5171\,kPa$, $\eta = 4.2302 \times 10^{-8}\,m^4/kg$, $\delta = 0.0805$, $b = 1.0430 \times 10^{15}\,/m^3$, $a_2 = 2.9648 \times 10^{29}\,Hz^2/m^3$, $a_3 = 2.1028 \times 10^{45}\,Hz^2/m^6$, and $a_4 = 1.4476 \times 10^{61}\,Hz^2/m^9$. It is interesting to emphasize the high values of the nonlinear parameters (see Fig. 4).

Three different models are used and compared in this section to

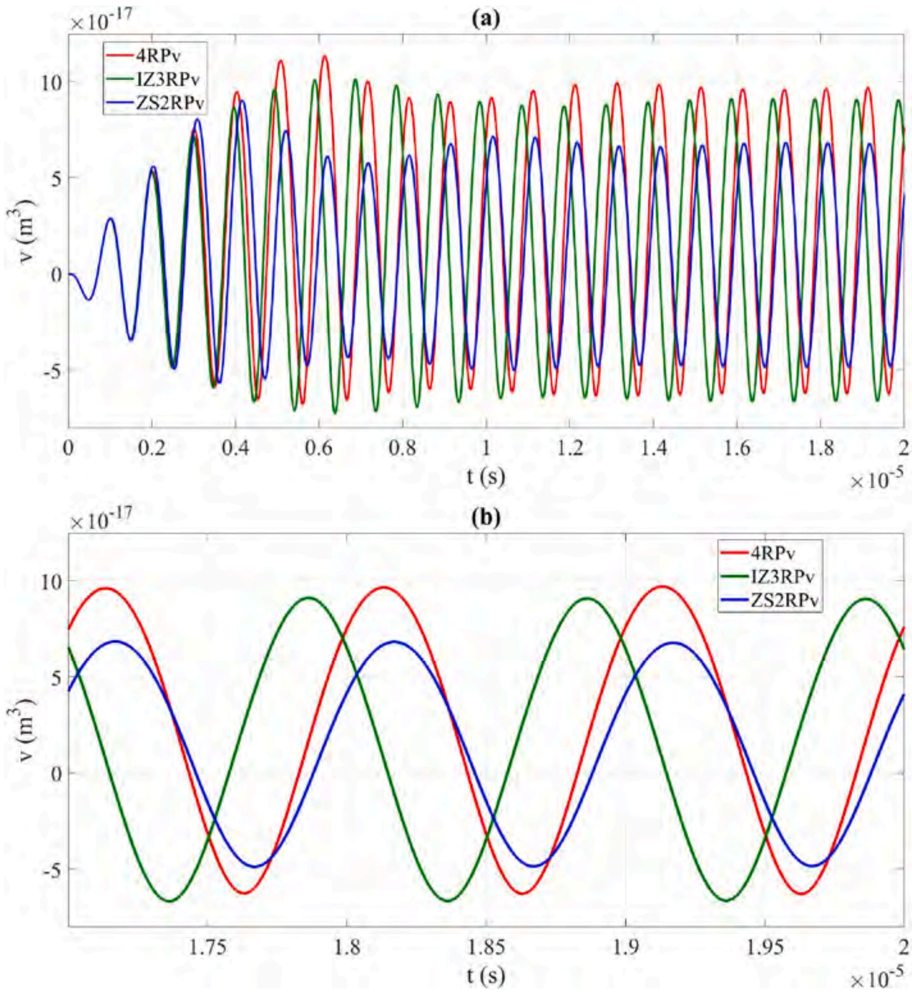

Fig. 3. Comparison of 4RPv, IZ3RPv, and ZS2RPv Models. $f = f_b$. High finite amplitude. Bubble volume variation (a), bubble volume variation during the last periods of the study (b).

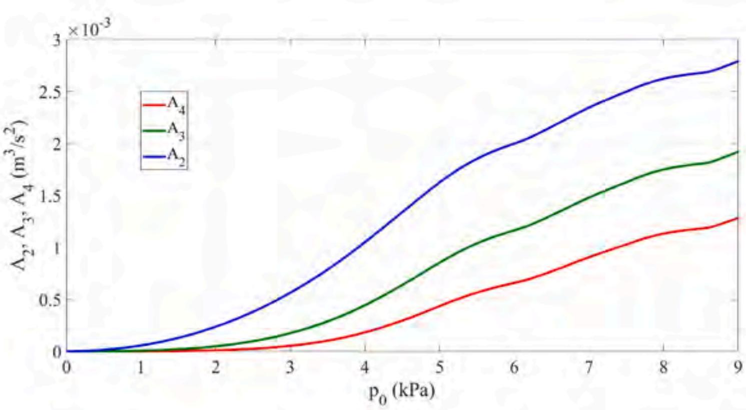

Fig. 4. 4RPv Model. $f = f_b$. Nonlinear terms A_2, A_3, A_4 vs. p_0.

determine the importance of the new nonlinear term in Eq. (19): the classic Rayleigh-Plesset equation in the volume frame derived by Zabolotskaya and Soluyan [10,12], which considers only nonlinear terms up to the second order, i.e., $a_2 \neq 0$ and $a_3 = a_4 = 0$ in Eq. (19), denoted by ZS2RPv Model; its improvement of third order approximation derived by Ilinskii and Zabolotskaya [11], for which $a_2 \neq 0$, $a_3 \neq 0$, and $a_4 = 0$ in Eq. (19), denoted by IZ3RPv Model; the new equation derived in 2.1, which considers all the nonlinear terms up to the fourth order, i.e., $a_2 \neq 0$, $a_3 \neq 0$, and $a_4 \neq 0$ in Eq. (19), denoted by 4RPv Model. It must be noted that in relation to Ref. [11] we do not carry out a comparison with the results presented in this reference since they include the interaction of several bubbles and the radiation of sound by bubbles, but we compare our results (fourth-order) to the ones obtained from an equivalent equation to the one developed in this

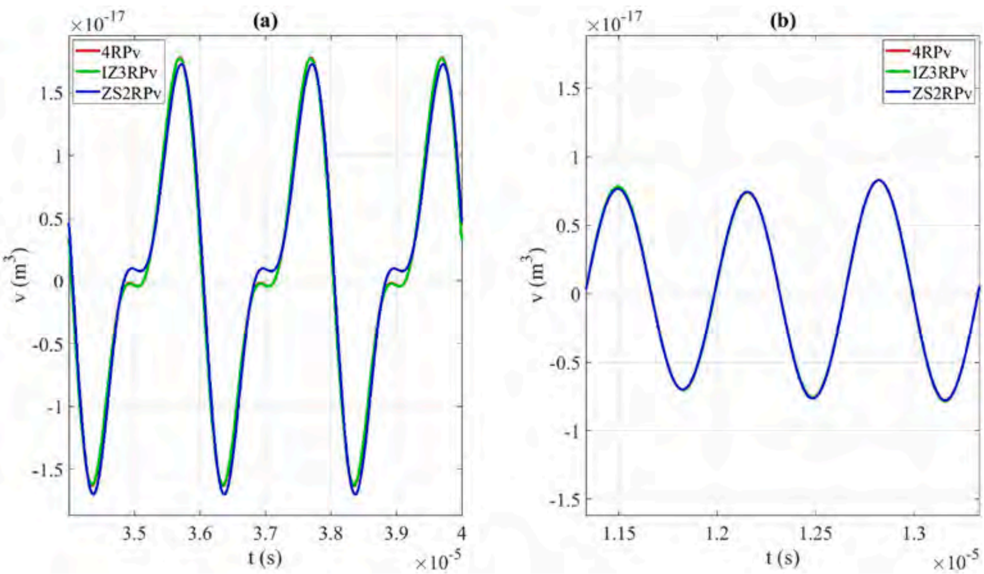

Fig. 5. Comparison of 4RPv, IZ3RPv, and ZS2RPv Models. High finite amplitude. Bubble volume variation during the last periods of the study. $f = 0.5f_b$ (a), $f = 1.5f_b$ (b).

reference in terms of nonlinearity due to the adiabatic gas law (third-order).

The ultrasonic wave excites the bubble at its resonance, $f = f_b$, and two amplitudes are contemplated, a high finite-amplitude $p_0 = 9\,kPa$ and an infinitesimal amplitude $p_0 = 1\,mPa$, studied during $T = 20/f$ (see Fig. 1). $\tau = 1 \times 10^{-5}$ in all the cases presented in this section.

In Figs. 1 and 2 are represented the pressure source signal (a) and the bubble volume variation (b) for an infinitesimal source amplitude (Fig. 1) and a high finite source amplitude (Fig. 2) obtained from 4RPv Model. Fig. 3 displays the bubble volume variation from 4RPv Model compared to the results given by ZS2RPv and IZ3RPv Models.

For infinitesimal amplitudes, Fig. 1, the nonlinear parameters a_2,a_3, a_4, and b multiply very small v-values in Eq. (19), the nonlinear terms in this equation are negligible, and the response of the system is linear (sine and symmetric signal). Note that this result is identical from ZS2RPv and IZ3RPv Models. However, for high finite amplitudes, Fig. 2, the signal shown is nonlinearly distorted and asymmetric in relation to the axis $v = 0$, since these nonlinear terms cannot be neglected, as the nonlinear parameters a_2,a_3, a_4, and b are multiplied by large v-values in Eq. (19).

It can be seen in Fig. 3 that the asymmetry of the signal is more pronounced with 4RPv Model than with IZ3RPv Model. The contribution of the coefficient a_4 in Eq. (19) is thus clear. It must be noted that the change of phase between 4RPv and ZS2RPv Models, which are almost in phase, in relation to IZ3RPv Model may be most likely due to a change of the acoustic stiffness factor of the bubble since the nonlinear terms a_2,a_3, and a_4 modify the relation between P_b and v when a higher-order approximation is carried out in the development of the adiabatic gas law, Eq. (2).

Fig. 4 represents the nonlinear terms, $A_2 = a_2 v_x^2$, $A_3 = a_3 v_x^3$, and $A_4 = a_4 v_x^4$, obtained varying the source amplitude p_0 from 1 Pa to 9 kPa, in which v_x denotes the maximal value of v reached during the last three periods of the study. This diagram allows us to observe the importance that each nonlinear term due to the adiabatic law has on the solution as the source amplitude increases. The diagram shows that the three coefficients are getting more important as amplitudes are increasing. After a strong cubic-type increase as p_0 is raised with quite moderate finite amplitudes ($<\approx 5\,kPa$), the three terms tend to a quasi-linear growth for the highest values of p_0. This results suggests that the omission of any of these terms, A_4 and A_3 in ZS2RPv Model (the most widely used in the volume-frame literature [10,12,16–18]), A_4 in IZ3RPv Model, can represent a drawback at high amplitudes. The effects derivated from the nonlinear behavior of the bubble, such as the primary Bjerknes forces or the coupling of the Rayleigh-Plesset model with the wave equation, would also be modified through the introduction of the a_4 term into the differential equation, 4RPv Model, in detriment to ZS2RPv Model which only assumes the existence of the term A_2 and to IZ3RPv Model which only assumes the existence of the terms A_2 and A_3.

From the above diagrams, it is clear that new nonlinear term in Eq. (19) allows the 4RPv Model to better describe the nonlinear behavior of the bubble when its response is the most nonlinear, at resonance.

Several cases assuming other source frequencies below and above the bubble resonance have shown very slight differences between 4RPv, IZ3RPv, and ZS2RPv Models, even at the high finite amplitude $p_0 = 9\,kPa$. Fig. 5 represents the corresponding results during the last periods of the study for $f = 0.5f_b$ (a) and $f = 1.5f_b$ (b). These simulations show that the new model equation, Eq. (19), including the new nonlinear term a_4 does not influence the results, and thus they suggest that the approximation employed in ZS2RPv Model is likely to be enough to track the nonlinear time bubble oscillations when the driving frequency is not close to the bubble resonance. Fig. 6 displays the difference, expressed through the discrete L_2-norm, in terms of v obtained at several amplitudes, from $p_0 = 1\,mPa$ up to $p_0 = 9\,kPa$ between ZS2RPv, v_{ZS2RPv}, and 4RPv, v_{4RPv}, Models on one hand (a), and between IZ3RPv, v_{IZ3RPv}, and 4RPv, v_{4RPv}, Models on the other hand (b), over the large range of source frequencies $f = [0.5f_b, 1.5f_b]$. The rest of parameters are kept identical to the previous case. The discrete L_2-norms of both differences at frequency f is defined by, where $v_{comp_{f,n}} = v_{ZS2RPv_{f,n}}$ or $v_{comp_{f,n}} = v_{IZ3RPv_{f,n}}$,

$$E_{v_f}^{L_2} = \sqrt{\sum_{n=1}^{N} \left(v_{4RPv_{f,n}} - v_{comp_{f,n}} \right)^2}. \tag{24}$$

It can be seen that for the lowest amplitudes the difference found is insignificant for all frequencies. This difference is effectively significant only at high amplitude and when the driving frequency is close to the bubble resonance, which confirms that the new nonlinear model, Eq. (19), is useful and necessary on a limited frequency range around f_b. The Fast Fourier Transform of the v signal indicates that higher harmonics are created when p_0 is raised, as well as a zero-frequency component, which are stronger for the highest p_0 amplitudes with 4RPv Model than with ZS2RPv Model, and even than IZ3RPv Model concerning the zero and second frequency components, as presented in Fig. 7 for the highest

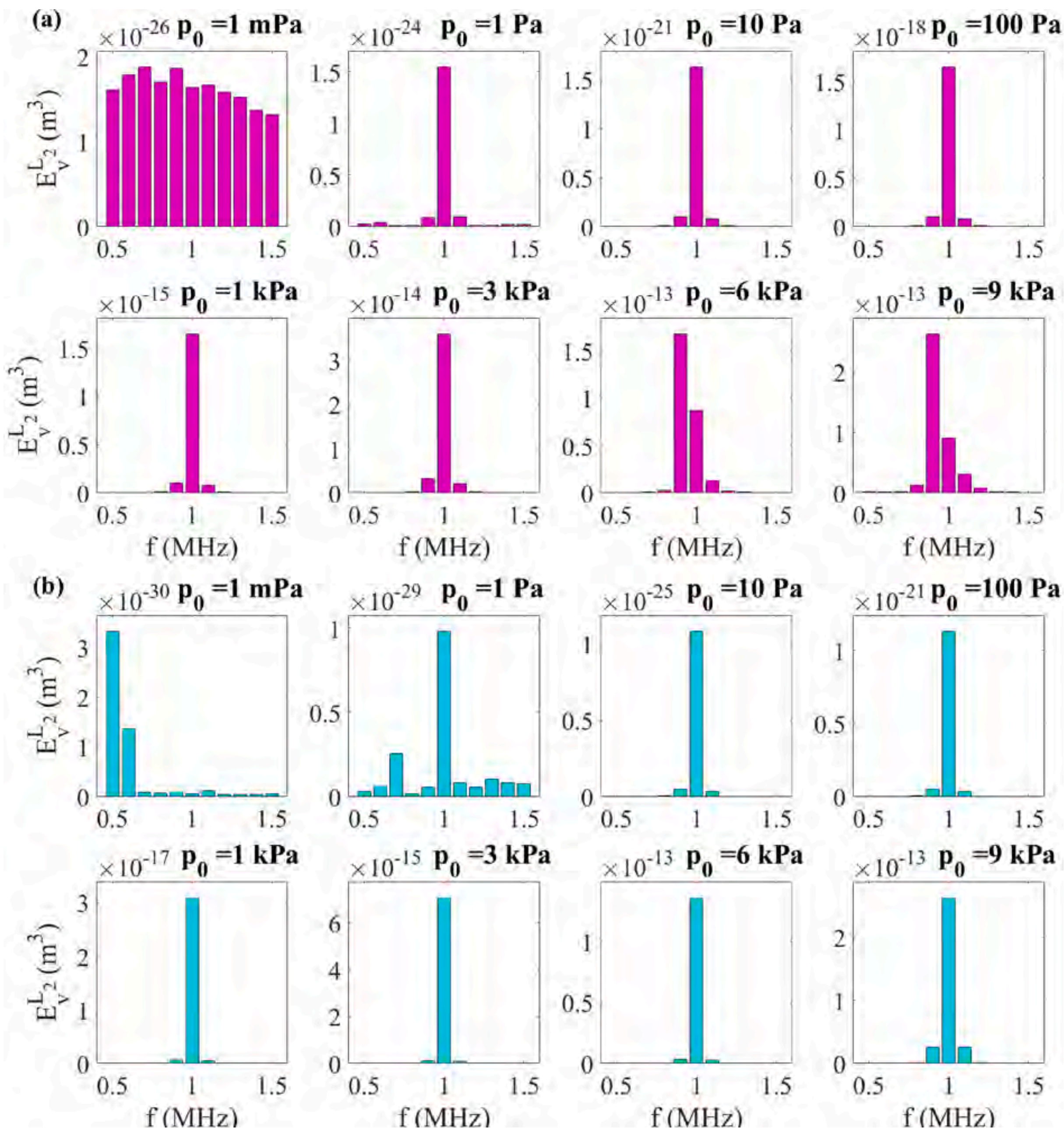

Fig. 6. Discrete L_2-norm vs. f of the difference in v between 4RPv and ZS2RPv Models (a) and between 4RPv and IZ3RPv Models (b).

finite amplitude used, $p_0 = 9\,kPa$. It can be noticed that the third-harmonic component cannot be evaluated by ZS2RPv Model because of its nonlinear restrictions but is reached by the approximations of 4RPv and IZ3RPv Models. The contribution of a_4 in Eq. (19) tends to increase the even-number frequency components. Whereas the harmonic components distort the signal, the effect of the zero-component frequency is the net increase of the mean volume of the bubble, which induces the asymmetry seen in Figs. 2 and 3, and thus could drop its resonance value (nonlinear resonance [17]). The latest point could also have an important effect on the evaluation of the Bjerknes forces.

The next study about the fourth-order term in the bubble equation developed in the volume frame will consider a population of multiple gas bubbles in a liquid by coupling the wave equation to the new model Eq. (19), which, as seen in this paper, is expected to be useful for driving frequencies close to the bubble resonance.

4. Conclusions

We have derived a nonlinear ordinary differential equation for modeling the nonlinear oscillations of a gas bubble placed in an ultrasonic field in terms of bubble-volume variations up to the fourth-order approximation within the Rayleigh-Plesset framework. The equation has been solved numerically and the results obtained from simulations have shown the usefulness of the new model, since the comparison to the classic second-order and third-order approximation equations derived in the 1960–70’s and 1990’s have revealed that the nonlinear behavior of the bubble at high finite amplitude differs when the nonlinearity of the phenomenon is higher, in the vicinity of the bubble resonance.

CRediT authorship contribution statement

Christian Vanhille: Conceptualization, Methodology, Software, Validation, Formal analysis, Investigation, Resources, Data curation,

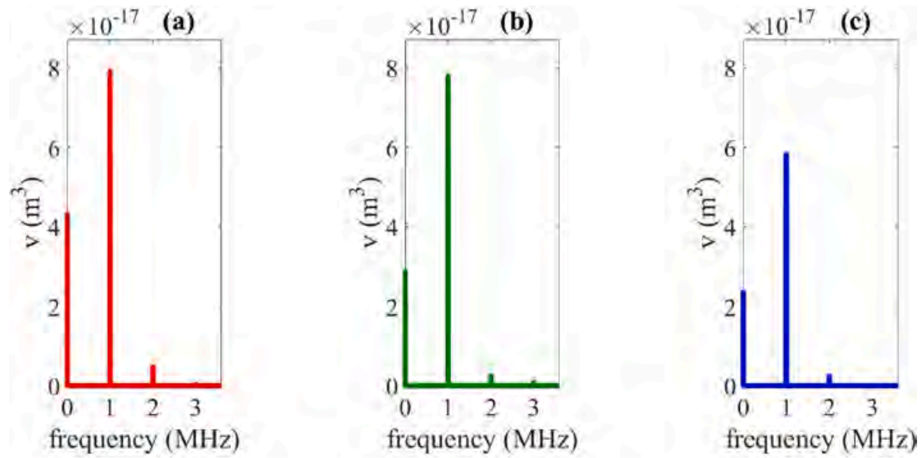

Fig. 7. Comparison of 4RPv, IZ3RPv, and ZS2RPv Models. $f = f_b$. High finite amplitude. Amplitude of bubble volume variation by Fast Fourier Transform. 4RPv (a), IZ3RPv (b), and ZS2RPv (c) Models.

Writing - original draft, Writing - review & editing, Visualization, Project administration, Funding acquisition.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

This work was supported by the National Agency for Research (Agencia Estatal de Investigación, AEI), Ministry of Science and Innovation of Spain (Ministerio de Ciencia e Innovación), and the European Regional Development Fund (FEDER) [Grant No. DPI2017-84758-P].

This work is dedicated to Dr. Cleofé Campos-Pozuelo.

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