Numerical modelling for the simulation of nonlinear ultrasound in liquids with gas bubbles

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Abstract. Several numerical models have been developed in different configurations to simulate the behaviour of finite-amplitude ultrasound when interacting with tiny gas bubbles in a liquid. Since this interaction is highly nonlinear, specific models must be developed to understand the propagation of the waves in this kind of dispersive media for which their nonlinear and attenuation coefficients, as well as the sound speed, are extremely dependent on the ratio of the driven frequency to the bubble resonance. The bubble volume variation is mathematically modelled in the time domain through a Rayleigh-Plesset equation with terms up to the second order, whereas the time-dependent acoustic field relies on the wave equation in one or several dimensions. Both differential equations are coupled and auxiliary conditions are imposed. The differential systems are solved by the developed numerical models. In this paper we study in a three-dimensional resonator with axial symmetry how new harmonics obtained by nonlinear distortion can be enhanced by taking the nonlinear resonance effect into account, and we show that the generation of new frequency components by nonlinear frequency mixing exists. We also analyse the stable cavitation phenomenon in a three-dimensional focused field with axial symmetry by considering a nonlinear dependence of bubble generation in the liquid and the existence of primary Bjerknes forces.

1. Introduction
Bubbly liquids are biphasic media for which a very small amount of gas bubbles can produce very high nonlinear effects on ultrasound without changing the liquid density and with relatively small pressure amplitudes [1-4]. Bubbles also produce dispersive effects and attenuation [1-4]. These media find huge interest in many industrial and medical applications [5-9]. However, the experimental study of such media is delicate since a stable population of homogeneous gas bubbles is not easy to obtain. The development of numerical tools able to simulate the behavior of finite-amplitude waves in bubbly liquids is thus necessary [1-4,6,10-12]. Moreover, many high-power ultrasound processes are based on acoustic cavitation, which includes two different regimes, stable and inertial [2,4,13,14]. Very active theoretical and experimental works have been performed [2-4,6], but a higher understanding of all the phenomenon involved in this framework is still needed. The complexity of the physical problem is such that the mathematical models must include restrictions to idealize the solution obtain after the solution procedure.

Linear responses of the system are obtained when infinitesimal acoustic pressure amplitudes are considered in a liquid, but nonlinear distortion affects the wave when finite amplitudes are assumed, i.e., harmonic components of the fundamental frequency are created and nonlinear combinations of signals of different frequencies generate other components [1-4,10-14]. Moreover, if those amplitudes are above the cavitation threshold, the generation of stable cavitation bubbles from the nuclei present in the liquid...
is triggered, and these bubbles oscillate nonlinearly [4,13,14]. For even higher amplitudes, inertial cavitation takes place, and the bubbles grow and collapse [4,13]. Primary Bjerknes forces are net radiation forces due to an acoustic wave on oscillating bubbles [3]. These phenomena have been studied in the literature [2-4,6,12,13,15].

In this paper we chose to study two aspects of ultrasound propagation in bubbly liquids, in three-dimensional volumes with axial symmetry. On the one hand, we propose to analyze the possibilities of a resonator to enhance the power of harmonics by using the nonlinear resonance effect [3]. We also aim to show that a difference component is generated from a dual-frequency source by nonlinear frequency mixing [16,17]. On the other hand, we perform a study of the stable cavitation process in a focused ultrasonic field, by considering a threshold-based nonlinear dependence of void fraction in the liquid on rarefaction acoustic pressure [4,13,18], and how primary Bjerknes forces act on these bubbles to form agglomerates [1,2,4,13,19]. These analysis are carried out by numerical simulations after adapting our ad-hoc numerical models [19-21]. To this end we consider a nonlinear coupled differential system formed by the wave equation and a Rayleigh-Plesset equation for which the bubble volume variation and the acoustic pressure are nonlinear dependent variables [2,3,11,21].

This mathematical model solved here considers the nonlinear mutual interaction of pressure and bubble vibration variables. In this sense, the nonlinearity of the bubbly medium is due to the presence of bubbles [3,17]. Also, these bubbles imply dispersion and attenuation of acoustic waves. Thus, the nonlinearity and sound speed in the bubbly liquid depends on the ratio of the driving frequency to the bubble resonance. This feature is extremely important for nonlinear waves with multi-frequency components [2,3,21].

Section 2 describes the physical problem we consider and the mathematical model we solve. It also defines how we simulate the stable cavitation process, including the nonlinear dependence law, and the equations we solve to evaluate the bubble motion. Section 3 presents the numerical simulations and discusses our results.

2. Models

2.1. Physical problem
A three-dimensional cylindrical volume of liquid of radius R and length Z in which a population of gas bubbles is evenly distributed is considered. The study is performed from time \( t = 0 \) s to T. Cylindrical coordinates with axial symmetry around \( \vec{e}_z \) \( (0 \leq \theta \leq 2\pi) \) are used, so that the coordinates \((z,r,t)\) are employed. The bubbly liquid volume is excited by an ultrasonic source at \( z = 0 \) m. The problem we solve is the nonlinear interaction of oscillating bubbles and ultrasound of finite amplitudes. The void fraction of gas in the liquid leads to high nonlinearity of the medium and to dispersion of features such like acoustic attenuation and nonlinearity.

2.2. Differential system
The system we describe here models the nonlinear interaction of ultrasound and gas bubble oscillations in the most general configuration: three-dimensional space domain and variable density of bubbles in the liquid. Both fields are defined by the bubble-volume variation \( v(z,r,t) \), i.e., its perturbation compared to its initial volume \( v_o \) (corresponding to the initial radius \( R_o \)), and the acoustic pressure \( p(z,r,t) \). The wave equation and a Rayleigh-Plesset equation are coupled to simulate the nonlinear mutual influence of bubbles and ultrasound, accounting for nonlinearity, dispersion, and attenuation due to the bubbles in the liquid [3,11,21]. In addition to the coupled differential system, auxiliary conditions are added to model the absence of acoustic and bubble perturbations at the outset, the axial symmetry, and the boundary conditions. The differential problem is then the following, in which partial derivatives are indicated by subscript on \( z, r \) and \( t \):
\[ v_n + \delta \alpha_{0} v_i + \omega_0^2 v = av^2 + b \left( 2vv_n + v_i^2 \right) - \eta p, \quad (z, r, t) \in [0, Z] \times [0, R] \times (0, T), \]
\[ p_n + p_i/r + p_{c_2} n - p_{c_1} n = -p_i N(z, r) v_i, \quad (z, r, t) \in [0, Z] \times [0, R] \times (0, T), \]
\[ p(z \neq 0) = v = p_i(z \neq 0) = v_i, \quad t = 0, (z, r) \in [0, Z] \times [0, R], \]
\[ p_i = 0, \quad r = 0, (z, t) \in [0, Z] \times (0, T), \]
\[ p_z = -p_i/c_i \text{ or } p = 0, \quad z = Z, (r, t) \in [0, R] \times (0, T), \]
\[ p = s, \quad z = 0, (r, t) \in [0, R] \times (0, T). \]

\( \alpha_0 \) is the first bubble resonance frequency, \( \delta = 4\mu_0/\omega_0 R_0^2 \) is the viscous damping coefficient of the bubble-liquid mixture, where \( \mu_0 \) is the kinematic viscosity of the liquid, \( \rho_1 \) is the equilibrium density of the liquid, \( c_i \) is the small-amplitude sound speed of the liquid, \( N(z, r) \) is the bubble density at the point \((z, r)\), \( s(r, t) \) is the function defining the excitation at the source, \( a = (\gamma + 1) \omega_0^2 / 2v_0 \) is the nonlinear coefficient related to the adiabatic gas law, where \( \gamma \) is the specific heat ratio of the gas, \( b = 1/6v_0 \) is the nonlinear coefficient accounting for the dynamic response of bubbles, and \( \eta = 4\pi R_0/\rho_1 \) is a constant.

For standing waves in cavities a bubbly liquid is considered and free-wall conditions are assumed. With a single-frequency source the following function is used: \( s(t) = p_0 \sin(\omega t) \), where \( p_0 \) is the amplitude and \( \omega = 2\pi f \) is the frequency, whereas when a dual-frequency source is considered the following function is used: \( s(t) = p_0 \sin(\omega_0 t) + p_0 \sin(\omega_1 t) \), where \( \omega_0 = 2\pi f_1 \) and \( \omega_2 = 2\pi f_2 \) are the primary frequencies.

When the stable cavitation process is assumed, a population with homogeneous density \( N_0 \) of very tiny spherical gas bubbles of radius \( R_0 \) is present in the liquid at the outset, simulating the gas nuclei from which the cavitation process can take place. A phased array composed by \( S \) spherical harmonic pressure sources, \( \rho = \rho \gamma_0 g\left( r \right) \sin(\omega t + \phi) \), where \( g \) is the spatial distribution and \( \phi \) is the phase, excited with phase delays is considered to create a focused pressure field. The whole contribution of the source is denoted by \( s(r, t) \). The finite-difference numerical model developed in [21] is adapted to account for bubble generation in such a way that the acoustic pressure field and the bubble-volume variation field are calculated at each space point on the fast time scale and at each space point on the slow time scale, i.e., on the interval \([0, T]\), whereas the bubble density in the liquid is evaluated on the slow time scale from the minimum rarefaction pressure \( p_-(z, r) \), in absolute value, reached at each space point during \( \Theta \) (several acoustic cycles), following the nonlinear law [18, 22]:

\[ N(z, r) = \begin{cases} 
N_0 \tan \left( \frac{p_-(z, r) - \Phi_c}{p_+ - \Phi_c} \pi \right) & \text{if } p_-(z, r) \geq \Phi_c, \quad (z, r) \in (0, Z) \times (0, R), \\
N_0 & \text{if } p_-(z, r) < \Phi_c.
\end{cases} \]
The bubble speed is then calculated. The motion of the nth bubble due to $B_0$ is tracked using the time evolution of its position vector in the $(z,r)$ plane at time $t$, $\mathbf{u}_n(t) = (z_n(t), r_n(t))$, i.e., the solution of the system formed by the dynamic equation, the initial position, $\mathbf{u}_{n_0} = \mathbf{u}_n(t = t_0)$, and the null initial speed,

$$M\mathbf{u}_n = \langle \overline{F}_n \rangle, \quad t \in (t_1, t_2),$$

$$\mathbf{u}_n = \mathbf{u}_{n_0}, \quad t = t_1,$$

$$\mathbf{u}_{n_1} = \mathbf{0}, \quad t = t_2,$$

$\langle \overline{F}_n \rangle$ is the average on $\Theta$ of $\overline{F}_n(t) = -\nabla p_n(t)$, i.e., $B_0$ applied to the nth bubble of mass $M = \langle V_n \rangle \rho_g + \rho_{l}/2$, and for which $V_n(t)$ is its volume, $\mathbf{u}_{n_1}$ is its speed, $\nabla$ is the gradient operator, $p_n(t)$ is the acoustic pressure at $\mathbf{u}_n(t)$, and $\rho_g$ is the equilibrium density of the gas. The solution of this differential system is obtained by applying a finite-difference scheme [19].

The main physical restrictions of the mathematical model used are described in [1-3, 11].

2.3. Numerical models

Two different numerical models have been developed to solve the coupled set of differential equations shown in Section 2.2. For standing waves, a finite-volume based numerical model was developed in [20] and is used in Section 3.1. For stable cavitation bubbles, a finite-difference based numerical model was developed in [21] and is used in Section 3.2. In these references the discretized equations are described in details.

3. Results and discussion

We consider that the liquid is water, $c_l = 1500 \text{m/s}$, $\rho_l = 1000 \text{kg/m}^3$, $\mu_d = 1.43 \times 10^{-3} \text{kg/s/m}^2$, $\mu_s = 1.43 \times 10^{-6} \text{m}^2 \text{s}^{-1}$, and the bubble gas is air, $\gamma = 1.4$, $\rho_{g} = 1.29 \text{kg/m}^3$. Two types of numerical simulations are performed to illustrate the use of the models developed in Section 2. On one hand, we study the nonlinear resonance effect in a three-dimensional cavity with axial symmetry to enhance the nonlinear harmonics of distorted ultrasound and the generation of new frequency components by nonlinear frequency mixing. On the other hand, we analyse the stable cavitation process in a three-dimensional focused field with axial symmetry and the effect of primary Bjerknes forces.

3.1. Nonlinear resonance and nonlinear frequency mixing in a standing-wave field

We consider a cylindrical cavity of dimensions $0.00175 \times 0.0035 \text{m}^2$ in the $(z,r)$ plane (figure 1) excited at its resonance frequency $f = 258 \text{kHz}$ in the bubbly liquid defined by $R_0 = 2.5 \mu\text{m}$ and $N = 5 \times 10^{11}$ bubbles $\text{m}^{-3}$, and for which the sound speed at $f$ is $c = 987.63 \text{m/s}$ [3]. A frequency sweep is applied around the resonance at two amplitudes at the source, $p_0 = 1 \text{Pa}$ and $p_0 = 15 \text{kPa}$. The maximum pressure amplitude reached in the cavity is presented in figure 2. For low amplitudes (linear regime), the highest value is obtained at the source frequency $f_\text{a} = 258 \text{kHz}$, which matches the resonance. For high amplitudes (nonlinear regime), the highest value is obtained at the source frequency $f_{\text{a}_n} = 255.01 \text{kHz}$, which is the nonlinear resonance of the cavity. This frequency does not match the linear resonance, a shift of the resonance of the cavity is produced when the amplitude is raised: $\Delta f = 2.99 \text{kHz}$. This means that a softening is undergone by the bubbly liquid when amplitudes are increased, i.e., the sound speed is lowered [23]. Figure 3 represents the difference of amplitude of the fundamental, second and third harmonics obtained in the resonator when the excitation is carried out at
\( f_{\text{res}} = 255.01 \text{kHz} \) and at \( f_\text{r} = 258 \text{kHz} \). The enhancement of the response in the cavity when the nonlinear resonance phenomenon is taken into account can be seen.

Figure 4 shows the amplitude of the difference frequency component obtained in the same cavity with the same bubbly medium by nonlinear mixing of two signals from the source, for which the primary frequencies are \( f_1 = 6 \times 10^3 \text{ Hz} \) and \( f_2 = 9.1758 \times 10^3 \text{ Hz} \), and the amplitude at the source \( p_0 = 12 \text{ kPa} \). The difference frequency is \( f_4 = f_2 - f_1 = 3.1758 \times 10^3 \text{ Hz} \), for which the sound speed in the bubbly liquid is \( c = 1215.7 \text{ m s}^{-1} \).

**Figure 1.** Cylindrical cavity excited at its resonance frequency.

**Figure 2.** Maximum pressure amplitude reached in the cavity with two amplitudes at the source.
3.2. Motion of stable cavitation bubbles in a focused field

We consider the source at \( f = 1 \text{MHz} \) emitting up to \( T = 8 \times 10^{-6} \text{s} \) in the liquid-nuclei mixture for which \( R = 0.01 \mu \text{m} \), \( N = 1 \times 10^{10} \text{bubbles m}^{-3} \), \( p' = 3.5 \text{MPa} \) and \( \Phi = 1.5 \text{MPa} \), confined in the cylinder of dimensions \( Z = 0.0075 \text{m} \) and \( R = 0.0045 \text{m} \). We assume that \( \Theta = 2/f \). For the medium with new stable cavitation bubbles we assume \( N = 2.5 \times 10^{10} \text{bubbles m}^{-3} \), \( R = 35 \mu \text{m} \), and the maximal void fraction is \( V = 0.449\% \). Figure 5a shows \( p \) in the \((z,r)\) plane for \( \Theta#1-\Theta#4 \). Since \( p > \Phi \) in some area from \( \Theta#2 \), new big stable cavitation bubbles are created. Figure 5b represents \( N \) in the \((z,r)\) plane at the end of \( \Theta#1-\Theta#4 \), which is used for the evaluation of acoustic pressure at \( \Theta#2-\Theta#5 \). These cavitation bubbles form a cloud around the focus that changes the propagation of ultrasound. The nonlinearity of the bubbly liquid raises in the cloud and the pressure signal is distorted.
nonlinearly. Figure 6 displays the frequency decomposition of $p$ on the symmetry axis ($\Theta \#5$). Higher harmonics appear in the spectrum: 2 MHz, 3 MHz, 4 MHz, which means that the effect of the nonlinearly oscillating big gas bubbles on ultrasound is the transfer of acoustic energy from the fundamental to higher harmonics. Note that the corresponding linear diagram at low amplitude would show one single frequency (the source frequency), since no new cavitation bubbles would be created.

Figure 7 represents the distribution in the $(z,r)$ plane at $\Theta \#5$ of $\langle F_r \rangle$ (a) and $\langle F_r \rangle$ (b) that affect the new cavitation bubbles, and their speed field (c). Around $z = 0.002\text{m}$ the bubbles are pushed toward the source and some are pushed away from the axis. Around $z \approx 0.0045\text{m}$ the bubbles agglomerate. This result, due to the combined effect of stable cavitation process and primary Bjerknes forces, is quite coherent with experimental data [24, 25].

Figure 5. (a) $p_-$ in the $(z,r)$ plane for $\Theta \#1 - \Theta \#4$. (b) $N$ in the $(z,r)$ plane at the end of $\Theta \#1 - \Theta \#4$. 
Figure 6. Frequency decomposition of $p$ on the symmetry axis ($\Theta$#5).
Figure 7. (a) Distribution of $<F_{zr}>$ in the $(z,r)$ plane ($\Theta#5$). (b) Distribution of $<F_{rr}>$ in the $(z,r)$ plane ($\Theta#5$). (c) Distribution of stable cavitation bubble speed in the $(z,r)$ plane ($\Theta#5$).

4. Acknowledgments
This work is supported by the National Agency for Research of Spain and the European Regional Development Fund through the grant DPI2017-84758-P.

Christian Vanhille dedicates this work to Dr. Cleofé Campos-Pozuelo.

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