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Journal of Sound and Vibration

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

Fast bubble dynamics and sizing

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ARTICLE INFO

Article history:

Received 15 October 2014

Received in revised form

1 June 2015

Accepted 20 June 2015

Handling Editor: L. Huang

ABSTRACT

Single bubble sizing is usually performed by measuring the resonant bubble response using the Dual Frequency Ultrasound Method. However, in practice, the use of millisecond-duration chirp-like waves yields nonlinear distortions of the bubble oscillations. In comparison with the resonant curve obtained under harmonic excitation, it was observed that the bubble dynamic response shifted by up to 20 percent of the resonant frequency with bubble radii of less than 100 μm . In the case of low pressure waves ($P < 5$ kPa), an approximate formula for the apparent frequency shift is derived. Simulated and experimental bubble responses are analyzed in the time–frequency domain using an enhanced concentrated (reassigned) spectrogram. The difference in the resonant frequency resulted from the persistence of the resonant mode in the bubble response. Numerical simulations in which these findings are extended to pairs of coupled bubbles and to bubble clouds are also presented.

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1. Introduction

Microbubble detection and sizing methods still remain to be found for a wide range of industrial and scientific purposes. Most conventional approaches focus on the presence of bubbles in liquids [1], which even at low concentrations may considerably impact the thermodynamic behavior of the fluid mixture involved. Oceanic measurements have helped us to understand role of bubbles (mass and energy transfers are bubble size dependent) in greenhouse gas cycles [2–4] and how they affect sound propagation [5]. Recent nuclear power studies have focused on the assessment of undesirable microbubble formation in liquid sodium or liquid CO₂ coolants [6–10] which can affect ultrasonic control systems as well as increasing the risk of gas pocket formation (a core safety issue). Bubbles are now being used to reduce scattering cross section of modern submarines and new bubble-filled anechoic coating materials (metamaterial properties) are currently being developed [11]. In other applications, microbubbles are used as bioactive food ingredients, green adjuncts for water or surface cleaning [12–15], ultrasound contrast agents [16–20] and as efficient pumps or activators in the field of microtechnology [21].

In bubbles driven by sound fields with sufficiently high pressure levels, the nonlinearities could be used to assess their size and concentration. Procedures of this kind are being used to grade the bubble occurring in the bloodstream and prevent the risks associated with surgery (cardiopulmonary bypass) induced gas emboli and decompression sickness [22–24]. Introduced or induced

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microbubbles can improve (pre-seeding) non-invasive surgery based on high intensity focused ultrasounds [25] or promote the delivery of drugs (sonoporation) [26] with enhanced therapeutic indexes. Nano-droplets of liquid perfluorocabone can be vaporized with ultrasounds, for example, once they have reached extravascular cancer cells thanks to the Enhanced Permeability and Retention (EPR) process. In order to extravasate neo-vasculature [27], the droplets must be less than 400 nm in diameter and then be transformed into micron-sized (ideally between 0.5 and 6 μm) echogenic bubbles.

With the Dual-Frequency Ultrasound Method (DFUM) [28,29], resonant bubbles are no longer liable to be confused with larger ones, as usually occurring in the case of linear and harmonic techniques [30,31]. In addition, the DFUM has proved to be efficient even when taken out to the surf zone to measure bubble sizes under breaking waves [2] or when working with a low mechanical index as well in in vitro and in vivo studies [32–34]. The DFUM is based on the fact that the resonance frequency of a bubble is inversely proportional to its radius. There are several ways of implementing DFUM [1]. The combination-frequency approach consists in transmitting two high frequencies: the difference between them is equal to the bubble resonance. This approach requires high level pressure waves of the order of 100 kPa to generate the bubble resonance. Alternatively, the modulation frequency approach requires less pressure (less than 5 kPa) and was therefore used in this study, where linear behavior is required. The frequency f_{imag} of the first wave, the “imaging wave”, is relatively high (High Frequency: HF) in comparison with the resonance frequency f_{res} of the bubble [28], and with the frequency f_{pump} of the so-called “pumping wave” (Low Frequency: LF). As the bubble oscillates under the LF pressure field, the instantaneous amplitude and frequency of the backscattered imaging wave are modulated by the cross-section of the bubble. The oscillations, which are most observable at sum-and-difference frequencies $f_{\text{beat}} = |f_{\text{imag}} \pm f_{\text{pump}}|$, therefore reach a maximum amplitude when $f_{\text{pump}} = f_{\text{res}}$. This procedure provides an efficient means of jointly detecting and sizing microbubbles. Multiple harmonic pumping waves have been recently used to further increase the range of radii of the bubbles characterized [35]. In addition, not only amplitude modulation but also phase modulation of the signals scattered by the pulsating bubbles can be exploited in order to determine their size [36].

In the classical DFUM implementation based on the assumption that the bubble behavior is stationary, sequential detection of mono-disperse bubble sets is carried out by applying stepped frequency bursts. However, these “long sweep duration” procedures are not appropriate for dealing with fleeting or moving bubbles (such as those crossing the tricuspid valve in the right ventricle in less than 20 ms). The aim of the present paper is to present a new signal processing approach based on short-duration excitations (of less than 10 ms) in order to overcome the above limitations.

The resonant behavior of a bubble activated by a chirp pumping wave shows a specific pattern of amplitude modulation, which determines the resonance frequency. However, as we will see in the next section, it has emerged that this resonant frequency is affected by the chirp duration, or rather, in a given frequency range, by the chirp rate. In the quasi-linear regime (i.e., at pump pressure amplitudes < 5 kPa), a numerical approach is proposed here for quantifying the frequency shift, based on the Harmonic (i.e., no frequency modulation) Resonance Frequency (HRF). The analytically estimated bias matched the numerical simulations based on the modified Rayleigh–Plesset equation. A dedicated time–frequency transformation was developed using the reassignment technique for use with both numerical and experimental data [37–39]. The high resolution obtained makes it possible to clearly identify the reason for the frequency shift. Predictions of bubble-pair and bubble-cloud responses are also presented.

2. Harmonic versus dynamic bubble resonances

2.1. Harmonic resonance frequency of a bubble

The volumetric pulsations of a single bubble, which is assumed to be spherical and small (in comparison with the acoustical wavelength), surrounded by an infinite incompressible medium can be described by the well-known nonlinear Rayleigh–Plesset model.

In order to finely simulate the bubble resonant behavior, the three damping mechanisms, corresponding to viscous losses at the bubble wall, sound radiation into the fluid and thermal losses, must all be included in the dynamic model. Assuming that the oscillations show polytropic behavior, the modified form of the Rayleigh–Plesset equation is

$$\rho_L \left[R\ddot{R} + \frac{3}{2}\dot{R}^2 \right] = P_B(t) - P_0 - P(t) + \frac{R}{c} \left(1 - \frac{\dot{R}}{c} \right) \frac{dP_B}{dt} \quad (1)$$

where ρ_L is the fluid density, R the instantaneous bubble radius, $P(t)$ the acoustic driving pressure, P_0 the static pressure in the fluid, and c the sound speed in the liquid. The term $P_B(t)$ is the pressure exerted on the liquid due to the pulsation of the bubble:

$$P_B(t) = \left(P_0 + \frac{2\sigma}{R_0} \right) \left(\frac{R_0}{R} \right)^{3\zeta} - \frac{2\sigma}{R} - 4\eta_L \frac{\dot{R}}{R} \quad (2)$$

where R_0 is the equilibrium bubble radius, σ the surface tension, η_L the fluid viscosity, ζ the polytropic exponent.

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