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To cite this article: Y N Zhang *et al* 2015 *IOP Conf. Ser.: Mater. Sci. Eng.* **72** 012003

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Bubble dynamics under acoustic excitation with multiple frequencies

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Abstract. Because of its magnificent mechanical and chemical effects, acoustic cavitation plays an important role in a broad range of biomedical, chemical and mechanical engineering problems. Particularly, irradiation of the multiple frequency acoustic wave could enhance the effects of cavitation. The advantages of employment of multi-frequency ultrasonic field include decreasing the cavitation thresholds, promoting cavitation nuclei generation, increasing the mass transfer and improving energy efficiency. Therefore, multi-frequency ultrasonic systems are employed in a variety of applications, e.g., to enhance the intensity of sonoluminescence, to increase efficiency of sonochemical reaction, to improve the accuracy of ultrasound imaging and the efficiency of tissue ablation. Compared to single-frequency systems, a lot of new features of bubble dynamics exist in multi-frequency systems, such as special properties of oscillating bubbles, unique resonances in the bubble response curves, and unusual chaotic behaviours. In present paper, the underlying mechanisms of the cavitation effects under multi-frequency acoustical excitation are also briefly introduced.

1. Introduction

Under radiation of acoustic waves, the bubbles will oscillate, termed as “acoustic cavitation” [1,2]. Acoustic cavitation has attracted much attention for many years because of its unique physical complexity [3], chemical applications [4] and biomedical significance [5]. Particularly, a considerable body of work has been produced treating the cavitation under multiple-frequency acoustic wave irradiation, i.e., two or more acoustic waves with the same or different frequencies acting on cavitation bubbles simultaneously (as shown in Figure 1).

In the literature, many positive effects of multi-frequency excitation have been reported. For example, the use of multi-frequency ultrasound field could lower the cavitation thresholds [6], generate more new cavitation nuclei, increase the active cavitation volume [8] and improve energy efficiency [9]. Hence, the multi-frequency systems are employed to enhance the intensity of sonoluminescence [10-17], to increase efficiency of sonochemical reaction [18-22], to improve the accuracy of ultrasound imaging [23-25] and the efficiency of tissue ablation [26,27].

The high energy focusing during bubble collapsing under acoustical wave could induce light emission, termed “sonoluminescence (SL)” [28]. Multi-bubble sonoluminescence (MBSL) and Single-bubble sonoluminescence (SBSL) were found by Marinesco and Trillat [29] and Gaitan et al. [30] respectively. For recent reviews on sonoluminescence, readers are referred to refs. [28,31,32]. Plenty



of works have proved that the use of multi frequency acoustical excitation is an efficient way to boost sonoluminescence. Holzfuss et al. [11] employed driving acoustic wave with a combination of the first and the second harmonics to increase the light emission during bubble oscillations up to 300% higher than the system with only fundamental frequency excitation. The relative phase and pressure amplitudes plays an important role on the intensity of sonoluminescence. Krefting et al. [13] investigated the influential parameters on the dual-frequency excited SBSL more systematically. By experimental measurements and numerical simulations, the region of light emission is mapped into the three-dimensional parameter space spanned by the two driving pressure amplitudes and their relative phase. The addition of the second frequency was able to amplify SBSL in particular parameter zone. Ciuti et al. [6,17] studied the factor of the enhancement of MBSL in ultrasound field with highly different frequencies. In most of the cases, the addition of low frequency into the high frequency field led to stronger light emission, except the intensities of both frequencies were much higher than the corresponding cavitation thresholds. For other recent results of sonoluminescence under multi frequency excitations, readers are referred to refs. [7,10,14,16,18].

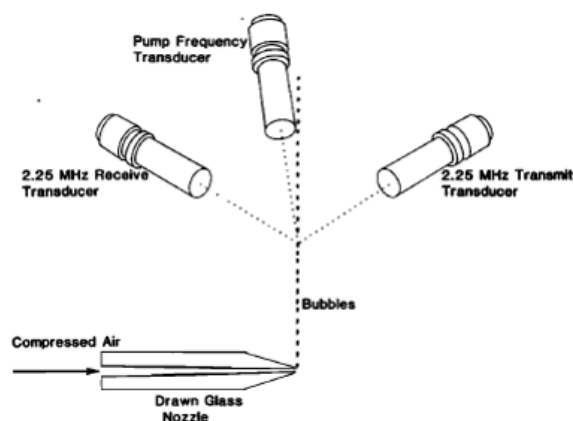


Figure 1. (Figure 3 of ref. [53]) Schematic of the multi-frequency system. Three ultrasonic transducers focus on air bubbles simultaneously. Two of them are transducers with the same frequencies (2.25 MHz) and the other one has a different frequency.

The high temperature and pressure created by bubble collapse can also promote chemical activities within or near the bubbles, which termed as “sonochemistry” [33]. The mechanisms and applications of sonochemistry were reviewed recently in a number of works [34–38]. The efficiency of sonochemical reactions could be enhanced by multi-frequency ultrasound systems. Feng et al. [21] employed both dual-frequency and three-frequency systems by combining ultrasonic transducers with frequencies of the kHz order and the MHz order. The release of iodine, the change of electroconductivity, and the fluorescence formation during sonication were all enhanced by multi-frequency sound source. In past decade, the application of multi-frequency systems was studied widely by researchers. Researchers also made efforts on optimizing the cavitation effects in sonochemical reactors [9,22,39,40]. Moholkar et al. [41,42] revealed that the mode and spatial distribution of the cavitation could be controlled by adjusting the parameters of the dual-frequency field. For instance, the phase difference between the two ultrasound waves had greater influence on production of radicals than the frequency ratio. And it is possible to overcome the directional sensitivity of the cavitation events and erosion of the sonicator surface by inducing the additional ultrasound waves. Kanthale et al. [18] investigated the effect of intensity and frequency of dual-frequency system numerically and compared the results with experimental results in literature. Their work indicated that there exists optimum value of the ultrasound intensity and frequencies.

Acoustic cavitation has wide application in the field of biomedicine. It can enhance the accuracy of ultrasound imaging with the help of the microbubbles [24,43], promote drug and gene transfer into tissue and cells [44,45], treat tumors and ablate tissues [46–47]. Recently, researchers tried to benefit these practices by induce multi-frequency systems. He et al. [26] introduced a high intensity focused ultrasound (HIFU) device which could work both at single-frequency mode and dual-frequency mode. The experimental results showed that the dual-frequency HIFU induced larger tissue lesion than the

conventional one under single-frequency mode, which meant that dual-frequency could improve the efficiency of tumor ablation. Guo et al. [27] placed tissues under the focus of HIFUs under single-frequency, dual-frequency and tri-frequency modes respectively. The multi-frequency mode could yield higher temperature during tissue ablation and higher raise rate of the temperature. The numerical simulation agreed well with the experimental results.

In present paper, the recent development of the bubble dynamics under multiple frequencies are reviewed together with the physical mechanisms of cavitation effects. The whole paper is organized as follows: in Sec.2, the oscillation of bubbles and its response to the multi-frequency acoustic waves are discussed together with its nonlinear features (e.g. chaos); in Sec.3, physical mechanisms relating with promotion of cavitation effects under multiple-frequency waves are briefly introduced.

2. Fundamentals of bubble dynamics under multiple frequencies

In the physical, chemical and biomedical effects of cavitation and corresponding applications mentioned above, oscillations of bubbles play an essential role. Therefore, bubble dynamics is one of the paramount problems in the cavitation dynamics. In this section, the basic features of bubble oscillations under multi-frequency excitations are introduced.

2.1. Oscillations of bubbles

For solving the radial oscillations of gas bubbles in liquids, various models have been developed [1,3,48-50]. By solving the bubble motion equations, the variation of bubble radius with time could be obtained as shown in Figures 2 and 3 (solutions from the Keller-Miskis model [51]). For single-frequency sinusoidal acoustic excitation (as shown in Figure 2), the gas bubble shows steady oscillation with the amplitude and frequency unchanged while for dual-frequency acoustic excitation (shown in Figure 3), the oscillation of gas bubble is much more complicated with the frequency and amplitude oscillating.

2.2. Response of bubbles

The bubble oscillators can be described by response curves, i.e., the amplitude of the steady state oscillation plotted versus the driving frequency. Lauterborn [52] gave a thorough investigation of the basic properties of nonlinear oscillations of gas bubbles in liquids. The response curves of a bubble to a single-frequency field were calculated and displayed in Figure 4. The expression n/m (Here, m and n are two integers) above the peaks represents the order of the resonance. Cases with $m=1$ and $n=2, 3, \dots$ correspond to harmonics; cases with $m=2, 3, \dots$ and $n=1$ correspond to subharmonics; cases with $m=2, 3, \dots$ and $n=2, 3, \dots$ correspond to ultraharmonics. For the definitions of these resonances, readers are referred to Page 284 of ref. [52]. However, the response curves of bubbles under multi-frequency excitations have not been studied yet.

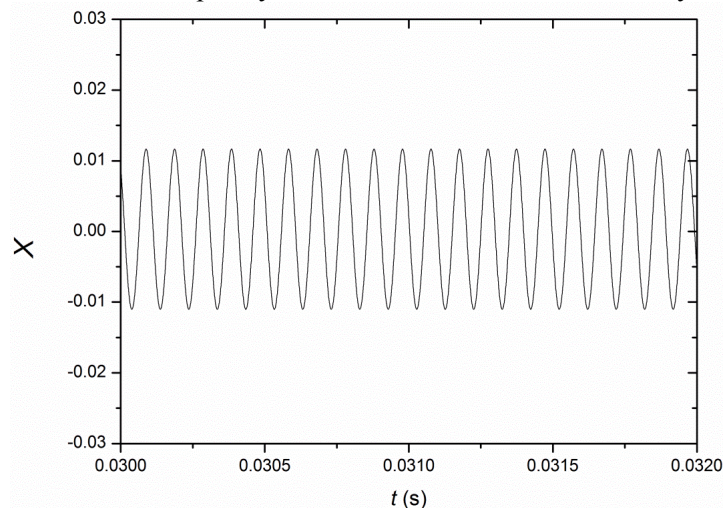


Figure 2 Instantaneous bubble radius excited by the single-frequency acoustic excitation against time. The equilibrium bubble radius: $R_0 = 10\mu m$. The angular frequency of sound wave: $\omega_s = 0.03\omega_0$. The pressure amplitude of sound wave: $P_A = 0.05P_0$. $X = (R - R_0)/R_0$. ω_0 is the natural frequency of the bubble. P_0 is the ambient pressure.

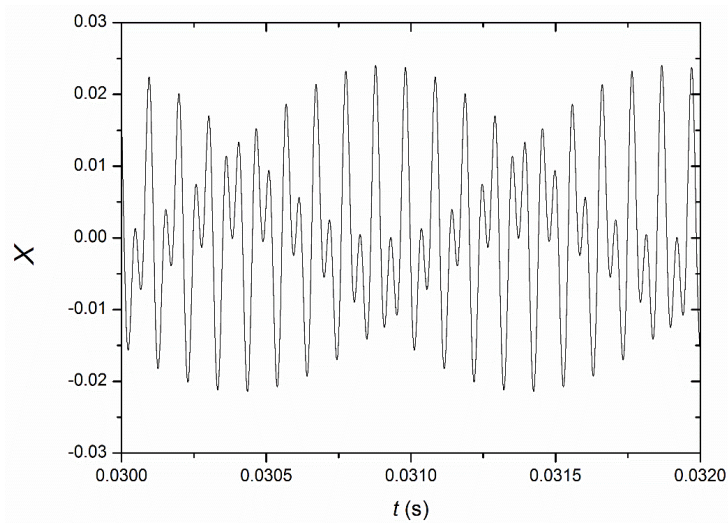


Figure 3 Instantaneous bubble radius excited by the dual-frequency acoustic excitation against time. The equilibrium bubble radius: $R_0 = 10\mu\text{m}$. The angular frequency of sound wave one: $\omega_1 = 0.03\omega_0$; sound wave two: $\omega_2 = 0.057\omega_0$. The pressure amplitude of sound wave: $P_{A1} = P_{A2} = 0.05P_0$. $X = (R - R_0)/R_0$. ω_0 is the natural frequency of the bubble. P_0 is the ambient pressure.

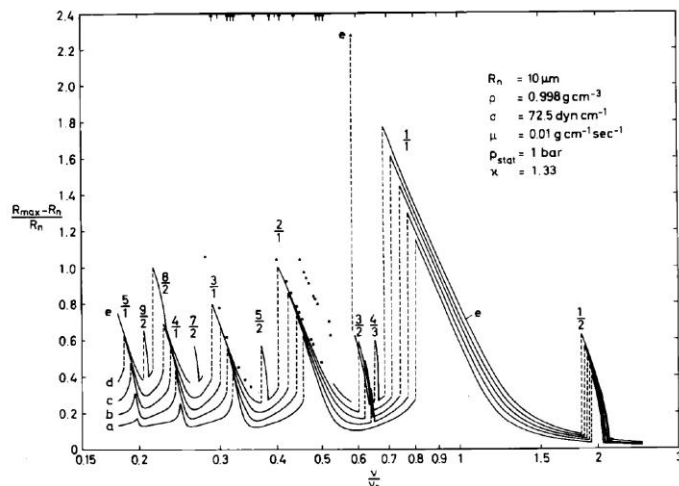


Figure 4 (Fig 3 of ref. [52]) Frequency response curves for a bubble in water with a radius at rest of $R_n = 10\mu\text{m}$ for different sound pressure amplitudes P_A of (a) 0.4, (b) 0.5, (c) 0.6, (d) 0.7, and (e) 0.8 bar. ν is the frequency of the driving sound field. ν_0 is the nature frequency of the bubble oscillation. R_{max} is the maximum radius of the bubble during its steady-state oscillation. The numbers marked above the peaks are the orders of the resonances, represented as n/m .

Via the Fourier transform, the “time domain” diagram (e.g., Figures 2 and 3) could transform to a “frequency domain” diagram, i.e., the power spectrum. Figure 5 is a typical power spectrum of bubble oscillation under single-frequency excitation. The corresponding frequencies of the “lines” are the driving frequency ν (main resonance), its integer multiples (harmonics), $\nu/2$ (subharmonic), $3\nu/2$ and $5\nu/2$ (ultraharmonics). Compared to the single frequency system, the echo of bubble oscillators in a dual-frequency acoustical field with frequencies f_1 and f_2 would contain the bands at $f_1 \pm f_2$ (Figure 6 shows the schematic spectrum of the bubble echo) besides the typical bands in single-frequency systems [53,54]. Furthermore, the echoes at $f_1 \pm f_2$ are much “sharper” than those at the main resonant frequency, which lead to more accurate bubble size measurement [53-55].

Nevertheless, in multi-frequency systems, the scattered echo contains more bands than the difference and sum frequencies of driving frequencies. Figure 7 [56] is an experimental spectra of scattered signal of bubbles excited by a dual-frequency field (with frequencies f_1 and f_2). In this figure, there are bands corresponding to main resonances (marked by ■), their harmonics (marked by □), subharmonics (marked by ▲) and sum and difference (marked by △). Besides these, there are other peaks (marked by ◆), of which corresponding frequencies are $2f_1 - f_2$ and $2f_2 - f_1$ and their magnitudes are at the same order with the harmonics. All of these suggest the unique properties of

bubble oscillations under multi-frequency excitations, which are worthy to investigate quantitatively and systematically.

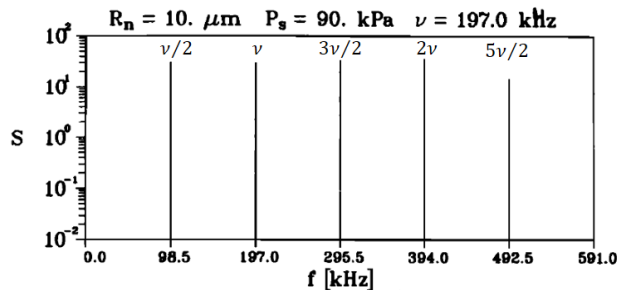


Figure 5. (Figure 13 of ref. [57]) Power spectrum of bubble oscillator. Equilibrium bubble radius $R_n = 10\mu\text{m}$. sound pressure amplitude P_s : 90kPa. Driving frequency ν : 197kHz

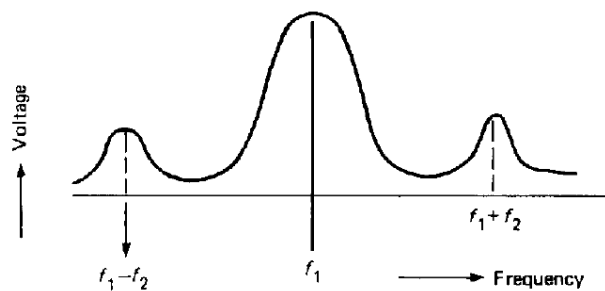


Figure 6. (Figure 1 of ref. [54]) The schematic spectrum of the bubble echo under dual-frequency excitation with frequency f_1 and f_2 .

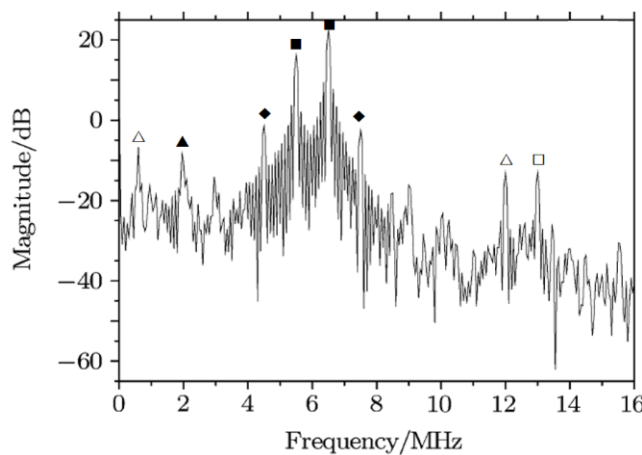


Figure 7. (Figure 3 of ref. [56]) Experimental spectra of the scattered signal from bubbles excited by a dual-frequency ultrasound field. The frequencies of the two sound waves are $f_1 = 5.5\text{MHz}$, $f_2 = 6.5\text{MHz}$, respectively. Sound pressure amplitude 0.5 MPa.

2.3. Chaotic oscillations

For a bubble oscillator forced by sound wave with period T , the period of its radial oscillation is equal to T according to the linear theory (as the oscillator shown in Figure 2). The phase space diagram [58] of the bubble oscillating with the driving frequency would be like Figure 8 (a), which is also named “limit cycle”. When the control parameter of the system, μ , becomes μ_1 , the period doubles to $2T$ [Figure 8 (b)], as the bubble oscillating at the subharmonic resonance. If the parameter further changes to μ_2 , the period reaches $4T$ [Figure 8 (c)]. Then after infinitely successive doubling, there is no certain period for bubble oscillating [Figure 8 (d)], i.e., it turns into *chaotic oscillation*. The changes of the state of the bubble at particular values of the parameter are called *bifurcations*. If the limit sets (named “attractors”) of the bubble oscillator are plotted against the control parameter, a bifurcation diagram can be obtained. Figure 9 (a) illustrates the bifurcation diagram of the normalized bubble radius driven by single-frequency sound wave. In the range of the parameters of the figure, the bubble oscillates chaotically.

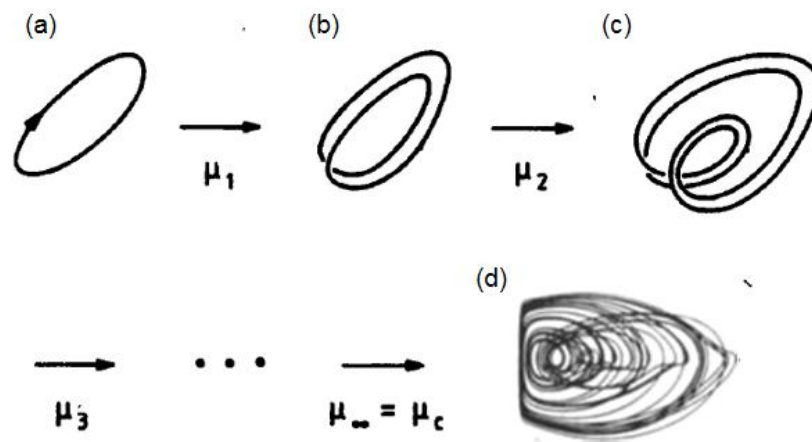


Figure 8. (Figure 8 of ref. [57]) Demonstration of route to chaos via an infinite cascade of period-doubling bifurcations. The trajectories are the phase space diagrams of bubble oscillation. μ_x indicates one of the parameters of the system.

In the chaos, a system can obey certain physical laws so that its future behaviour is deterministic while the system could exhibit unpredictable behaviour owing to the sensitivity to initial conditions [59]. It widely exists in dynamical systems. Lauterborn and his colleagues [57,60-62], have introduced research methods of chaos and deepen the understanding of the bifurcation structure and strange attractors of acoustical bubble oscillation. Because of its unpredictability, chaotic oscillation should be avoided in the practices of sonochemistry, ultrasound enhanced drug delivery and surgery. The addition of the second forcing term is one method to control chaos, which has been successively applied in pendulum system [63], Duffing oscillator [64,65] and electric circuit [66]. Behnia et al. [67] found that the addition of the second sound wave, i.e., the usage of the dual-frequency system, could also reduce the chaotic oscillations of the bubbles to the regular ones (as the comparison of the bifurcation diagrams between single- and dual- frequency shown in Figure 9). They discussed the influence of the frequency of the second wave and the phase difference between two waves. And numerical simulations were performed on the progress of HIFU tumor ablation.

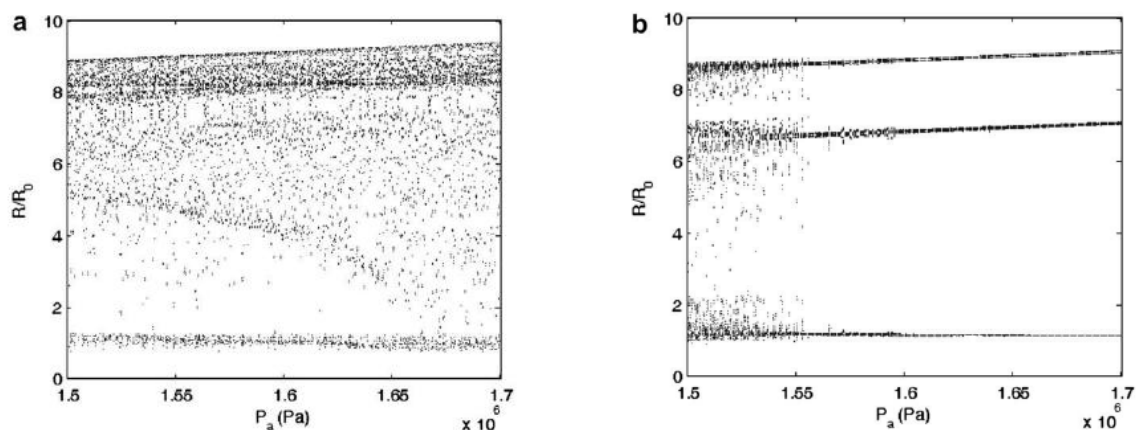


Figure 9. (Figure 1 of ref. [67]) Bifurcation diagrams of the normalized bubble radius with the initial radius $10\mu\text{m}$. a) Single-frequency excitation, driving frequency 200kHz. b) Dual-frequency excitation, driving frequencies 200kHz and 500kHz.

3. Physical mechanisms

The underlying mechanisms of the effects of multi-frequency acoustical excitations are still not clear. Moreover, for different cavitation systems, e.g., single-bubble cavitation and multi-bubble cavitation, the mechanisms of multi-frequency actions are also different. In this section, some of the mechanisms are introduced briefly.

3.1. Dissociation hypothesis

Ketterling and Apfel [68] explained the multi-frequency sonoluminescence using phase space diagrams based on the dissociation hypothesis (DH) initially proposed by Lohse and Hilgenfeldt [69]. Lohse et al. [69,70] proposed that because of the high temperature during SBSL, the nitrogen and oxygen in air bubbles may dissociate to free radicals $\bullet O$ and $\bullet N$, which will compose water solvable chemicals (e.g., NO_3^- and NH_4^+) in subsequent reactions. Meanwhile, the inert gas can accumulate in bubbles under strongly acoustically driven [70,71]. Therefore, the mass transfer through chemical reaction and rectified diffusion should be considered in SBSL. Multi-frequency acoustical excitation could enhance these effects.

Based on Ketterling and Apfel [68], a phase diagram was constructed based on calculation of the equation of bubble motion, diffusive equilibrium and the Mach criterion (assuming the ratio of the bubble wall velocity and speed of gas is larger than one), which separated the response of the bubbles into four regions, i.e., stable SL, unstable SL, stable non-SL and unstable non-SL. Compared to the experimental data of Holzfuss et al. [11], an excellent quantitative agreement was found.

3.2. Nucleation

For multi bubble cavitation, Cuiti et al. [6,17] proposed that in an ultrasound composed by two highly different frequencies, the large amount of the new nuclei generated by the added low frequency (LF) is the main factor of the enhancement of the light intensity of MBSL.

According to Figure 10, at the moment of switching the LF field off the SL intensity did not fall down immediately but, on the contrary, it increased firstly and then decreased smoothly. This phenomenon suggested that the fragments induced by bubble collapse became new nuclei with smaller radii than the initial equilibrium radii which were more likely to collapse in the high frequency (HF) field. This mechanism could also explain easily the “after effect” of the LF field on the intensity of sonoluminescence generated by the HF field in the case of switch-on the LF and HF fields sequentially (as shown in Figure 11). New nuclei generated by LF could live for such a sufficient long time that a higher sonoluminescence intensity occurred in the HF field.

Feng et al. [21] investigated the cavitation yield from the aspect of sonochemistry. They also pointed out that the production of new bubbles by the LF field is one possible mechanism of the enhancement of sonochemical efficiency.

3.3. Suppression of forming the stable clusters of cavitation bubbles

During multi-bubble cavitation, the bubbles in clusters are close to each other and interact strongly by shock waves and Bjerknes forces [72]. Therefore, bubbles deform under these interactions in the early stage of collapse. The nonspherical collapse is less efficient than spherical collapse in terms of the energy concentration, which is considered to be one of the reasons decreasing the intensity of MBSL by bubble cluster [73]. The added low frequency sound wave can impede the formation of bubble cluster by the shock waves and liquid microjets produced by collapse of large LF bubbles. So that, the total efficiency of the energy concentrated by cavitation bubbles may raise in the multi-frequency systems.

3.4. Periodical decrease in the total quasi-static pressure in the LF field

If the frequency of the LF field is substantially (10 times or more) lower than that of the HF field, then the LF field is quasi-static in relation to the HF field. The quasi-static pressure during the negative pressure amplitude half-period of the LF field lowers the cavitation threshold of the HF field by

increasing bubble size and increasing the number of bubbles driven by that field. In the compression half-period, the increase of the LF-field quasi-static pressure may cause an increase of the efficiency of the HF bubbles collapses [74-76].

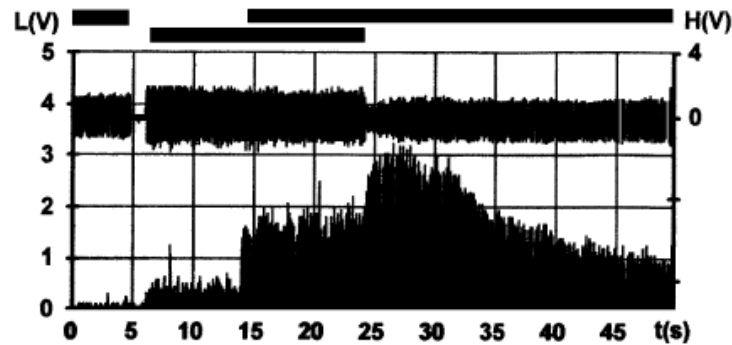


Figure 10. (Fig 2 of ref. [17]) Time history of the hydrophone output H (upper record) and of the photomultiplier L (lower record, which indicates the intensity of light emission). High frequency (HF) field parameters: pulse period 100 ms; pulse duration 2 ms; driving voltage $U_{HF} = 55V$. Low frequency (LF) field parameters: $U_{LF} = 750V$. The intensity of the ultrasound: $I_{HF} > I_{HF,th}$, $I_{LF} \leq I_{LF,th}$ (the subscript th represents the threshold of cavitation).

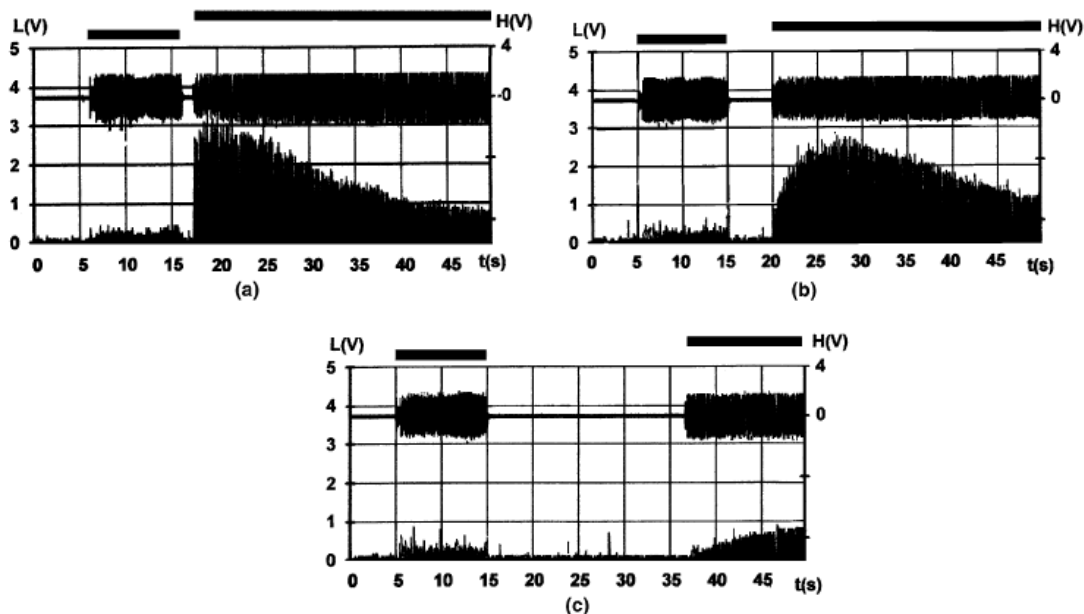


Figure 11. (Figure 3 of ref. [17]) Time history of the hydrophone output H (upper record) and of the photomultiplier L (lower record) for different time intervals Δt between low frequency (LF) field off and high frequency (HF) field on: $\Delta t \approx 2s$ (a), $5s$ (b), $22.5s$ (c). Other parameters are the same as in Figure 10.

4. Conclusion

With the increasing interests on the effects of acoustical cavitation, the multi-frequency acoustical systems also attract more and more attentions in the fields of sonochemistry, ultrasound imaging, and ultrasound enhanced biomedicine *etc.* The ability of multi-frequency systems on boosting the sonoluminescence, improving sonochemical efficiency, and promoting tumor treatment has been proved both theoretically and experimentally. And researchers have also done lots of efforts on the optimization of the multi-frequency systems.

However, compared to the wide applications of the multi-frequency acoustical systems, the mechanisms of the multi-frequency induced effects are not clear and the investigations of the fundamental issues of bubble dynamics under multi-frequency excitations are lack. For instance, the studies of Behnia et al. [67] were performed in a very narrow range of parameter zone (the amplitude of sound pressure was from 1.55 MPa to 1.7 Mpa). Actually, multi-frequency excitation may lower the critical pressure amplitude for chaotic oscillations [77], which means the addition of the second sound wave may also induce chaos. Furthermore, as far as we know, many basic problems of bubble oscillations, e.g., acoustical scattering cross section and Bjernes forces, have not been studied in the multi-frequency systems. Therefore, for a deeper understanding of bubble behaviour under multi-frequency excitation and better applications of multi-frequency systems in the fields mentioned before, a systematic investigation of the bubble dynamics under multi-frequency excitation is necessary.

Acknowledgement

The authors acknowledge the financial support from China Scholarship Council, the Ph.D. tuition fees from the University of Warwick and the EPSRC WIMRC grant (RESCM 9219), "the Fundamental Research Funds for the Central Universities" (Project No.: 2014ZD09).

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