

## Symmetric mode resonance of bubbles near a rigid boundary - the nonlinear case with time delay effects

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### Abstract

A fundamental understanding of the effect of a surface on the resonance frequency of bubbles will be useful in the future development of diagnostic medical ultrasound equipment, and specifically in the area of targeted contrast agents for the screening and possible treatment of colon cancer. In this work we turn to the wall effects on the nonlinear resonance frequency response of air bubbles in water, following on from an earlier work which considered linear interactions (E. M. B. Payne, S. Illesinghe, A. Ooi, R. Manasseh, J. Acoust. Soc. Am. 118, 2841-2849 (2005)). Numerical results for micron-sized bubbles near a rigid boundary are presented, showing the shift in frequency caused by the presence of the boundary and the presence of other bubbles. Time delay effects are also included, showing a damping of the frequency response. Simulations are limited to the special case where all bubbles are in phase (i.e., the symmetric mode), which refers to the case where all bubbles have the same initial conditions and are subjected to the same excitation pressure field. As a result they have identical time histories. An experimental method for measuring the frequency response of a single bubble attached to a surface is also briefly mentioned.

### Introduction

Nonlinear oscillations of bubbles have been studied extensively over the years ([1, 2, 3, 4]). Understanding the various mechanisms and different types of nonlinear behaviour is important especially in the field of medical ultrasound diagnostics, where coated microspheres (known as contrast agents) are injected into the body for the purpose of highlighting blockages or injured areas.

The current practice in medical contrast imaging is to drive these contrast agents at ultrasonic frequencies, thereby exciting them into nonlinear oscillations [5]. The frequency of these oscillations is then detected on an ultrasound scanning machine, which highlights the position of the group of bubbles. In targeted contrast agent imaging, the idea is that the bubbles target a specific site in the body. A bubble which is attached to its target site will thus emit a different response from a bubble which is freely flowing [6]. The difference in frequency response can be attributed to the presence of the surface, which alters the bubble dynamics [7, 8]. Modelling the effect of a surface on the frequency response of a system of bubbles is therefore important in being able to distinguish those bubbles which have attached from those which are freely flowing.

In this paper, we model a group of bubbles near a rigid surface, and show how the frequency response shifts due to the presence of the surface, as well as the presence of other bubbles. We also consider the effect of time delay, and show how time delay dampens the maximum bubble radius reached.

Time delay effects for coupled bubble system have been studied

only in recent times, and have been limited to linear oscillations [9]. We show that time delays can have a significant effect on the nonlinear frequency response, and, in addition to the other effects, may help explain discrepancies between theory and experiment.

### Theory

The theoretical model used in the present study is the Rayleigh-Plesset equation with an additional term that represents the bubble-bubble interaction,

$$R_i \ddot{R}_i + \frac{3}{2} \dot{R}_i^2 - \frac{p_{mi}}{\rho} = -\frac{p_{ext}(t)}{\rho} - \sum_{j=1, j \neq i}^N \frac{1}{r_{ij}} \frac{d(R_j^2 \dot{R}_j)}{dt}, \quad (1)$$

$$p_{mi} = p_v + \left( P_0 + \frac{2\sigma}{R_{i0}} \right) \left( \frac{R_{i0}}{R_i} \right)^{3\kappa} - \frac{2\sigma}{R_i} - \frac{4\mu}{R_i} \dot{R}_i - P_0, \quad (2)$$

where  $R_i$  is the time dependent radius of bubble  $i$ ,  $p_{ext}(t)$  is the driving pressure signal,  $\rho$  is the density of liquid water,  $r_{ij}$  is the distance between the centers of bubbles  $i$  and  $j$ ,  $p_v$  is the vapor pressure,  $P_0$  is the atmospheric pressure,  $\sigma$  is the surface tension,  $R_{i0}$  is the equilibrium radius of bubble  $i$ , and  $\mu$  is the viscosity of liquid water. This equation assumes that all the bubbles oscillate spherically and remain spherical throughout their cycle. The last term of equation (1) denotes the pressure of the sounds emitted by the other bubbles measured at the position of bubble  $i$ , and represents bubble-bubble coupling through sound. Equation (1) also assumes that the spherical bubbles are in an unbounded domain.

### A rigid wall

To include the effect of a rigid wall, we use the mirror image theory, and model the presence of the wall by assuming a mirror image of the real bubble system [7]. Assume that the distance between the center of bubble  $i$  and the wall is  $D_i$ . For simplicity, we take all  $D_i$  to have the same value equal to  $D$ , and so equation (1) is rewritten into

$$R_i \ddot{R}_i + \frac{3}{2} \dot{R}_i^2 - \frac{p_{mi}}{\rho} = -\frac{p_{ext}(t)}{\rho} - \frac{1}{2D} \frac{d(R_i^2 \dot{R}_i)}{dt} - \sum_{j=1, j \neq i}^N \left( \frac{1}{r_{ij}} + \frac{1}{s_{ij}} \right) \frac{d(R_j^2 \dot{R}_j)}{dt}, \quad (3)$$

where  $s_{ij} = (r_{ij}^2 + 4D^2)^{1/2}$  is the distance between the centers of bubble  $i$  and the mirror image of bubble  $j$ . The second term and the last term on the right-hand side, respectively, denote the amplitudes of the sounds emitted by the mirror image of bubble  $i$  and by bubble  $j$  and its mirror image.

### Time delay

Time delay is modelled by assuming a finite sound propagation between bubble  $i$  and bubble  $j$ . No time delay is modelled

between image bubbles (i.e., between bubble  $i$  and its image), nor is time delay modelled between bubble  $i$  and the image of bubble  $j$ . Thus, with these assumptions, equation (3) becomes

$$R_i \ddot{R}_i + \frac{3}{2} \dot{R}_i^2 - \frac{p_{mi}}{\rho} = -\frac{p_{ext}(t)}{\rho} - \frac{1}{2D} \frac{d(R_i^2 \dot{R}_i)}{dt} - \sum_{j=1, j \neq i}^N \left( \frac{1}{r_{ij}} \frac{d}{dt} \left( R_j^2(t-\tau) \dot{R}_j(t-\tau) \right) + \frac{1}{s_{ij}} \frac{d}{dt} \left( R_j^2 \dot{R}_j \right) \right), \quad (4)$$

where  $\tau = \frac{r_{ij}}{c}$  represents the finite time delay between bubble  $i$  and bubble  $j$ , and  $c$  is the speed of sound in liquid water.

For simplicity, we assume that all bubbles oscillate in phase ( $R_i = R_j = R$ ), have the same equilibrium radius ( $R_{i0} = R_{j0} = R_0$ ), and have the same distance between them ( $r_{ij} = r$ ). Equation (4) thus simplifies to a single nonlinear ordinary differential equation

$$R \ddot{R} + \frac{3}{2} \dot{R}^2 - \frac{p_m}{\rho} = -\frac{p_{ext}}{\rho} - \frac{1}{2D} \left( 2R \dot{R}^2 + R^2 \ddot{R} \right) - (N_{bub} - 1) \left( \frac{1}{r} \left( 2R(t-\tau) \dot{R}^2(t-\tau) + R^2(t-\tau) \ddot{R}(t-\tau) \right) + \frac{1}{s} \left( 2R \dot{R}^2 + R^2 \ddot{R} \right) \right), \quad (5)$$

where  $d(R^2 \dot{R})/dt = 2R \dot{R}^2 + R^2 \ddot{R}$ , and  $N_{bub}$  denotes the number of bubbles in the system. Equation (5) is only valid for  $N_{bub} \leq 3$ , and represents the special case where all bubbles have the same time history and are in phase.

To clarify what we mean by ‘‘in phase’’, consider two bubbles (bubble 1 and bubble 2), separated by some distance  $r$  in an unbounded domain. The bubbles have the same equilibrium radii and are subjected to the same external excitation pressure field. The external excitation pressure drives both bubbles into oscillation, resulting in an additional pressure field radiating from each individual bubble – at the same instant in time. Assuming a time delay of  $\tau$  seconds, then bubble 1’s pressure field will take  $\tau$  seconds to reach bubble 2, and bubble 2’s pressure field will take  $\tau$  seconds to reach bubble 1 (i.e., each bubble’s pressure field takes the same time to reach its respective neighbour). Because the bubbles are the same size and are driven by the same external pressure field, each one will respond in the same way to their respective neighbour’s delayed pressure field. Thus, the bubbles will have identical time histories and are considered to be ‘‘in phase’’. The same reasoning can be extended to arrangements with higher numbers of bubbles, where the bubbles are spaced equally from each other.

### Numerical Method

A simple Euler time-stepping scheme was used to solve equation (5). A time delay history buffer was needed to store the history of  $R$ ,  $\dot{R}$  and  $\ddot{R}$ , and used to calculate  $R(t-\tau)$ ,  $\dot{R}(t-\tau)$ , and  $\ddot{R}(t-\tau)$ . In all the simulations the bubble system was forced by an oscillating pressure field  $p_{ext}(t) = P_A \sin(2\pi f_{ext} t)$ , where  $P_A$  and  $f_{ext}$  are, respectively, the amplitude and frequency of the external pressure field.

Table 1 lists the physical parameter values that were used in simulations.

In all of the simulations, the system was initialised with  $R(t=0) = R_0$ ,  $\dot{R}(t=0) = 0$ , and  $\ddot{R}(t=0) = 0$ . Figure 1 shows a typical time series for a simulation with  $N_{bub} = 1$  and  $D = \infty$  (i.e.,

Symbol	Value	Units
$\rho$	1000	kg/m <sup>3</sup>
$\mu$	0.001	kg/ms
$\sigma$	0.072	N/m
$\kappa$	1	
$c$	1480	m/s
$P_0$	0.1013	MPa
$P_A$	0.01	MPa
$P_v$	2330	Pa

Table 1: Physical parameters used in simulations.

a single bubble in an unbounded domain). One can see that the radius  $R(t)$  goes through an initial transient phase before settling down to a steady-state oscillation. If the applied pressure amplitude  $P_A$  is too large then  $R(t)$  undergoes large fluctuations and does not settle down to a periodic behaviour. In such cases, the Rayleigh-Plesset equation needs modification [10, 11]. To avoid such situations  $P_A$  was set to 0.01 MPa.

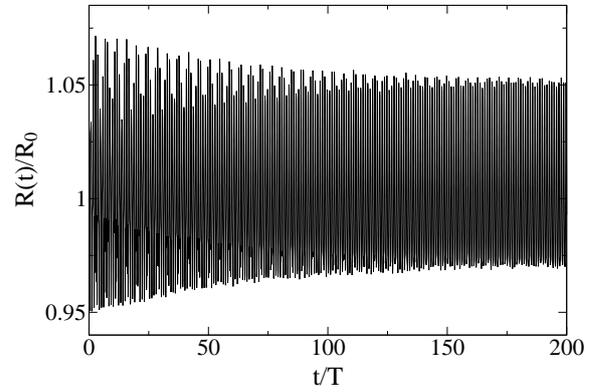


Figure 1: Time series response for a single bubble ( $N_{bub} = 1$ ) in an unbounded domain ( $D = \infty$ ).  $R_0 = 100 \mu\text{m}$ ,  $P_A = 0.01 \text{ MPa}$ ,  $f_{ext}/f_0 = 0.3$ .

Figure 2 shows a typical frequency response curve for a bubble of  $R_0 = 100 \mu\text{m}$  without time delay. This represents a plot of  $(R_{max} - R_0)/R_0$  versus  $f_{ext}/f_0$ , where  $R_{max}$  is the maximum radius of the bubble during its steady-state oscillation, and  $f_0 = \frac{1}{2\pi R_0} \sqrt{\frac{3\kappa P_0}{\rho}}$  is the linear natural frequency for a single bubble in an unbounded domain. The main resonance peak occurs in the region around  $f_{ext}/f_0 = 1$ , while harmonic resonances occur to the left and subharmonic resonances to the right of the main resonance peak. Such behaviour is in general agreement with the single-bubble calculations of Lauterborn [1].

To ensure that any effects from the initial transients can be ignored, data samples for the calculation of  $R_{max}$  were only taken from  $150 < (t/T) < 200$ , where

$$T = \max(T_0, T_{ext}), \quad T_0 = 1/f_0, \quad T_{ext} = 1/f_{ext}. \quad (6)$$

For all simulations a time step increment of  $1 \times 10^{-9}$  seconds was sufficient to ensure stability of the solution. Further decreasing the time step increment (to  $1 \times 10^{-10}$  seconds) yielded identical results and thus such accuracy was not required.

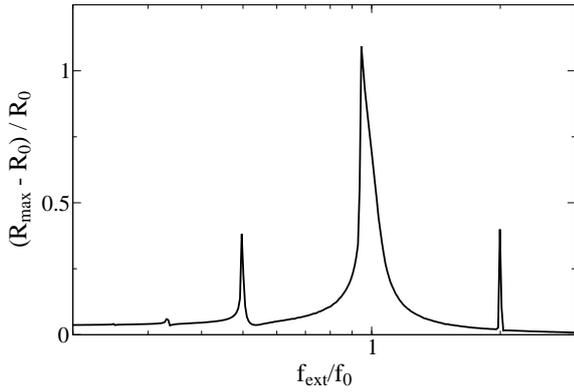


Figure 2: Frequency response for a single bubble ( $N_{bub} = 1$ ,  $R_0 = 100 \mu\text{m}$ ) in an unbounded domain ( $D = \infty$ ,  $r = \infty$ ), without time delay.

### Results

This section presents numerical results showing the effect of a rigid surface, nearby bubbles, and time delay on the frequency response of a system of bubbles.

#### The effect of a surface

Figure 3 shows a clear shift in the main resonance frequency towards lower frequencies as the distance to the boundary decreases. The same is true for the harmonic and subharmonic resonance peaks. As shown in previous work (for linear theory [7]), the effect of the wall is to increase the mass loading on the system, and thereby lower the system's resonance frequency. As the distance to the boundary decreases, the mass loading effect increases; further decreasing the resonance frequency. Another effect of the surface is that as the distance to the boundary decreases,  $R_{max}$  tends to decrease.

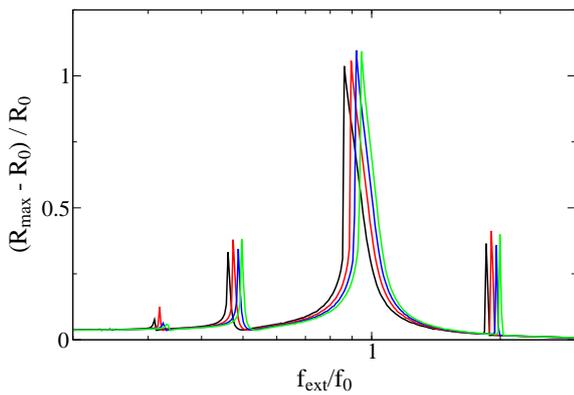


Figure 3: Frequency response for a single bubble ( $R_0 = 100 \mu\text{m}$ ) without time delay, with varying separation distance to a rigid boundary. From left to right: —  $D = 3R_0$ , —  $D = 5R_0$ , —  $D = 10R_0$ , —  $D = 50R_0$ .

#### The effect of nearby bubbles

Figure 4 shows the effect of the number of bubbles on the frequency response for up to  $N_{bub} = 3$ . As the number of bubbles in the system is increased, the resonance peaks shift towards lower frequencies. This can be explained by the fact that nearby bubbles add to the effective mass of the system, and thus lower the resonance frequency. This is in agreement with the linear

theory in [7]. Furthermore,  $R_{max}$  decreases as  $N_{bub}$  increases.

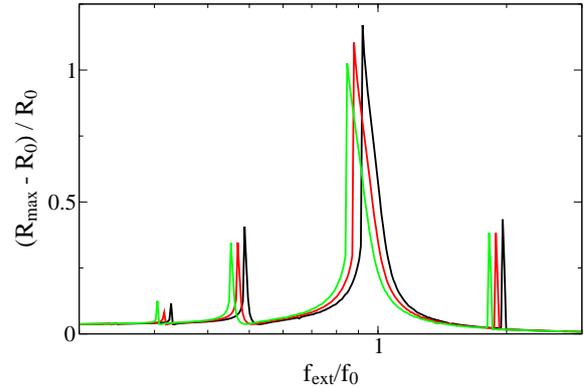


Figure 4: Frequency response (from right to left) for —  $N_{bub} = 1$ , —  $N_{bub} = 2$ , —  $N_{bub} = 3$ , near a rigid boundary, without time delay ( $R_0 = 100 \mu\text{m}$ ,  $D = 1.2 \text{ mm}$ ,  $r = 2 \text{ mm}$ ).

#### The effect of time delay

The effect of time delay on the frequency response for  $N_{bub} = 2$  and  $N_{bub} = 3$  are shown in figures 5 and 6 respectively. In both cases there is no obvious shift in the frequency response as a result of time delay, although there is a significant reduction in  $R_{max}$  around the main resonance peak. This is also true for the harmonics and subharmonics of the system. In particular, in figure 6 for  $N_{bub} = 3$ , the subharmonic around  $f_{ext}/f_0 = 1.8$  appears to be completely damped out.

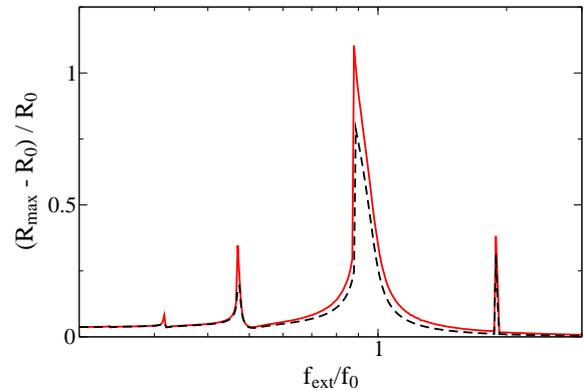


Figure 5: Frequency response for  $N_{bub} = 2$  near a wall ( $R_0 = 100 \mu\text{m}$ ,  $D = 1.2 \text{ mm}$ ,  $r = 2 \text{ mm}$ ). — without time delay, ---- with time delay.

Previous investigations of time delay effects [9] show that time delay alters the damping rather than the resonance frequency of the system. To confirm that this holds for all parameter values, a more in-depth investigation than presented here is needed.

#### Experiments

Holt and Crum [3] give experimental results for the resonance curves of an oscillating, acoustically levitated air bubble in water. Their results clearly show peaks in experimental data around the second harmonic resonance, coinciding with their numerical results. However, there seems to be no equivalent study for the case where the bubble is oscillating near a surface, or near other bubbles. The aim of future experiments, therefore,

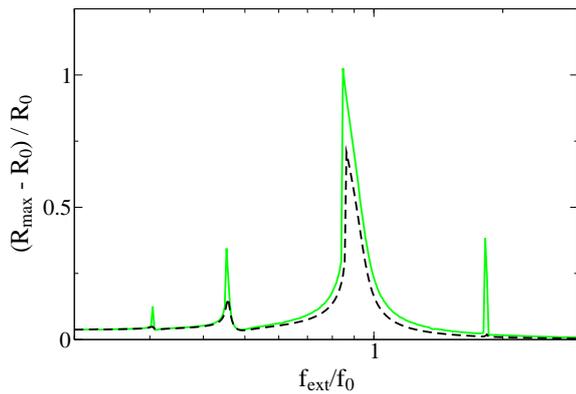


Figure 6: Frequency response for  $N_{bub} = 3$  near a wall ( $R_0 = 100 \mu\text{m}$ ,  $D = 1.2 \text{ mm}$ ,  $r = 2 \text{ mm}$ ). — without time delay, ---- with time delay.

is to measure the resonance response of a bubble near and attached to a rigid surface. This can be done by measuring the maximum radius of the bubble during its oscillation cycle (a stroboscopic technique is planned), while sweeping the excitation frequency over the expected resonance range, and identifying the peaks in the frequency response. Allowing for time delays (which were neglected in the study of Holt and Crum), may lead to better agreement with experimental data, as was the case for the study of the pressure distribution around a rising chain of bubbles [9].

### Conclusions

We have shown that, as for linear theory, the effect of a surface on the nonlinear frequency response is to cause a distinct shift towards lower frequencies. As the surface is brought closer to the bubbles, the frequency response shifts further towards lower frequencies because of the increased mass loading on the system. We have also shown that as the number of bubbles increases, the frequency response shifts towards lower frequencies. The magnitude of the resonance peaks also reduced as a result of the above factors.

For the parameters considered in this paper, the inclusion of time delay significantly dampened the harmonics and subharmonics of the frequency response, with simulations showing a significant reduction in the height of the peaks. This may help to explain future experimental results, and ultimately lead to the fine-tuning of diagnostic tools.

In terms of all the physically achievable combinations of driving pressure, bubble size, distance to the surface, and distance between bubbles, this work has only just touched the surface, and because of the nonlinearity of the response, different trends may result for other combinations. The assumptions involved in modelling time delay also need to be thoroughly explored before any general conclusions can be made regarding the effect of time delay on the resonance response.

### References

- [1] Lauterborn, W., Numerical Investigation of nonlinear oscillations of gas bubbles in liquids, *J. Acoust. Soc. Am.*, **59**, 1976, 283–293.
- [2] Leighton, T.G., Wilkinspn, M., Walton, A.J. and Field, J.E., Studies of non-linear bubble oscillations in a simulated acoustic field, *Eur. J. Phys.*, **11**, 1990, 352–358.
- [3] Holt, R.G. and Crum, L.A., Acoustically forced oscillations of air bubbles in water: Experimental results, *J. Acoust. Soc. Am.*, **91**, 1992, 1924–1932.
- [4] Feng, Z.C. and Leal, L.G., Nonlinear Bubble Dynamics, *Annu. Rev. Fluid. Mech.*, **29**, 1997, 201–243.
- [5] Lindner, J.R., Microbubbles in medical imaging: current applications and future directions, *Nature Reviews*, **3**, 2004, 527–532.
- [6] Zhao, S., Ferrara, K.W. and Dayton, P.A., Asymmetric oscillation of adherent targeted ultrasound contrast agents, *Appl. Phys. Lett.*, **87**, 2005.
- [7] Payne, E.M.B., Illesinghe, S.J., Ooi, A. and Manasseh, R., Symmetric mode resonance of bubbles attached to a rigid boundary, *J. Acoust. Soc. Am.*, **118**, 2005, 2841–2849.
- [8] Qin, S. and Ferrara, K.W., The natural frequency of nonlinear oscillations of ultrasound contrast agents in microvessels, *Ultrasound Med. Bio.*, **33**, 2007, 1140–1148.
- [9] Doinikov, A.A., Manasseh, R., Ooi, A., Time delays in coupled microbubble systems, *J. Acoust. Soc. Am.*, **117**, 2005, 47–50.
- [10] Keller, J.B. and Miksis, M., Bubble oscillations of large amplitude, *J. Acoust. Soc. Am.*, **68**, 1980, 628–633.
- [11] Lauterborn, W. and Mettin, R., Nonlinear Bubble Dynamics: Response Curves and More, *Proceedings of the NATO Advanced Study Institute, Leavenworth (WA), USA, 18–29 Aug 1997*, In: L.A. Crum et al. (eds.): Sonochemistry and Sonoluminescence, Kluwer Academic Publishers, Dordrecht, 1999, 63–72.