NONLINEAR SCATTERING OF ULTRASOUND BY BUBBLES:
NUMERICAL AND EXPERIMENTAL INVESTIGATIONS WITH
APPLICATION TO THEIR DETECTION

by

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February, 1982
Nonlinear Scattering of Ultrasound by Bubbles: Numerical and Experimental Investigations with Application to their Detection

by

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A Thesis submitted in conformity with the requirements for the Degree of Doctor of Philosophy in the University of Toronto

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I. INTRODUCTION

A. Overview

This research has been motivated by the need for information about gas bubbles which form in persons who are exposed to large reductions in ambient pressure. Three occupational groups routinely experience such pressure reductions: divers [Bennett and Elliott 1975], caisson workers [McCallum 1967], and aviators [Fryer 1969]. During decompression, or up to several hours afterward, decompression sickness may develop, with consequences ranging from mild pain to death. Over the long term, the cumulative effects of decompression may lead to tissue damage such as bone necrosis [Beckman, Elliott, and Smith 1974].

To date, noninvasive methods of monitoring the appearance of bubbles "in vivo" have been based on the scattering of ultrasound [Evans 1975]. Of the several techniques available, the most generally used relies on detection of the Doppler shifted component of ultrasound scattered by the moving targets in the blood [Spencer 1977]. Use of the Doppler principle eliminates the large masking signal from stationary tissue and the low frequency movement artifacts. At present, however, this technique is difficult for individuals untrained in its use, produces subjective results [Kisman 1977], and is inherently limited to the detection of moving bubbles. Some workers have used pulsed ultrasonic imaging (B Scan) [Rubissow and Mackay 1971, Mackay and Rubissow 1977, Daniels, Paton, and Smith 1977, 1979], to detect both stationary and moving bubbles, and localize their sites of formation. This technique is more costly and more difficult to use, especially with human subjects.

Further advances in the ability to detect bubbles may follow from an examination of some aspects of the physics of bubbles in an ultrasonic field. Beginning with Rayleigh [1917], bubble dynamics has attracted the attention of many workers, undoubtedly because it has been interesting for both theoretical and practical reasons [Flynn, 1964].

The response of a spherical bubble to an ultrasonic field is intrinsically nonlinear, which makes exact analytical solution of the problem virtually impossible unless simplifying assumptions are made. Welsby and Safar [1969] have proposed, on theoretical grounds, that this nonlinearity could be used to advantage for the detection of bubbles. This suggestion was tested experimentally with inconclusive results by Evans [1975], by Martin, Hudgens, and Wonn [1973], and by Moulinier [1978], who also discussed theoretical considerations. None of these studies, however, considered the detailed nature of the bubble dynamics.

It is the aim of this thesis to examine the nonlinearity of the oscillations of a bubble in ultrasound from a more fundamental perspective, to obtain basic information which can be used in a rational evaluation of the proposed technique. Both numerical investigations of a theoretical model, and experimental investigations are reported.
The theoretical model considers a spherical bubble, initially at rest, in an infinite liquid, in which a plane ultrasonic wave of relatively long wavelength (compared to the size of the bubble) is introduced. The actual case of a bubble in tissue or blood is far more complicated. Nevertheless, the model studied provides basic information to predict conditions under which nonlinear effects should be observable. A comparison is made with the predictions of a linearized model [Nishi 1975] and a previous numerical study of the nonlinear bubble problem [Lauterborn 1976].

The experimental model is more complex than the theoretical model, but simpler than the case of bubbles in tissue or blood. A stream of individual nitrogen bubbles rises through a beam of ultrasound of relatively long wavelength in water confined by a glass tank. Primarily because of the limitations of the ultrasonic transducers, it is not possible to make a detailed comparison between the numerical and experimental results. Instead, the level of nonlinearity indicated by the numerical results for the corresponding experimental conditions is used to predict whether nonlinear effects should be observable. The aim of the experiments was therefore simply to search for nonlinear effects in scattering by bubbles for conditions which might be considered for detection of bubbles in man. The implications of these results for a practical bubble detector are discussed in Appendix A. While the research has been directed toward diving medical applications, it may also be of interest in underwater surveillance because of the important influence bubbles have on the propagation of sound beams [Wildt 1968].

B. Review of the Literature on Acoustic Scattering by Bubbles

Scattering of sound by a gaseous sphere was considered by Rayleigh [1894, pp. 282-284], who also derived an equation for the collapse of a spherical cavity [1917]. This problem was also considered by Lamb [1932, Section 91(a)]. Minnaert [1933] provided a reasonably accurate explanation for the sounds generated by freely oscillating bubbles formed at an orifice, and derived a formula for the resonance frequency of a bubble, assuming adiabatic changes in volume of the gas within the bubble. Smith [1935] discussed bubble resonance and the destructive mechanical effects of oscillating bubbles on nearby solid objects. Plesset [1949] examined the growth and collapse of vapour bubbles in a flow past a surface. Noltingk and Neppiras [1950] studied the dynamics of a cavitation bubble formed in an ultrasonic field. They obtained numerical solutions for Rayleigh's equation, modified to allow for surface tension and an ultrasonic disturbance, assuming isothermal behaviour. In a subsequent paper, Neppiras and Noltingk [1951] discussed the combinations of the parameters, ultrasonic pressure amplitude, ultrasonic frequency, hydrostatic pressure, and bubble radius, which would lead to cavitation. Poritsky [1952] examined the effects of liquid viscosity and surface tension on the collapse and growth of a bubble. Esche [1952] performed the first spectral measurements of cavitation noise, obtaining evidence of harmonics and subharmonics.

Since these early investigations, the literature on the subject of bubble dynamics and bubble acoustics has grown considerably. Among
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ABSTRACT

The magnitude of the nonlinear effect in the scattering of ultrasound by nitrogen bubbles in water is examined for ultrasonic frequencies and amplitudes typical of those used in diagnostic medical instruments. The research is directed towards determining whether this effect could be used to detect bubbles in blood or tissue, for application in decompression research. The theory of bubble dynamics is reviewed. Numerical solutions of the modified Rayleigh equation, including the effects of acoustic, thermal, and viscous damping, and the dependence of the polytropic gas constant on frequency, are presented. A system for the spectral analysis of a single ultrasonic pulse, scattered by bubbles, is described, and the results of experiments, intended to complement the numerical investigations, are reported. For conditions typical of diagnostic ultrasound, it is shown that nonlinear scattering is significant (second harmonic greater than -40 dB with respect to the fundamental) only for the population of bubbles which are close to, or smaller than, the resonance size, for the ultrasonic frequency in use. Therefore, the use of nonlinear scattering for the detection of bubbles is limited to cases where the ultrasonic frequency is close to the resonance frequency of the bubbles, or requires variation of the ultrasonic frequency.
the papers concentrating on bubble dynamics and cavitation, there are several excellent review articles. Flynn [1964], has given definitions of such terms as 'cavitation', which often had been given different interpretations. Four models of bubble dynamics, with increasing levels of sophistication, were presented. The effect of dissipation on bubble motion was discussed, as were transient cavities, bubble nuclei, thresholds for bubble formation, and surface instabilities. The effects of cavitation on the surrounding media were described and various cavitation measures proposed. The most sophisticated of the bubble models reviewed by Flynn relied on the theory of finite amplitude effects in large bubble oscillations, discussed by Cole [1948]. The models of bubble dynamics were also discussed by Akulichev [1971], and Beyer [1974]. Akulichev presented a numerical comparison between the different models, and experimental evidence to corroborate them. A comprehensive presentation of the theory of bubble dynamics was given by Hsieh [1965]. He began with a general formulation of the problem, and followed with various simplifications, which included the assumptions of spherical symmetry and a uniform interior. Then he showed how equations describing various classes of the problems of bubble dynamics, such as collapse and oscillation, are derived. In a recent review article by Plesset and Prosperetti [1976], theoretical and experimental aspects of gas bubble dynamics, vapour bubble dynamics, and surface instabilities are discussed. Most relevant to this study is the section on gas bubble dynamics, which, beginning with Rayleigh's equation, considers linearized solutions, perturbation solutions, and numerical solutions of the nonlinear problem. The literature on the dynamics of nonspherical bubbles was also reviewed by these authors.

Lauterborn [1976] investigated the nonlinear oscillations of gas bubbles in liquids. His paper was the inspiration for the numerical part of this study, and two of his results were used for comparison, as a check of the method employed in this study. Other papers addressing the nonlinear aspects of bubble dynamics are those of Welsby and Safar [1969], Zabolotskaya and Soluyan [1972], and the many papers using a perturbation approach, employing asymptotic expansions, by Prosperetti [1974, 1975, 1976, 1977(a)]. His work has concentrated on elucidating some interesting phenomena of cavitation, such as the occurrence of subharmonic and ultraharmonic oscillations (oscillations with dominant spectral components at other than the driving frequencies or its harmonics), and the existence of a threshold amplitude for their appearance. In experimental investigations of the subharmonic response of stable bubbles, described by Neppiras [1969], it was shown that bubbles can be forced to vibrate at the first subharmonic (one half of the driving frequency) if excited at a frequency which is approximately twice their radial resonance frequency. Another approach which employed perturbation and asymptotic expansion was offered by Nayfeh and Saric [1973]. Recently, Keller and Miksis [1980] have derived an equation to deal with large amplitude bubble oscillations including the effects of acoustic radiation, viscosity, and surface tension.

There is a broad division between those papers which discuss bubble dynamics and cavitation, and those which discuss bubble acoustics,
although some authors, notably Plesset, straddle that division. Bubble acoustics generally assumes small amplitude radial vibrations and uses a linearized model for the bubble motion. Strasberg [1956] used such a model to discuss the pressure radiated by bubbles, and showed that only volume oscillations produce significant sound pressures. Devin [1959] presented a derivation of the damping constants for radially pulsating bubbles. Expressions for the total resonance damping coefficient, being the sum of thermal, acoustic, and viscous components, were derived, as was a correction for Minnaert's [1933] formula for the resonance frequency of the bubble. Hsieh and Plesset [1961], presented a theory of viscous and thermal absorption by a bubble following the classical Rayleigh approach. Kapustina [1970] reviewed some results from the linear theory of bubble motion in an ultrasonic field, and summarized theoretical and experimental findings relating to damping constants. Chapman and Plesset [1971] solved linearized equations of motion and energy for freely-oscillating gas bubbles to obtain expressions for the thermal damping constant and the polytropic exponent. Nishi [1975] considered scattering and absorption of sound waves by bubbles in viscous fluids using the classical Rayleigh approach. Results were obtained for bubbles in both water and blood at different hydrostatic pressures, the purpose being to determine the feasibility of using ultrasound to detect, in blood, bubbles caused by decompression. Use of ultrasound to detect bubbles in nonbiological applications was discussed by Medwin [1975, 1977]. Of particular importance to this study, is the paper by Prosperetti [1977(b)], in which expressions for the polytropic exponent and the thermal damping constant were developed without the assumption of resonance. The values of these parameters were shown to be dependent on the driving frequency. Recently, a new approach to acoustical scattering problems has been developed and applied to spherical bubbles by Gaunaurd and Uberall [1978], Gaunaurd, Scharnhorst, and Uberall [1979], and Uberall et al. [1979]. Some of these contributions are considered in greater detail in the next chapter, in which those aspects of the theory of acoustically driven bubble oscillations which are essential to an understanding of the problem addressed by this thesis are reviewed.

The application of ultrasound to detection of bubbles in blood has been the subject of many papers. A history of the technique to the last decade was given by Evans [1975] and was the subject of a workshop at the 1977 Annual Scientific Meeting of the Undersea Medical Society. Theoretical discussions of the technique were given by Nishi [1977] and Evans [1977]. The application of pulsed ultrasonic imaging techniques to detect bubbles in tissue was described by Rubisow and Mackay [1971], and is currently being used by Daniels, Paton, and Smith [1977, 1979]. Many papers have appeared on the application of Doppler ultrasound to decompression experiments. The history has been reviewed by Spencer [1977]. Combined theoretical and experimental results, both "in vitro" and "in vivo", with rabbits, were reported by Nishi [1972] and Nishi and Livingstone [1973]. Among the more recent contributions with human divers, are those of Johanson and Postles [1977], Guillerm and Masurel [1977], and Nishi, Kissman, Eaton, Buckingham, and Masurel [1981]. These papers indicate current practice in decompression research for bubble detection with the Doppler
technique. Monjaret, Guillerm, and Masurel [1975] determined experimentally the relationship between bubble size and amplitude of the scattered Doppler signal, verifying Nishi's [1975] theoretical predictions. These authors then developed an instrument to estimate the volume of gas detected. More recently, work on processing of Doppler ultrasonic signals has been reported by Kisman [1977] and Belcher [1980].

As already mentioned, the use of nonlinear scattering of ultrasound as a diagnostic technique for detecting bubbles was proposed by Welsby and Safar [1969]. They discussed the effects that a uniformly-distributed population of microbubbles would have on the acoustic nonlinearity of the medium. Evans [1975], with the cooperation of Welsby, tested the suggestions of Welsby and Safar experimentally. The results were inconclusive, although promising, and further research was recommended. Martin, Hudgens, and Wonn [1973], also tested the technique, and reported on a series of hyperbaric exposures in a chamber, with human subjects. These authors were optimistic about their results, but the technique was not developed further or used by them. Moulinier [1978] developed an instrument to monitor changes in the ratio of the second harmonic to the fundamental component of ultrasound transmitted through a medium to a receiver. The instrument underwent extensive "in vitro" and "in vivo" trials, the latter performed on rabbits, with a Doppler ultrasonic instrument providing a reference. His results also left a number of questions unanswered, and further development has not been pursued. The work of these authors is considered in greater detail in Chapter V.

II. A REVIEW OF THE DYNAMICS OF BUBBLES IN ULTRASONIC FIELDS

In this section, those aspects of the theory of bubble dynamics which are required for the description of the response of a bubble to an ultrasonic field are reviewed. First the forces acting on a stationary spherical bubble are described. Then a derivation of Rayleigh's equation for the collapse of a spherical cavity is given, and the additional considerations necessary for the case of a gas bubble in an ultrasonic field are described. This is followed by a discussion of solutions quoted in the literature, which were obtained by linearizing the bubble equation, by keeping second and third order terms, or by using numerical methods. The incompressible fluid model of bubble oscillations is shown to be appropriate for medical diagnostic ultrasonics. The section concludes with a review of some of the available experimental evidence pertaining to the nonlinearity of bubble oscillations.

A. Static Model

The forces acting on the surface of a stationary spherical bubble, of radius R, in an infinite liquid medium, are depicted in Figure 2.1. Interior forces are the gas pressure \( p_g \) and the vapour pressure \( p_v \). These are balanced by the hydrostatic pressure \( p_{L0} \) and the surface tension \( \sigma/2R \) (true for a sphere), where \( \sigma \) is the surface tension coefficient. The balance of forces can be written

\[
p_g + p_v = p_{L0} + \frac{2\sigma}{R}.
\]  

(1)
Figure 2.1 The forces acting on the surface of a stationary bubble of radius $R$, neglecting gravity. The hydrostatic pressure is $P_L$; $P_g$ is the gas pressure; $P_v$ the vapour pressure; and $\sigma$ is the surface energy per unit area.

This defines an equilibrium radius

$$R_e = \frac{2\sigma}{(P_g + P_v - P_L)}$$

which may be unstable.

B. Rayleigh’s Equation for the Collapse of a Spherical Cavity

The collapse of a spherical cavity in an incompressible fluid was considered by Lord Rayleigh [1917]. The equation of motion he derived served as the basis for subsequent models of bubble dynamics. The following derivation of Rayleigh’s equation follows closely that of Beyer [1974].

Consider an isolated spherical bubble of radius $R$, centered at the origin in an incompressible inviscid fluid. The equation of motion (Euler’s equation) at a point $r$ in the fluid ($r > R$) with velocity $u$ is [Skudrzyk p. 275]

$$\frac{3u}{\partial t} + \frac{3u}{\partial r} - \frac{1}{\rho_0} \frac{3p_L}{\partial r} = 0$$

where $p_L$ is the pressure in the fluid and $\rho_0$ is the density of the fluid. The equation of continuity is

$$\frac{\partial (ru)}{\partial r} = 0.$$  

For irrotational flow, one may introduce a velocity potential, $\phi$, defined by

$$u = -\frac{3\phi}{\partial r}.$$
The equation of motion is integrated from \( r \) to \( \infty \):

\[
\frac{\partial u}{\partial t} + \frac{\partial}{\partial r} \left( ur \right) = \frac{1}{\rho_0} \frac{\partial p_L}{\partial r}.
\] (6)

Substituting for \( u \) with Eq. (5) and rearranging the second term gives

\[
- \frac{3}{\rho_0} \int \frac{\partial \phi}{\partial r} \, dr + \int \frac{\partial}{\partial r} \left( \frac{u^2}{2} \right) \, dr = \int \frac{1}{\rho_0} \frac{3p_L}{\partial r} \, dr + \frac{1}{\rho_0} \left[ p_L(\infty) - p_L(r) \right].
\] (7)

Since there are no disturbances at \( r = \infty \), one may put \( \phi(\infty) = 0 \). Equation (7) becomes

\[
- \frac{3\phi}{\rho_0} + \frac{u^2}{2} - \frac{p_L(r) - p_L(\infty)}{\rho_0} = 0.
\] (8)

Likewise, integration of the equation of continuity gives

\[
u r^2 = f(t).
\] (9)

For a bubble of radius \( R \), let the surface velocity be designated \( u \), then, at any instant, the particle velocity is

\[
u(r) = \frac{ur^2}{2} = \frac{3\phi}{2r},
\] (10)

and the velocity potential is

\[
\phi(r) = ur^2/r.
\] (11)

Substituting for \( \phi \) in Eq. (8) gives

\[
\frac{1}{r} \frac{d}{dt} \left( \frac{UR^2}{2} \right) - \frac{1}{2} \frac{U^2R^4}{r^4} + \frac{1}{\rho_0} \left[ p_L(\infty) - p_L(r) \right] = 0,
\] (12)

\[
(1/r)R^2 \dot{U} + (2/r)BUR - U^2R^4/2r^4 + \rho_0^{-1} \left[ p_L(\infty) - p_L(r) \right] = 0.
\]

At the bubble walls this becomes

\[
\overline{R} \ddot{U} + (3/2)R^2 + \rho_0^{-1} \left[ p_L(\infty) - p_L(R) \right] = 0,
\] (13)

or equivalently

\[
\dot{U} + \frac{3U^2}{2R} + (\rho_0 R)^{-1} \left[ p_L(\infty) - p_L(R) \right] = 0.
\] (14)

For a spherical void (i.e., \( p_g \) and \( p_v = 0 \) with initial radius \( R_0 \), and with \( p_v(R) \) replaced by \( p_{LO} \), Rayleigh's solution gives the bubble collapse velocity as

\[
U^2 = \frac{2p_{LO}}{3\rho_0} [(R_0/R) - 1],
\] (15)

and the time for collapse as

\[
t_c = 0.915R_0 (\rho_0/\rho_{LO})^{1/2}.
\] (16)

C. Rayleigh Model Modified to Include Gas Bubbles in an Ultrasonic Field

The Rayleigh model was modified by Noltingk and Neppiras [1950] and others to include the effects of a time-varying pressure field and the presence of gas in the cavity [Flynn 1964]. The wavelength of the sound is considered to be much greater than the radius of the bubble,
and the assumption of incompressibility remains valid for a local solution. Two terms representing the hydrostatic pressure and sound pressure replace \( p(\omega) \):

\[
P_L(\omega) = P_{L0} - P_m \sin(\omega t). \tag{17}
\]

The boundary condition at the surface of the bubble is given by Eq. (1). The gas pressure \( p_g \) for radius \( R \) may be expressed in terms of initial pressure \( p_{g0} \) for a bubble initially at rest (\( R_0 = R_e \)) as

\[
p_g = p_{g0}(R_0/R)^{3\gamma}, \tag{18}
\]

where \( \gamma = 1 \) for isothermal expansion, and \( \gamma = c_p/c_v \) for adiabatic expansion; for a diatomic gas \( c_p/c_v = 1.4 \). In fact, the oscillations would be somewhere between the isothermal and adiabatic extremes, so Eq. (19) is replaced by

\[
p_g = p_{g0}(R_0/R)^{3\kappa}, \tag{19}
\]

where \( 1 < \kappa < 1.4 \).

Prosperetti [1977] has published expressions for \( \kappa \) based on a linearized theory which takes into account the heat exchange between the gas and the liquid. Numerical results are conveniently summarized in a graph of \( \kappa \) vs. \( G_1 \) with \( G_2 \) as a parameter, where \( G_1 \) and \( G_2 \) are nondimensional quantities:

\[
G_1 = \frac{MD}{\gamma R T g g}, \tag{20}
\]

and

\[
G_2 = \frac{\omega R_0^2}{D_g}, \tag{21}
\]

where \( D_g \) is the thermal diffusivity of the gas, \( R_g \) is the universal gas constant, \( T_m \) is the absolute temperature of the liquid far from the bubble, and \( M \) is the molecular weight of the gas. The depth that heat penetrates, by conduction, into the bubble during one cycle is termed the thermal penetration depth. \( G_1 \) is the square of the ratio of the thermal penetration depth to the wavelength of sound in the gas, and \( G_2 \) is the square of the ratio of the bubble radius to the thermal penetration depth.

The value of \( \kappa \) is frequency-dependent because heat conduction out of the bubble depends on three parameters, two of which are frequency-dependent. The three parameters are: bubble radius, the wavelength of the sound field in the gas \( \lambda_g \), and the thermal penetration depth \( L_g \). The parameters \( \lambda_g \) and \( L_g \) are given by Plesset and Prosperetti [1977]:

\[
\lambda_g = 2\pi(yRT_g/M)^{1/2}/\omega, \tag{22}
\]

\[
L_g = (D_g \omega)^{1/2}. \tag{23}
\]

At moderately high frequencies, \( (L_g \ll R_0 \ll \lambda_g) \), adiabatic behaviour would be observed, and at low frequencies, \( (R_0 \ll L_g \ll \lambda_g) \), isothermal behaviour would be expected. This covers the regime of interest in this thesis. For example, at 2 MHz in nitrogen, \( \lambda_g = 177 \mu m \) and \( L_g = 1.38 \mu m \). At 20 kHz, \( \lambda_g = 13.8 \mu m \).

With these considerations, Rayleigh’s equation may be written

\[
R \bar{R} + (3/2)\bar{R}^2 = \rho_0^{-1}[(p_{L0} + 2\omega/R_0 - \rho_v)(R_0/\bar{R})^3 \kappa
\]

\[
+ \rho_v - 2\omega/R - p_{L0} + p_m \sin(\omega t)], \tag{24}
\]
or equivalently

$$\dot{\mathbf{u}} + 3\mathbf{u}^2/2\mathbf{R} = (\rho_0)\mathbf{R}^{-1}[(P_{L0} + 2\mathbf{c}/\mathbf{R} - P_v)(R_0/\mathbf{R})^3 \kappa$$

$$+ \mathbf{P}_v - 2\mathbf{c}/\mathbf{R} - P_{L0} + P_m \sin\omega t], \quad (25)$$

where the driving pressure is represented by the term $P_m \sin\omega t$.

D. Solutions Obtained by Linearizing the Bubble Equation

Let $\mathbf{R} = \mathbf{R}_0 + \mathbf{r}$, with $\mathbf{r} \ll \mathbf{R}_0$. Then, by expanding $1/\mathbf{R}$ in a Taylor series, and keeping only linear terms, Eq. (25) can be simplified to

$$\frac{d^2 \mathbf{r}}{dt^2} + \omega_0^2 \mathbf{r} = \frac{P_m}{\rho_0 \mathbf{R}_0} \sin\omega t, \quad (26)$$

with

$$\omega_0^2 = (\mathbf{R}_0^2 \rho_0)^{-1}[3\kappa(P_{L0} + 2\mathbf{c}/\mathbf{R}_0 - P_v) - 2\mathbf{c}/\mathbf{R}_0], \quad (27)$$

where $\omega_0$ is the resonance frequency. Equation (26) is the familiar equation for a driven, undamped, harmonic oscillator. Thus, in the linear approximation, the bubble and surrounding fluid form a resonant system. The linear, undamped, resonance frequency is useful for comparison with the resonance frequency for the nonlinear, damped case, where it depends on the amplitude of the oscillations.

Nishi [1975] investigated the scattering and absorption of ultrasound by gas bubbles in water and blood using conventional scattering theory. Beginning with the equation of continuity for compressible fluids, the momentum equation, including gas viscosity, the equation of state, and the energy equation, he introduced small amplitude approximations and obtained a set of four linear equations in four unknowns: density, velocity, temperature, and pressure. He derived expressions for the scattering, absorption, and total cross sections for a bubble in ultrasound. His development included the effects of thermal and acoustic damping and damping due to viscosity of both the liquid and the gas. Numerical results were presented for scattering and absorption cross sections versus bubble radius at 5 MHz, for both water and blood, with hydrostatic pressure as a parameter. His numerical results also explored frequency dependence and the effects of viscosity, and were presented, as well, in the form of scattered intensities at 1 cm from the bubble.

A general linear solution to the problem of scattering of waves by cavities in visco-elastic media has recently been reported [Gaunaud et al. 1979, Gaunaud and Uberall 1978]. For the case of gas bubbles in water, these authors state that their results recover those of Nishi [1975].

Uberall et al. [1979] used this technique to show that circumferential waves are set up on the surface of the bubble, and that, when the frequency of excitation is such that the circumference equals an integer plus one-half wavelengths, the phase matching of the circumnavigating waves gives rise to resonances. The one-half wavelength term is present because the waves experience a phase jump of $-\pi/2$, when passing through convergence zones in the incident direction ($\theta=0$)
and the opposite pole ($\theta = \pi$). However, the effects of these resonances, except for the monopole case, are confined largely to the motion of gas inside the cavity. Influences on the external medium are reduced by the boundary conditions, which permit slippage between the gas and the liquid. Since, in addition, these effects occur when the wavelength is of the same order or smaller than the bubble dimensions, a regime outside our practical interest, they will not be further considered.

E. Solutions of the Nonlinear Bubble Equation

Zabolotskaya and Soluyan [1972] studied the generation of second harmonic distortion by bubbles. Their model started with a version of Rayleigh's equation which does not include the effect of surface tension; hence it would not apply to bubbles much less than about 50 $\mu$m in radius. (For a bubble of radius 50 $\mu$m at a pressure of 1 bar, surface tension adds a pressure of 0.029 bars.) They transformed their equation into one in terms of volume, and used a small amplitude approximation to keep only first and second order terms.

Their equation was then

$$\ddot{V} + \omega_0^2 V - \alpha V^2 + \beta [2\dot{V} + \dot{V}^2] + \delta_t \dot{V} = \xi \cos(\omega t + \phi)$$

(28)

with

$$\omega_0^2 = 3\gamma p_0/\rho_0 R_0^3,$$

$$\alpha = 38(r + 1)\omega_0^2,$$

$$\beta = 1/8\pi R_0^3,$$

$$\delta_t = 4\pi \rho_0/\rho_0,$$

and $V$ represents the volume perturbations; $\delta_t$ is the total damping coefficient at resonance.

Zabolotskaya and Soluyan stated that this equation has a solution which includes a term at the driving frequency $\omega$ and one at twice the frequency $2\omega$. They gave the amplitude of the second harmonic volume oscillations as

$$V_2 = \frac{\eta^2(a - 3\omega_0^2)A^2}{2[(\omega_0^2 - \omega^2)^2 + \omega \delta_t^2][(\omega_0^2 - 4\omega^2)^2 + 16\omega \delta_t^2]^{1/2}}.$$

(29)

According to this result, the amplitude of the second harmonic peaks at two frequencies - one half the resonance frequency, and the resonance frequency, the latter peak being of higher amplitude. A plot of this equation is shown in Figure 2.2.

Prosperetti [1974] investigated steady state nonlinear oscillations of bubbles using an asymptotic expansion method. The equation solved was

$$R \ddot{R} + (3/2) \dot{R}^2 = \frac{1}{p_0} [p_L(t) - 2\alpha/R - (4\mu/R)\dot{R}],$$

(30)

where

$$p_L(t) = p_{LO}(1 - p_m \cos\phi_0),$$

$$p_0(t) = p_{BO}(R/R_0)^{3\gamma},$$

and $\mu$ is the coefficient of shear viscosity. The substitution $R =
Second Harmonic Volume Oscillations

![Graph](image)

Figure 2.2. A plot of the expression of Zabolotskaya and Soluyan [1971] for the amplitude of the second harmonic volume oscillations $V_2$, for a bubble with an initial resting radius $R_0$ of 50 μm. The abscissa represents the driving frequency. The resonance frequency of the bubble is 65 kHz.

$R_0(1+x)$ was made, the power series expanded, and terms up to the third order were kept. Prosperetti solved the equation thus obtained, and put the solution in a similar form for the case of second and third harmonics and the one-half and one-third subharmonics. A slightly different form was required for the fundamental resonance. As these equations are lengthy, and will not be required here, they will not be written out, but a qualitative description of their implications will be given.

The term 'harmonic' as used in the literature of bubble dynamics does not refer to the frequency components of a real signal, but rather to peaks in the frequency response of the bubble. When the frequency of oscillation of the bubble corresponds to the driving frequency, but also corresponds to an integer ($n$) submultiple ($1/n$) of the resonance frequency, that oscillation is termed a harmonic of order 'n'. When, instead, the frequency of oscillation is a submultiple of the driving frequency, but corresponds to the resonance frequency of the bubble, that oscillation is termed a subharmonic. Ultraharmonics refer to oscillations which have a dominant frequency different from both the resonance and driving frequencies, but integer multiples or submultiples of both. For example, an oscillation with frequency one third of the resonance frequency and one half of the driving frequency is termed an ultraharmonic of order 3/2. In this thesis, when the terms fundamental, first harmonic, second harmonic, etc., are used, they have their usual meaning in terms of the frequency components of a real signal.
In the region of the one-half subharmonic there were three branches of the solution for the subharmonic amplitude; one proved to be zero, while another was unstable. The solution demonstrated the existence of a subharmonic threshold given by

\[ \xi = \frac{(3\Omega - W)^{3/2}}{9\Omega^{2} - W} \frac{24\nu_{\text{eff}}}{R_{0}(\rho_{g} \rho_{0}^{2})}, \]  

(31)

where

\[ W = 2\nu_{\text{eff}} \rho_{0} \rho_{g}^{2}, \]

\( \nu_{\text{eff}} \) is an effective viscosity, accounting for thermal, acoustic, and viscous damping, and \( \xi \) is defined by

\[ \xi = (1-\nu) \rho_{m}. \]

The existence of a threshold for subharmonic oscillations has been observed experimentally [Esche 1952, Neppiras 1969].

In the region of the harmonics, the characteristic equation is a cubic and a zero amplitude solution is no longer valid. Also, there is no threshold for harmonic oscillations, although for sufficiently large driving pressures, the solution can become multi-valued. Far from resonance, the amplitude is very small. Good agreement was obtained with Lauterborn's numerical solutions, to be described next.

Lauterborn [1976] carried out extensive numerical investigations of bubble oscillations with a view to elucidating the occurrence of subharmonic components in cavitation noise. His model was basically that of Eq. (25), with the introduction of a term to include the effects of viscous damping. The equation solved was

\[ \ddot{R} + \frac{(3/2)R^{2}}{\rho_{g} \rho_{0}^{2}} + (3/2)R \frac{\rho_{v}}{\rho_{g} \rho_{0}^{2}} + \frac{\rho_{0}}{\rho_{g} \rho_{0}^{2}} \frac{(4\nu/R)R}{R_{0} \rho_{g}^{2}} \]

\[ = (4\nu/R) \frac{R}{R_{0}} \rho_{v} \sin \omega t, \]

(32)

where

\[ \rho_{g} \rho_{0}^{2} = \rho_{L} - \rho_{v} + 2\nu/R_{0}, \]

and \( \nu \) is the coefficient of shear viscosity. This equation differs from that of Prosperetti only in the inclusion of the vapour pressure \( \rho_{v} \).

Lauterborn's calculations were done for three different bubble radii: 0.1, 1, and 10 \( \mu \)m, corresponding to cases of heavy, moderate, and light viscous damping respectively. The other parameters varied were frequency and driving pressure. Vapour pressure was assumed to be small and constant (0.0233 bar), and the polytropic gas constant was assigned the value 1.33.

Lauterborn's results show that fairly complicated bubble motions can occur at moderate sound pressure levels - a few tenths of a bar. Harmonics, subharmonics, and ultraharmonics were observed in the numerical solutions. Lauterborn summarized his results in the form of graphs showing amplitude of the bubble radial response as a function of frequency, with driving pressure as a parameter. These curves
might be termed excitation spectra. They show that increased amplitude broadens resonances and shifts them toward lower frequencies, and that the onset of ultraharmonics and subharmonics appears to have a threshold which agrees with the analytical formula (Eq. 32) deduced by Prosperetti [1974].

Bubble oscillations tended to become large and unstable at amplitudes close to the Blake threshold for uncontrolled growth beyond a critical radius [Flynn 1964] given by

$$P_t = P_{LO} - P_v - 4\sigma/(3\sqrt{R_0}(1 + (P_{LO} - P_v)R_0/2\sigma)^{1/2}), \quad (33)$$

a result derived from static theory. The critical radius is

$$R_c = 4\sigma/3|P_{LO} - P_v|. \quad (34)$$

The bubble oscillations for the regime Lauterborn studied demonstrated nonlinear behaviour, but the transition from linear to nonlinear behaviour was not discussed. The details of this transition, for conditions common in diagnostic medical ultrasonics, are the primary interest of this study.

F. Models Including Higher Order Effects

Most of the models for bubble oscillations discussed so far assume that the surrounding liquid is incompressible. In such models the speed of sound is infinite, and no account is taken of dissipation due to acoustic radiation, unless it is included as part of a phenomenologically-derived damping parameter.

Several more complex models exist to take care of acoustic radiation damping and other higher order effects which become more important as bubble oscillations grow in amplitude. These models are reviewed by Flynn [1964], Akulichev [1971], and Beyer [1974]. The next most sophisticated model introduces the acoustic approximation, in which a slight compressibility is assigned to the liquid; some energy is therefore lost through propagation of a spherical acoustic wave. Viscosity is included only in the boundary condition

$$p_L(t) = p_g + p_v - 2\sigma/R - 4\mu \nu/R. \quad (35)$$

The next level of complexity is found in a model attributed to Herring and Flynn (see Akulichev [1971]). In it, energy storage due to liquid compressibility is included. The differences between the predictions of this model and those of the acoustic approximation appear only in the final stages of bubble collapse.

Finally, the finite amplitude model deals with cases for which the bubble wall velocity approaches or exceeds the speed of sound in the liquid. This model is attributed to Kirkwood and Bethe, who developed it in 1942 to describe underwater explosions, and to Gilmore who applied it to bubbles (see Flynn [1964] or Cole [1948]).

A comparison of the predictions of these three models, carried out by Akulichev [1971], shows that they agree up to pressure ampli-
tudes of around 40 bars. Therefore, to describe the motion of a bub-
ble in low amplitude ultrasound, the incompressible fluid model is
adequate.

G. Nonspherical Bubbles

In all models discussed to this point, it was assumed that the
bubbles were spherical. Relaxation of this restriction greatly com-
plicates the analysis [Plesset and Prosperetti 1976]. Although, in an
unbounded fluid, bubbles can only become nonspherical by amplification
of pre-existing perturbations, in fact the necessary conditions are
generally present. Buoyancy, proximity to boundaries, and the
inherent instability of contracting bubbles are examples of perturbing
factors.

In analyses where bubbles may be nonspherical, the bubble surface
is usually specified in terms of spherical harmonics. Plesset [1954]
solved the bubble equation for a free surface, thus specified, in an
unbounded, incompressible, inviscid fluid assuming small amplitude
effects. Bubble instability and eventual break up during collapse was
predicted, an effect that has been observed experimentally. The col-
lapse of a bubble near a boundary was studied numerically by Plesset
and Chapman [1971] and their predictions were verified experimentally
by Lauterborn and Bolle [1975]. The considerable amount of other work
which has been done in this field is reviewed by Plesset and Pros-
peretti [1976]. In the numerical analysis which follows, only spheri-
cal bubbles are considered.

H. Experimental Results

Most available experimental results which demonstrate the non-
linear response of bubbles to ultrasound have been obtained for cavi-
tating bubbles. Usually these bubbles were produced ultrasonically in
fields of at least moderate amplitude. The experiments to be reported
in Chapter IV differ fundamentally in that the bubbles were not pro-
duced by cavitation and the fields were pulsed and generally of lower
amplitude and higher frequency. This is a regime of greater interest
to diagnostic medical ultrasonics, since it is hoped that bubbles will
be detected - not produced.

Esche [1952] performed the first spectral measurements of the
sound field emitted by bubbles. These measurements spanned a fre-
quency range from static to 3.3 MHz using a variety of transmission,
reception, and monitoring techniques. He observed the presence of
harmonics, ultraharmonics, and subharmonics superimposed on a contin-
um. The harmonics and ultraharmonics occurred first with increasing
sound pressure amplitude, while the appearance of subharmonics and the
continuum coincided with the violent growth and collapse of the bub-
bles. Esche used the appearance of the continuum to define the onset
of cavitation. An especially strong subharmonic was often observed at
one half the driving frequency and a one-third harmonic was observed
when the sound field intensity was sufficiently high.

Akulichev [1972] performed similar experiments, but in addition,
used high speed photography to observe the bubble motions. A barium
titanate ring transducer was driven at 10 kHz so that cavitation was produced along the axis of the transducer. The sound pressure in the ring was measured using a hydrophone with flat frequency response up to 500 kHz, and a spectrum analyser. The spectra were rich in harmonic content. In tap water, cavitation was well developed at 10 kHz for sound pressure amplitudes of 0.4 bars; subharmonics were evident at 0.6 bars; and a continuum accompanied vigorous cavitation at 0.8 bars.

Akulichev compared photographic records of bubble growth and collapse obtained in experiments with numerical predictions obtained using the incompressible fluid, acoustic, and finite amplitude models. These showed remarkable agreement for a bubble with equilibrium radius 1.0 μm, in sound fields with pressure amplitudes 1.75, 2.0, and 2.75 bars, and frequency 10 kHz.

Neppiras [1969] conducted experiments with single bubbles, in which he showed that bubbles can be forced to oscillate at the first subharmonic if the driving frequency is close to twice the radial resonance frequency of the bubbles. His experiments showed that the subharmonic response grows rapidly beyond a certain level, and that this threshold is slightly lower for stable bubbles than the threshold in a liquid without bubbles, where the subharmonic appears with the onset of cavitation. The existence of other low frequency spectral components, possibly caused by the shock excitation of large bubbles was also demonstrated.

III. NUMERICAL INVESTIGATIONS

A numerical study was carried out in order to determine the non-linearity of the response of bubbles to ultrasound at frequencies and levels encountered in medical diagnostic ultrasonics. In this way, the efficacy of using this nonlinearity as an aid in bubble detection or identification could be determined.

A. The Equation Solved

The equation solved was basically that studied by Lauterborn [1976] (Eq. 33), except that the viscous damping term was replaced by a total damping term which included the viscous, acoustic, and thermal components. The form of this term is

\[ \dot{\delta}_t = \dot{\delta}_\mu + \dot{\delta}_{ac} + \dot{\delta}_{th} \]  

(36)

where

\[ \dot{\delta}_\mu = \frac{4\mu}{\omega_0 R_0^2}, \]  

(37)

\[ \dot{\delta}_{ac} = (\omega R_0/c)[1 + (\omega R_0/c)^2]^{-1}, \]  

(38)

\[ \dot{\delta}_{th} = (\omega_0/\omega)^2 B, \]  

(39)

and B is the thermal damping constant of Prosperetti [1977]. The origin of Equations (36) through (39) is discussed in Appendix B. The ordinary, second order, nonlinear, differential equation solved was

\[ \rho_0 \ddot{R} + \frac{3}{2} \dot{R}^2 = p_0 \left( \frac{R_0}{R} \right)^3 + p_v - p_{L_0} - \frac{2\alpha}{R} \]  

(40)

\[ - \delta_\mu \omega_0 R \dot{R} - p(t), \]
where
\[ P_{g0} = \frac{2\sigma}{R_0} + P_{LO} - P_v. \]

The values of \( \kappa \) and \( B \) are dependent on the equilibrium radius and frequency, and were usually obtained from tables provided by Prosperetti [1977], interpolating between tabulated values where necessary. For the initial studies, values of the resonance damping constant as given by Nishi [1975] were used, and for studies of free oscillations, values of \( \kappa \) and \( \delta_r \) were obtained from Chapman and Plesset [1971].

### B. Method

Equation (40) was first written as a system of two first-order equations:

\[ U = \dot{R}, \]
\[ \rho_0 \dot{R}^2 + \frac{3}{2} \rho_0 U^2 = P_{g0} \frac{R_0}{R} \dot{R}^2 + P_v - P_{LO} - \frac{2\sigma}{R} - \dot{\omega} \rho_0 U. \]  

This set of equations was solved using the fourth-order predictor-corrector pair of algorithms of Adams-Bashforth and Adams-Moulton [Conte and de Boor, pp. 336-367]. For this algorithm, four starting points are required. The first point was determined by the initial conditions, and the next three were obtained using the fourth-order Runge-Kutta algorithm. An estimate of the next point in the solution is made using the predictor, and then that value is used by the corrector to obtain an improved estimate. In principle, the process of correcting can be repeated any number of times. An advantage of this algorithm is that the difference between the predicted and corrected values can be used to estimate the magnitude of the local discretization error [Conte and de Boor, p. 352], and, if desired, this error can be reduced, either by repeating the correction process, or by reducing the step size. Since it is usually not efficient to use more than one correction before changing the step size, the practice of making only one correction was adopted here.

To define the problem, a number of parameters and initial conditions had to be specified. Most of the parameters were not varied but were set to values appropriate for 20°C water at 1 atm. These parameters are listed in Table 3.1.

<table>
<thead>
<tr>
<th>TABLE 3.1. Parameters of the problem.</th>
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<tbody>
<tr>
<td>Hydrostatic pressure</td>
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<tr>
<td>Density of water</td>
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<tr>
<td>Vapour pressure</td>
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<tr>
<td>Coefficient of surface tension</td>
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Initially, the bubble was at rest and, usually, at its equilibrium radius, which was specified at run time. A number of parameters which describe the nature of the ultrasonic excitation also had to be specified. The excitation was of the form

\[ p(t) = A f(t) \cos(\omega t + \phi), \]

where
\[ f(t) = \begin{cases} 
0.5 - 0.5 \cos(\pi t / t_m), & 0 \leq t \leq t_m, \\
1.0, & t_m < t < t_f - t_m, \\
0.5 - 0.5 \cos[\pi(t_f - t) / t_m], & t_f - t_m < t < t_f;
\end{cases} \]

and
\[ A = \text{amplitude of excitation}, \]
\[ \omega = \text{frequency of excitation}, \]
\[ \phi = \text{phase of excitation}. \]
$t_f$ was the width of the pulse and $t_m$ was defined as a percentage of $t_f$, both specified at run time. The purpose of $f(t)$ was to simulate ultrasonic pulses from real transducers, which do not respond instantaneously to voltage steps. The parameter, $A$, was the amplitude of the ultrasound in bars; $\phi$ was the phase, normally set to $\pi/2$; and $\omega$ was the angular frequency. When pulse excitation was used, the pulse width was also specified. Other parameters associated with the algorithm and specified at run time were: step size, in terms of the number of points per ultrasonic period; and the total number of points, $N$, to be obtained in the solution.

The solution was presented as an $N \times 3$ array, with the three columns being time, radial velocity, and radius. This array was then reduced to one of dimensions $1024 \times 2$, usually representing time and radial velocity. The length of the reduced array was restricted to $1024$ for input to a Fourier transform program. Radial velocity was normally included in the reduced array because ultrasonic transducers respond to pressure, and the pressure radiated by a spherical source is proportional to the derivative with respect to time of its volume velocity [Kinsler and Frey, p. 164]. The far-field pressure from a spherical source can be written

$$p = (A/r)\sin(\omega t - kr),$$

where $A$, the complex amplitude is given by

$$A = R_0 U_0 \rho_0 c R_0 (kR_0 + j) / (1 + k^2 R_0^2 (\cos kR_0 + jsinkR_0)).$$

The magnitude of $A$ is

$$|A| = \frac{R_0 U_0 \rho_0 c (kR_0)}{(1 + k^2 R_0^2)^{1/2}}. \quad (47)$$

For $k R_0 \ll 1$, this reduces to

$$A = R_0^2 U_0 \rho_0 \omega. \quad (48)$$

The time domain representation of the solution was plotted and the array was passed to a Fast Fourier Transform (FFT) program which calculated the periodogram [Bendat and Piersol p. 315, Oppenheim and Schafer, p. 542]. The periodogram $I_N(w)$ may be defined as follows:

The discrete Fourier transform of a sequence $x(n)$, $0 \leq n < N-1$, is

$$X(e^{jw}) = \sum_{n=0}^{N-1} x(n) e^{-jwn},$$

and

$$I_N(w) = \frac{1}{N} |X(e^{jw})|^2. \quad (50)$$

The periodogram may be considered a raw estimate of the power spectral density of the measured data. The discrete Fourier transform uses a truncated series of data, which is equivalent to multiplying the original data by a box car function. In the frequency domain, this is equivalent to performing a convolution of the data with a function having a narrow main lobe (dependent on the duration of the data sequence) and significant side lobes (up to 20% of the main lobe), half of which are negative. This procedure may lead to the
The computation of erroneous power results [Bendat and Piersol, p. 316].
The problem has been alleviated by multiplying the original data with
a cosine taper, Eq. (44), instead of the boxcar function. The power
spectrum of this function has a wider main lobe and smaller side
lobes. The cosine taper was convenient to use because it was provided
as an option with the periodogram program.

The logarithm of the periodogram of the radial velocity was plot-
ted. Examples of plots of time and frequency domain representations
of the excitation pulse (f = 1 MHz, A = 0.017 bar, width = 16 µs,
taper = 40%), and of the bubble’s response (R = 2 µm), are shown in
Figures 3.1 and 3.2. Nonlinearity of the bubble’s response can be
detected by the appearance of harmonics in the periodogram. The level
of these harmonics, relative to the fundamental, can be obtained in
terms of dB re 1 m/s, by subtracting the ordinate values and mul-
tiplying by 10 (since the periodogram is proportional to velocity
squared). The periodogram gives relative information about the levels
of the harmonics, because the calculated magnitude is dependent on the
pulse duration, the width of the time window, and the amount of the
cosine taper used. In the example, the second harmonic of the radial
velocity response is 30 dB below the fundamental, and the third har-
monic is 63 dB down. To adjust for the frequency dependence of a
spherical source (Eq. 46), correction factors were applied to the har-
monic levels obtained from the radial velocity response. In the case
of a very small bubble (kR0 << 1) the pressure amplitude is propor-
tional to frequency. Hence the correction factors would be 6.02 dB
for the second harmonic and 9.54 dB for the third harmonic. These
Corrections would apply to the case in Figure 3.2. Hence, in this case, the radiated pressure would have second and third harmonic components 24 and 54.5 dB below the fundamental component, respectively.

C. Results

1. Frequency Much Greater Than Resonance

Since the numerical investigations were intended to complement experimental investigations, the cases studied numerically often reflected the conditions present in the experiments. Thus, the earliest simulations were for bubbles of radius 50 μm in a continuous ultrasonic field, of frequency 1 MHz, which was simply turned on at time zero. The linear resonance frequency, neglecting damping, is given by Chapman and Plesset [1971]

$$\omega_0^2 = \frac{3K\rho_0(1 + 2\sigma/\rho_0R_0)/\rho_0R_0^2 - 2\sigma_0R_0^3}{\rho_0R_0^2}.$$  (51)

For a bubble of radius 50 μm, this gives 64 kHz, approximately. Hence, the driving frequency was greater than the resonance frequency by a factor of 16. The amplitude was varied between 0.1 and 1.2 bars. The value of $K$ was set to 1.33, following the example of Lauterborn [1976], and $\sigma$ was set to its resonance value, $\sigma_t = 0.096$, as given by Nishi [1975]. Although, in light of Prosperetti's work, these values were not accurate, it was observed that for frequencies much above resonance, the nonlinearity of the bubble's response was not sensitive to variation of these parameters. Prosperetti's results indicate $K = 1.36$ and $\delta_t = 0.20$. Despite the relatively high driving amplitudes...
chosen, the results demonstrated that the second harmonic component was always less than -70 dB with respect to the fundamental. By any practical definition, therefore, the bubble was behaving linearly. (Most sinusoidal electronic oscillators have a higher distortion specification.)

In later numerical studies, the form of the driving pressure was changed to that of a pulsed sinusoid, because this change had proved to be necessary in the experiments. The initial pulsed studies involved a bubble of radius 50 μm, driven by ultrasound of frequency 2 MHz, for pulse durations ranging from 1 to 16 μs, and pulse amplitudes ranging from 0.05 to 0.4 bars. To simulate better the response of a practical transducer (Section IV. B. 4.), different amounts of cosine tapering were tried, Eq. (44). Then, while the taper was set to a constant 10%, and the total pulse width was set to eight cycles of the ultrasonic wave, the ultrasonic frequency and amplitude were varied.

A second harmonic component was not evident until the frequency was reduced to 128 kHz, approximately twice the bubble resonance frequency. Then, for a driving amplitude of 0.2 bars, the second harmonic component was 35 dB below the fundamental. Nevertheless, for frequencies far above resonance, one effect of the bubble response was evident - the lower frequency components were increased at the expense of the higher frequency components, when compared with the periodogram of the driving pulse. This was an effect of the transient resonant oscillation, excited by the pulse, which appeared as a modulation of the response at the driving frequency. The effect, illustrated in Figure 3.3, was particularly noticeable for sharp pulses. In Figures 3.3(a) and 3.3(b), a numerical excitation pulse of frequency 500 kHz, width 16 μs, amplitude 0.05 bars and taper 10%, is shown along with its periodogram. The calculated response of a bubble, of radius 50 μm, to the driving pulse in Figure 3.3(a), is shown in Figure 3.3(c). The periodogram of the bubble's response is shown in Figure 3.3(d); it can be seen that the lower frequency components are emphasized, particularly in the neighbourhood of the bubble's resonance frequency.

2. Near Resonance

Although a frequency of 1 MHz is at the low end of the range (1-10 MHz) commonly used in diagnostic medical ultrasonics, it is too high to produce a nonlinear effect in a bubble of radius 50 μm. As this was evident in the preliminary CW studies, the frequency was reduced, and varied between 60 and 70 kHz, a range which includes the linear resonance frequency of the bubble. The amplitude was set to 0.1 bars originally, and the same values of κ and δ were used, 1.33 and 0.096, respectively. The value for δ was appropriate, having been obtained with the assumption of resonance, but the value for κ was high. For comparison, Prosperetti's [1977] results give 1.173 as the appropriate value for this parameter.

Figure 3.4 shows the logarithm of the fundamental and second harmonic components of the bubble radial velocity oscillations, and the logarithm of the ratio of these components, as a function of
The fundamental component peaks at 64 kHz, while the second harmonic component is maximal between 63.5 and 64 kHz. The maximum value of the ratio of the second harmonic to fundamental component is approximately -8.3 dB at 63.5 kHz.

Near resonance, the nonlinearity was sensitive to variation in \( \delta_t \). When \( \delta_t \) was reduced by 4.2%, the ratio of the second harmonic to fundamental increased by 8.6%. When \( \delta_t \) was increased by 4.2%, this ratio decreased by 5.9%.

When the form of the excitation was changed to a pulsed sinusoid, similar values for the ratio of second harmonic to fundamental components were observed. For pulsed excitation, with a 65 kHz centre frequency, the sensitivity of bubble nonlinearity to variation of the damping constant was further examined. The results, plotted in Figure 3.5, show that the amplitude and nonlinearity of the response becomes more sensitive to changes in the value of the total damping as it is reduced below about 0.1. Since the theoretically-predicted value for the damping constant lies on the rapidly changing part of this curve, a measurement of the ratio of second harmonic to fundamental components, could serve as a check of the theoretical value, given by Eq. (36) through (39).

The effect of varying the polytropic gas constant was also studied for these conditions. A shift of the resonance frequency, given by Eq. (51), was the primary effect of this variation. The nonlinearity of the response, as indicated by the ratio of second harmonic to
Figure 3.4 The logarithm of the radial velocity response (magnitude squared), as a function of frequency, for a bubble of radius 50 μm in an ultrasonic field of amplitude 0.1 bars.
(a) The fundamental component.
(b) The second harmonic component.
(c) The ratio of second harmonic to fundamental components.

Bubble Radial Velocity vs. Frequency

Figure 3.5 A plot showing the effect of altering the damping constant on the nonlinearity of the response of a bubble of radius 50 μm, driven at resonance.
(a) The fundamental component of the radial velocity.
(b) The second harmonic relative to the fundamental.
(c) The third harmonic relative to the fundamental.
fundamental components in Table 3.2, was not much altered, especially when the driving frequency was allowed to track the resonance frequency.

**TABLE 3.2 The Effect of the Polytropic Exponent on Bubble Nonlinearity**

<table>
<thead>
<tr>
<th>$\kappa$</th>
<th>$f$ (kHz)</th>
<th>$f_r$ (kHz)</th>
<th>$U_1$ (dB)</th>
<th>$U_2/U_1$ (dB)</th>
<th>$U_3/U_1$ (dB)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.40</td>
<td>65.0</td>
<td>65.6</td>
<td>-16.2</td>
<td>-17.5</td>
<td>-33.2</td>
</tr>
<tr>
<td>1.33</td>
<td>65.0</td>
<td>64.0</td>
<td>-16.3</td>
<td>-18.0</td>
<td>-33.8</td>
</tr>
<tr>
<td>1.22</td>
<td>65.0</td>
<td>61.3</td>
<td>-19.9</td>
<td>-19.8</td>
<td>-37.5</td>
</tr>
<tr>
<td>1.22</td>
<td>61.3</td>
<td>61.3</td>
<td>-16.1</td>
<td>-17.4</td>
<td>-31.7</td>
</tr>
</tbody>
</table>

In this table, $\kappa$ is the polytropic exponent; $f$ is the driving ultrasonic frequency; $f_r$ is the linear resonance frequency of the bubble; $U_1$ is the fundamental component of the bubble's radial velocity, in dB re. 1 m-s$^{-1}$; and $U_2$ and $U_3$ refer to the second and third harmonic components respectively.

3. Frequency Much Less Than Resonance

In the initial CW numerical investigations, no cases were studied for which the ultrasonic frequency was much less than the resonance frequency of the bubble. In the initial pulse investigations, however, some cases were studied, for which the driving frequency was one half, one third, or two thirds of the resonance frequency. The equilibrium radius of the bubble was 50 $\mu$m, and the amplitude of the ultrasonic pulse was 0.05 bars. The results showed heavy levels of distortion. The greatest value for the ratio of the second harmonic to fundamental component, was observed for a driving frequency equal to one half the resonance frequency, but the overall level of the response was reduced. Likewise, the ratio of the third harmonic to the fundamental component was greatest when the driving frequency was one third the resonance frequency. These results are summarised in Table 3.3. Similar results for smaller bubbles at higher resonance frequencies are presented in Section III. B. 4.

**TABLE 3.3 Summary of Results for Frequencies Below Resonance**

<table>
<thead>
<tr>
<th>$R_e$ (µm)</th>
<th>$f$ (kHz)</th>
<th>$f/f_r$</th>
<th>$A$ (bar)</th>
<th>$U_1$ (dB)</th>
<th>$U_2/U_1$ (dB)</th>
<th>$U_3/U_1$ (dB)</th>
</tr>
</thead>
<tbody>
<tr>
<td>50</td>
<td>32.0</td>
<td>0.500</td>
<td>0.05</td>
<td>-40.3</td>
<td>-3.3</td>
<td>-21</td>
</tr>
<tr>
<td>50</td>
<td>21.3</td>
<td>0.333</td>
<td>0.05</td>
<td>-45.3</td>
<td>-22</td>
<td>-5.8</td>
</tr>
<tr>
<td>50</td>
<td>42.6</td>
<td>0.667</td>
<td>0.05</td>
<td>-36</td>
<td>-20</td>
<td>-38</td>
</tr>
<tr>
<td>5</td>
<td>100</td>
<td>0.165</td>
<td>0.017</td>
<td>-50</td>
<td>-39</td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>100</td>
<td>0.165</td>
<td>0.054</td>
<td>-40</td>
<td>-29</td>
<td>-56</td>
</tr>
<tr>
<td>3</td>
<td>100</td>
<td>0.100</td>
<td>0.017</td>
<td>-56</td>
<td>-40</td>
<td></td>
</tr>
<tr>
<td>3</td>
<td>100</td>
<td>0.100</td>
<td>0.054</td>
<td>-46</td>
<td>-30</td>
<td>-70</td>
</tr>
<tr>
<td>2</td>
<td>100</td>
<td>0.059</td>
<td>0.017</td>
<td>-60</td>
<td>-42</td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>100</td>
<td>0.059</td>
<td>0.054</td>
<td>-50</td>
<td>-32</td>
<td>-64</td>
</tr>
</tbody>
</table>

In this table, $R_e$ is equilibrium radius of the bubble. $f$ is the excitation frequency, $f_r$ is the linear resonance frequency of the bubble, $A$ is the amplitude of the ultrasound, $U_1$ is the magnitude of the fundamental component of the bubble's radial velocity response in dB re. 1 m-s$^{-1}$, and $U_2$ and $U_3$ refer to the second and third harmonic components.

In addition, Table 3.3 presents some later results for an ultrasonic frequency of 100 kHz and bubbles of radius 2, 3 and 5 $\mu$m. These values were chosen to represent the practical lower limits of frequency and bubble size for medical ultrasonic bubble detection. The results showed greater distortion than for the case where the driving frequency was much greater than the resonance frequency. To obtain acceptable accuracy in these solutions, the step size used had to be reduced greatly in comparison with the step size for the cases of resonance and higher frequencies. For the case in which the
equilibrium radius of the bubble was 2 \textmu m, and the pulse amplitude was 0.054 bars, each cycle of the driving frequency was divided into 4096 steps, and a total of 131,072 points were calculated. The maximum value of the local discretization error [Conte and de Boor, p. 352], in any of the steps, was 0.78 per cent.

The physical reason that, in the low frequency case, the response of the bubble is more heavily distorted than in the high frequency case can be simply understood. For lower frequencies, although the radial velocities involved are smaller, the bubble undergoes a greater relative change in size. In the limit of very low frequencies, the bubble would either tend to grow indefinitely, or to collapse, were it not stabilized by one or more other mechanisms. These mechanisms are considered in bubble nucleation theory [eg. Yount, Kunkle, Darrigo, Ingle, Yeung, and Beckman, 1977].

4. Variation of Bubble Size for Fixed Frequency

Most ultrasonic instruments operate at a single frequency owing to bandwidth limitations of the transducers. Also, to avoid possible harmful biological effects, medical diagnostic instruments are constrained to use low ultrasonic amplitudes. Therefore, in a more detailed numerical study, frequency and amplitude were kept fixed while bubble size was varied, to determine what population of bubbles would contribute significantly to nonlinear scattering of the ultrasonic pulse.

---

**a. Frequency 1 MHz**

The frequency 1 MHz is at the low end of the range of frequencies normally used in medical diagnostic ultrasonics (1 to 10 MHz). A pulse width of 16 \textmu s was chosen, with a cosine taper of 40 %, to give a pulse shape similar to one which would be easily attainable in practice. This pulse and its periodogram were shown in Figure 3.1. The amplitude of the pulse was 0.017 bars, which, for plane wave excitation, corresponds to a peak ultrasonic intensity of 10 mW-cm$^{-2}$. Bubble size was varied between 1 and 20 \textmu m, and the fundamental, second harmonic, and third harmonic components of the radial velocity response were noted. The values of $\kappa$ and $\xi$ used were obtained from Prosperetti [1977]. The analysis was repeated for a pulse amplitude of 0.054 bars, corresponding to a peak intensity of 100 mW-cm$^{-2}$. Figure 3.6(a) shows the fundamental, second and third harmonic components as obtained from the bubble velocity periodograms. Figure 3.6(b) shows the same plot for amplitude 0.054 bars. Plots of the ratio of the second and third harmonics to the fundamental for both amplitudes are shown in Figures 3.6(c) and 3.6(d). In these figures, the resonance size is 3.2 \textmu m, and the second harmonic exhibits a second peak at half resonance size. The ratio of second harmonic to fundamental components is greatest for bubbles of radius 1.6 \textmu m, and is significant only for bubbles in the range 1 to 4 \textmu m. Supplementary plots showing the damping constant, polytropic constant, and the resonance frequency vs. bubble size are given in Figure 3.7. The increase in the damping constant for small bubbles is due to the increasing importance of viscosity. For the smallest bubbles, the polytropic exponent is almost equal to one, corresponding to
Figure 3.6 (a) First, second, and third harmonic levels of bubble radial velocity, as a function of bubble size for pulsed ultrasound, of frequency 1 MHz, pulse width 16 μs, and amplitude 0.017 bars.
(b) As in (a), but for pulse amplitude 0.054 bars.
(c) The level of the second harmonic component, relative to the fundamental, for amplitudes 0.017 and 0.054 bars.
(d) The level of the third harmonic component relative to the fundamental, for amplitudes 0.017 and 0.054 bars.

Figure 3.7 (a) The total damping constant used in the 1 MHz simulation, as a function of the radius of the bubble at rest.
(b) The polytropic exponent as a function of the radius of the bubble at rest.
(c) The linear resonance frequency as a function of the radius of the bubble at rest.
isothermal oscillations, while for the larger bubbles, it increases towards, but does not reach 1.4, the adiabatic limit for diatomic gases. Figure 3.7(c) is a plot of Eq. (51). The dependence of the bubble resonance frequency on driving frequency is implied by the dependence of the polytropic exponent on frequency.

b. Frequency 500 kHz  Numerical results were also obtained for an ultrasonic frequency of 500 kHz. The width of the pulse was doubled, but the shape remained the same. Again the two pulse amplitudes, 0.017 and 0.054 bars were used. These results are presented in Figures 3.8 and 3.9.

The plots at 500 kHz are similar to those at 1 MHz, except shifted towards larger bubble radii. In addition, for the third harmonic component, a third peak has become visible. For this frequency, the scattering is significantly nonlinear for bubbles up to 8 \( \mu \text{m} \) in radius. Plots of the damping constant, polytropic exponent, and resonance frequency vs. bubble size are given in Figure 3.9.

Figures 3.10 to 3.13 present plots of the radial velocity in the time and frequency domains for four bubble sizes of interest. Figure 3.10 shows the response for a bubble of radius 2.2 \( \mu \text{m} \). The waveform appears to be relatively undistorted, even though this represents a maximum for the ratio of third harmonic to fundamental. In contrast, the response for a bubble of radius 3.2 \( \mu \text{m} \), shown in Figure 3.11, appears flattened. This represents a maximum in the ratio of second harmonic to fundamental scattering. The response for a bubble near
resonance is shown in Figure 3.12. The high $Q$, associated with resonance, is evident in the long time taken for the response to grow and decay. As well, all harmonic components exhibit an absolute maximum at resonance. Finally, Figure 3.13 shows the response of a bubble of radius 7.5 μm – larger than resonance size. The periodogram for this case shows an interesting asymmetry towards lower frequencies, indicating that the natural frequency of the bubble is less than the driving frequency. Although the reverse was true for bubbles smaller than resonance size, the effect was less noticeable.

5. Comparison with Experiments

In the next chapter, numerical results are presented along with experimental results with corresponding values for the frequency, pulse amplitude, pulse width, and bubble size. The frequencies used were 70.5, 245, 506, and 1,690 kHz, and pulse amplitudes were in the range 0.05 to 1.0 bars. The bubble radii observed ranged from 55 to 190 μm.

Also in the next chapter, some numerical results are presented to complement numerical observations of a freely oscillating gas bubble. To obtain these results, the bubble was assumed to remain spherical and an initial departure from equilibrium was specified.

Figure 3.9 (a) The total damping constant used in the 500 kHz simulation, as a function of the radius of the bubble at rest. (b) The polytropic exponent as a function of the radius of the bubble at rest. (c) The linear resonance frequency as a function of the radius of the bubble at rest.
Figure 3.10 (a) The radial velocity response of a bubble of radius $2.2 \mu m$ to a pulse of ultrasound of frequency 500 kHz, width 32 $\mu s$, and amplitude 0.054 bars.

(b) The periodogram of the response in (a).

Figure 3.11 (a) The radial velocity response of a bubble of radius $3.2 \mu m$ to a pulse of ultrasound of frequency 500 kHz, width 32 $\mu s$, and amplitude 0.054 bars.

(b) The periodogram of the response in (a).
Figure 3.12 (a) The radial velocity response of a bubble of radius 6.5 μm to a pulse of ultrasound of frequency 500 kHz, width 32 μs, and amplitude 0.054 bars. (b) The periodogram of the response in (a).

Figure 3.13 (a) The radial velocity response of a bubble of radius 7.5 μm to a pulse of ultrasound of frequency 500 kHz, width 32 μs, and amplitude 0.054 bars. (b) The periodogram of the response in (a).
6. Comparison with Results Available in the Literature

To confirm the validity of the numerical results obtained here, a comparison was carried out with some of the results published by Lauterborn [1976], Nishi [1975], and Zabolotskaya and Soluyan [1971].

a. Comparison with Lauterborn

As a check, a comparison was made with the published numerical results of Lauterborn [1976] for the two cases, from among his many solutions, for which he presented radius-time curves and power spectra. Both cases were for a bubble of equilibrium radius 10 μm, excited by a continuous sinusoidal pressure of amplitude 0.7 bars. In one case, the driving frequency was 646.46 kHz, being 1.95 times the resonance frequency, and in the other case, the frequency was 122.66 kHz, or 0.37 times the resonance frequency.

In both cases, the effective polytropic constant, $\kappa$, was taken to be 1.33, and only the viscous contribution to damping was considered. With respect to these choices, Lauterborn probably did not have access to Prosperetti's [1977] results when his calculations were performed, although these authors evidently did correspond on related matters. For the polytropic exponent, Prosperetti's results indicate values of $\kappa = 1.17$ at 646.46 kHz and 1.03 at 122.66 kHz. Lauterborn pointed out that, although neglected in his solution, thermal and acoustic damping would be important for bubbles of radius 10 μm. (Lauterborn's calculations were for bubbles of radius 0.1, 1, and 10 μm. For the smallest of those sizes, viscous damping is dominant.)

For the case, $f/f_r = 1.95$, Lauterborn showed one period of a subharmonic oscillation, which was termed a steady-state solution. Time zero on his graph cannot correspond to the beginning of the excitation, so it is not clear when the result shown was obtained with respect to the beginning of the excitation. A similar subharmonic response was obtained in this study, using what are believed to be the same initial conditions. The first 100 μs of the solution is shown in Figure 3.14. The subharmonic appears to grow for about the first 40 μs, and, afterwards, its amplitude is still modulated by a lower frequency component. The agreement with Lauterborn's result for this case is good. The periodogram also bears a strong resemblance to his.

For the sake of interest, these calculations were repeated using for the value of the polytropic exponent, $\kappa = 1.17$, and for the dimensionless thermal damping constant of Prosperetti, $B = 0.088$, which were obtained from Prosperetti's [1977] results. The values of the damping constants were 0.027, 0.021, and 0.010, for the acoustic, thermal and viscous contributions, respectively. The total damping constant was 0.058. Thus, as Lauterborn expected, the neglect of acoustic and thermal damping was not a good approximation for the 10 μm radius bubble. In this case (Figures 3.15(a) and 3.15(b)), although a subharmonic is originally present, it decays, leaving a solution which is, however, rich in harmonics. For interest, the radial velocity response is also shown (Figures 3.15(c) and 3.15(d)).

In the other case shown by Lauterborn, he obtained an ultraharmonic response. Although the same initial conditions were specified...
The first 100 μs of radial bubble oscillations for continuous excitation. The parameters were: frequency 646.46 kHz, amplitude 0.7 bars, $R_0$ 10 μm, $k$ 1.17, including thermal viscous damping, and acoustic damping.

(a) The relative bubble radius.
(b) The periodogram of the response in (a).
(c) The bubble radial velocity.
(d) The periodogram of the response in (c).
in this study, a similar ultraharmonic was present only initially, decaying eventually to a steady state response with most of the energy at the driving frequency (Fig. 3.16). This response had a very high harmonic content. The discrepancy with Lauterborn's result is likely due to a difference in the form of the viscous damping constant. This difference becomes important for large amplitude oscillations (Appendix B).

The solutions shown in Figure 3.17 were obtained using values of 1.03 and 0.051 for $K$ and $B$, respectively. In this simulation, the shape of the driving pulse was given a 10% cosine taper, which allowed the response to settle to a steady state value more quickly. Its eventual form did not differ noticeably from the response obtained without taper, despite the nonlinear nature of the problem. This solution differs greatly from the one shown in Figure 3.16. For comparison, the radial velocity response is also shown. In the radial velocity response, the third and fourth harmonic components both exceed the fundamental.

b. Comparison with Nishi. As a check of the accuracy of the algorithm, a comparison with the linearized results of Nishi [1975] was also carried out. Nishi published plots of the ratio of scat-
ttering intensity to incident intensity at 1 cm, vs. bubble radius, for excitation by a continuous plane wave at 5 MHz, with hydrostatic pressure as a parameter. For comparison, numerical calculations were made for bubbles of radius 1 and 10 μm, excited at 5 MHz by a pulse, tapered 40% at both ends, with amplitude 0.1 bars, and width 32 μs.

Figure 3.16 The first 120 μs of radial displacement for continuous excitation, obtained for comparison with the results of Lauterborn [1976]. The parameters were: frequency 122.66 kHz, amplitude 0.7 bars, $R_b$ 10 μm, $\kappa$ 1.33, with viscous damping only.
(a) The relative bubble radius.
(b) The periodogram of the response in (a).
The first 120 μs of radial bubble oscillations for cosine-tapered, continuous excitation. The parameters were: frequency 122.66 kHz, amplitude 0.7 bars, R₀ 10 μm, k = 1.03, including thermal viscous damping, and acoustic damping.

(a) The relative bubble radius.
(b) The periodogram of the response in (a).
(c) The bubble radial velocity.
(d) The periodogram of the response in (c).

For the larger bubble, the ratio of excitation to resonance frequency was 14.97, the velocity amplitude was about 0.03 m/s, and the solution possessed no noticeable harmonic content. For the smaller bubble, the ratio of excitation to resonance frequency was 1.28, the radial velocity was approximately 0.75 m/s, and the solution was noticeably nonlinear. The ratio of the second harmonic to fundamental component of radial velocity was -31 dB.

The scattering ratio at 1 cm was calculated using

\[ \frac{I_s}{I_i} = (\rho_\omega R_0^2 U/r_p)^2 \]  

based on a well known result for the intensity of radiation from small spherical sources and in plane waves [Kinsler and Frey, pp. 163 and 118]. For the 10 μm bubble this ratio was 8.85 × 10⁻⁷, and for the 1 μm bubble it was 5.53 × 10⁻⁸, in excellent agreement with Nishi's plotted results.

### Comparison with Zabolotskaya and Soluyan

As a check of the validity of the expression given by Zabolotskaya and Soluyan [1971] (Eq. 29), this expression was evaluated and compared with some numerical results (Table 3.4). To evaluate Eq. 29, the value used for the damping constant was the same as that calculated in the numerical simulation. The agreement was good for most cases, including even that for the smallest bubble size tested at 1 MHz. Since the expression of Zabolotskaya and Soluyan does not include the effects of surface tension, it was not expected to be accurate for bubbles of radius much less than 50 μm. The largest discrepancy was observed for bub-
bles larger than resonance size. Also, when the amplitude of the
driving pressure was increased from 0.017 to 0.054 bars, at 1 MHz, the
discrepancy became larger, as would be expected, since the expression
of Zabolotskaya and Soluyan assumes small amplitude oscillations.

<table>
<thead>
<tr>
<th>Table 3.4 Comparison with Zabolotskaya and Soluyan [1971]</th>
</tr>
</thead>
<tbody>
<tr>
<td>f (MHz)</td>
</tr>
<tr>
<td>---------</td>
</tr>
<tr>
<td>1.00</td>
</tr>
<tr>
<td>1.00</td>
</tr>
<tr>
<td>1.00</td>
</tr>
<tr>
<td>1.00</td>
</tr>
<tr>
<td>1.00</td>
</tr>
<tr>
<td>1.00</td>
</tr>
<tr>
<td>0.50</td>
</tr>
<tr>
<td>0.50</td>
</tr>
<tr>
<td>0.50</td>
</tr>
<tr>
<td>0.50</td>
</tr>
<tr>
<td>0.055</td>
</tr>
<tr>
<td>0.0625</td>
</tr>
<tr>
<td>0.065</td>
</tr>
<tr>
<td>0.075</td>
</tr>
<tr>
<td>0.125</td>
</tr>
</tbody>
</table>

In this table, \( V_{2zs} \) represents the second harmonic component of the
bubble volume oscillations [1971], obtained using the expression of
Zabolotskaya and Soluyan, and \( V_{2N} \) represents the corresponding numerical
result, obtained in this study.

IV. EXPERIMENTAL INVESTIGATIONS

A. Approach

In most previous experimental investigations of nonlinear bubble
dynamics, high intensity ultrasound was used to create cavitation bub-
bles, and the bubble oscillations were then observed either optically or acoustically. The optical techniques were used to determine radius-
time curves, while the acoustical methods were used to obtain spectral information [Esche 1952, Akulichev 1971]. Neppiras [1969] studied nonlinear, especially subharmonic, oscillations of single or small numbers of gas bubbles, which were produced by forcing gas through a needle. Studies have also been carried out by Evans [1975], Martin et al. [1973], and Moulinier [1978], using bubbles produced by decompressing water or animal tissue, for the purpose of determining the effectiveness of detecting the second harmonic of scattered ultrasound to indicate the presence of decompression bubbles. Evans [1975] regarded the results of his experiments as inconclusive but promising. Martin et al. [1973] were optimistic about their results, while Moulinier [1978] concluded that the technique required further test and development. The results of these authors are discussed in Chapter V.

The experiments to be reported here differ from previous investiga-
tions in several ways. First, bubbles were formed singly, or in a
well-defined stream, as was the case in the experiments of Neppiras
[1969], but not in either the cavitation or decompression experiments.
This permits measurement of the bubble size, which is important for comparison with theoretical results, and it eliminates the complicating effects caused by the presence of a field of bubbles of various sizes. Second, the ultrasonic amplitudes employed were lower than those required to produce either cavitation, or a subharmonic response. This was also the case in the decompression experiments, of Evans [1975], Martin et al. [1973], and Moulinier [1978], for which the goal was to develop a diagnostic technique that would not damage tissue. The decompression experimenters did not, however, examine in detail the nonlinearity of the bubble response. They limited their studies to monitoring either the level of the second harmonic, or the ratio of the second harmonic to the fundamental for the purpose of bubble detection. In this study, a more fundamental approach has been adopted, and attention was not restricted to the second harmonic. Finally, in this thesis pulsed ultrasound was used, and, unlike previous studies, spectral analysis was done on single pulses of scattered ultrasound.

The numerical investigations indicate that the bubbles which will behave in a significantly nonlinear manner are of a size such that their resonance frequency is close to the excitation frequency, or to a lesser extent, a harmonic or subharmonic of the excitation frequency. Changing the ultrasonic frequency is inconvenient, however, because piezoelectric transducers are most sensitive to one frequency only [Heuter and Bolt, p. 86]. In the case of edge-clamped discs, this frequency corresponds to the thickness resonance. Away from this frequency the transducer response is still not flat, because of resonances associated with other dimensions, such as the radial resonance in circular discs. It is difficult, therefore, to match the frequency of the ultrasound to the bubble resonance frequency.

Large disc transducers can produce well-defined beams of ultrasound. Despite the directional nature of the transducers, it was observed, in preliminary CW backscattering experiments, that reflections from surfaces of the tank mask the contribution from the bubbles when only one or a few bubbles contribute to the scattering. Furthermore, when it is possible to detect bubbles, the bubble signal appears as an amplitude modulation of the background signal. This poses two problems: first, the presence of a large background signal makes it more difficult to detect the bubbles; and second, to analyse the spectrum of the ultrasound scattered by the bubbles, it is necessary to allow for the large fundamental component due to the reflections.

This masking can be reduced by a variety of techniques. One method is to use a very large number of bubbles distributed throughout the tank, effectively increasing the portion of the signal due to scattering by bubbles. It is difficult, however, to control the size and spatial distribution of such bubbles, and their interaction poses a complicated theoretical problem [Welsby and Safar, 1969].

A second method is to line the enclosure with sound absorbing material. Initially, tests were made with several materials available in the laboratory, including foam and neoprene rubber, but the reduction observed was inadequate. Special purpose materials for acoustic
lining of water-filled tanks are available commercially, which reduce reflected energy by 30 dB at frequencies above 1 MHz, but by less than 10 dB at frequencies below 100 kHz [Barge 1978, Corsaro, Klunder, and Jarzynski 1980]. Because much greater than 10 dB reduction of reflected energy was required to unmask the bubble signal at the lowest frequencies (70.5 kHz), and because of the high cost of the special rubber materials, another solution was sought.

A third method of reducing the masking of the bubble signal employs the Doppler shift caused by the movement of the bubbles through the field of view of the transducers. If the amount of the shift is great enough, the shifted component can be separated from the static, unshifted, background signal. The two components have significant bandwidths, however, and the Doppler component is generally of much lower magnitude, so that it may be buried in the skirt of the background signal, if the shift is small. The magnitude and bandwidth of the signals, and the properties of the filters used (the steepness of their cutoff and the amount of their out-of-band rejection) determine the amount of the Doppler shift needed for separation. The Doppler shift is proportional to the transmitted frequency and to the component of the particle velocity projected in the direction of the transducers. It was found experimentally that, for ultrasound in the MHz region, this frequency shift was sufficient to detect the passage of single bubbles of radius greater than 50 μm, since larger bubbles rise more rapidly. (The speed of their rise is proportional to the square of their radius, as derived by Stokes [see Prandtl and Tietjens 1937, or Lamb 1932].) To approach resonance conditions it was necessary to use frequencies below 100 kHz, where the Doppler shift, amounting to less than 1 Hz for bubbles of 50 μm radius, was insufficient.

The problem of avoiding the interference caused by reflections from tank walls and the liquid - air interface was largely overcome by using pulsed ultrasound and a gated receiver, the method generally used to simulate free field conditions in water-filled enclosures [Bobber 1970]. As shown in Figure 4.1 (b), the signal is sampled only during the interval between the end of the pulse transmission and the first return of echoes from surfaces in the enclosure other than the bubble targets. The next pulse is not transmitted until the echoes have sufficiently decayed. The use of this technique eliminated noise from electromagnetic coupling, which occurred during pulse transmission, and solved the problem of excessive acoustic background signal, except at the lowest frequency used, 70.5 kHz. At that frequency, some background signal had to be tolerated because, for the tank used, the pulse widths were too long to prevent reflections from arriving before the end of the pulse. The minimum pulse width achievable was dependent on the response of the transducers (Section IV. B. 4.). The use of pulsed ultrasound conferred a second benefit, in that, because the pulse consisted of a gated sine wave, the transmitted energy was spread over a wider range about the centre frequency, increasing the range of bubble sizes for which significant nonlinear behaviour could be expected. On the other hand, pulse operation posed the problem of devising a method to acquire and analyse the scattered pressure pulses. This challenge was met by building an apparatus capable of
capturing and digitizing a single ultrasonic pulse for later reproduction and analysis using a computer.

In any experiment in which nonlinear effects are to be observed, adequate account must be taken of the nonlinearity which is bound to be present in the apparatus itself, and which imposes a limitation on the ability of the apparatus to distinguish nonlinear effects. For this reason, it was found inadvisable to use a transducer tuned to the second harmonic of the transmitted ultrasound as the receiver, although this was done by Evans [1975], and Moulinier [1978]. It was observed that, with such an arrangement, the receiving transducer may behave nonlinearly. An increase in the amplitude of the fundamental (for example, due to scattering by a passing bubble) could then lead to an increase in the observed second harmonic level with respect to the fundamental. This was verified using small steel balls as targets, since these represent linear scatterers. Furthermore, it was observed using a spectrum analyser, that the second harmonic level could change spontaneously, and was dependent on the orientation of the receiving transducer with respect to the transmitter. To avoid this problem and to achieve sufficient sensitivity for bubble detection, the receiving transducer was usually identical to the transmitter, and operated at resonance.

As stated previously, the aim of these experiments was to search for nonlinear effects in the scattering of ultrasound by bubbles, for conditions which might apply to detection of bubbles in blood and tissue, to determine whether the nonlinearity would be an effective indicator of the presence of bubbles. The appearance of nonlinearity would be indicated by an increase in the level of the second and third harmonic components relative to the fundamental, over the levels observed from scattering by other, presumably linear, scatterers. (In the latter case, the residual second and third harmonic components would represent the system noise.) Thus, average periodograms obtained from scattering by bubbles were compared visually with average periodograms obtained using a reference scatterer, usually an aluminum rod. (The value of $kR$ for the rod, where $R$ is the radius of the rod, varied from 1.5 at 70.5 kHz to 36 at 1.69 MHz. For that range, the reflection factor is almost constant at about 0.7 [Skudrzyk, p. 448]; at lower frequencies it drops off.) A rough comparison with theory was effected by obtaining numerical solutions to the model discussed in Chapter III for the bubble radii and approximate ultrasonic amplitudes used in the experiments, and from those solutions, determining the predicted levels of the second and third harmonics of the radiated pressure, relative to the fundamental.

This comparison was complicated by a number of experimental factors. In the first place, there were usually several bubbles in the ultrasonic beam. The bubbles were, however, of approximately equal size, and were usually well-separated. This affected the amplitude, but not the harmonic content of the received signal, and it was the latter property which was of interest. In fact, the larger amplitude improved the signal-to-noise ratio of the captured pulse. Secondly, the amplitude distribution across the ultrasonic beam was non-uniform, especially in the case of the highest frequency transducers (1.69
MHz), for which the bubbles were in the near-field. If a bubble were in a position of reduced amplitude, its response would be more linear. During preparations for the experiments, care was taken to aim the experiments for maximum scattered signal. Finally, the frequency response of the receiving transducers was not flat. The lack of transducers with flat frequency response, as well as adequate sensitivity and directionality, was one of the two most important experimental limitations. The other was the inability to produce bubbles as small as desired.

Details of the experimental apparatus are given in the next section.

B. Apparatus

In outline, the experimental apparatus consisted of a glass water-filled tank to contain the experiment, a bubble generator, a microscope and camera for viewing and recording the bubbles, and transducers for transmitting and receiving the ultrasound (Fig. 4.1). The transmitter included a CW signal source, a radio frequency (RF) switch gated by a pulse generator, and a 50 dB power amplifier. The receiver included a broadband amplifier, and an instrument for sampling and digitizing the analogue signal. A delayed replica of the pulse used to gate the RF switch triggered the receiver. Signal analysis was done using a minicomputer. The experimental procedure consisted of: generation of a stream of bubbles; visual observation of the bubbles and recording them photographically for subsequent sizing; transmission of a pulse of ultrasound; capture of the pulse scattered by the bubbles; and, finally, spectral analysis of the captured pulse.

1. Bubble Generator

To generate bubbles, nitrogen was forced under pressure through a stainless steel needle or a glass micropipette. The needle was 5 cm long with a nominal bore of 25 μm and an outer diameter of 90 μm. The pipettes were formed using a pipette puller. As the micropipette orifice produced by this method, was too small to permit bubble formation, the tips were carefully broken while viewing them under a microscope. An outer diameter between 10 and 20 μm was optimum for production of uniform, small bubbles. A precision pressure regulator
Figure 4.1 A schematic of the experimental apparatus. Bubbles were formed by forcing nitrogen through a needle. The bubbles were observed with a microscope and photographed for sizing. A pulse of ultrasound was transmitted toward the bubbles. The pulse scattered by the bubbles was received, captured, digitized, and sent to a minicomputer for plotting and spectral analysis.

Figure 4.1 (b) The effect of range gating as viewed with an oscilloscope.
(1) The pulse applied to the transmitting transducer.
(2) The receiver trigger pulse, with variable delay, and the receiver window.
(3) The received signal.
   (i) The direct r.f. leakage
   (ii) The pulse scattered by the target (bubbles). The delay of the receiver gate is adjusted to correspond to the time of arrival of this pulse.
   (iii) Echoes from other reflectors. These are allowed to decay before transmission of the next pulse.

provided the operating pressure (less than 1 bar) that was required to drive gas through the needle or pipette orifice.

The needle was immersed in the water and the bubbles rose to the surface through the overlapping fields of view of the transmitting and receiving transducers, which were positioned adjacent to each other at a shallow angle (backscattering), and aimed for the maximum bubble signal. A surgical microscope (magnification 500 times) with camera, was trained on the tip of the needle. Photographs of the bubbles were taken as they rose away from the needle, appearing as dark spots against a bright background (Fig. 4.2). The contrast was accentuated by taping shiny foil to the opposite side of the tank. Black and white film was used (Kodak Tri X). The best exposures were obtained by using the maximum lens aperture (f 14) at top shutter speed (1/500 s), and by pushing the developing process three stops (in Dufine) for an effective film speed of ASA 3200. Measurements of bubble radius could then be made by comparing the size of the image of the bubble with that of the needle, which had been measured independently under another microscope having a calibrated graticule.

The accuracy of this method of measurement was about ± 12 %, being limited by the sharpness of the photographic image. The standard deviation of a typical group of measurements on twenty bubbles, was usually a few per cent of the mean. Measurements which were made on bubbles over a period of hours usually agreed to within a few per cent, attesting to good reproducibility of the bubble production method.
Figure 4.2 A stream of bubbles produced by the bubble generator, of radii $74 \pm 11 \, \mu m$. The outer diameter of the stainless steel needle is $94 \pm 4 \, \mu m$. 
Bubble size and rate of production could be controlled over a limited range by varying the regulator pressure. The smallest bubbles were obtained for pressures just above the threshold level of formation at the end of the needle. Rate of production was faster for small bubbles than for larger ones. Depending on the rate of production, and the size of the transducers, several bubbles were usually in the ultrasonic beam at one time. The bubbles ranged in radius from 55 to 350 μm, but the smallest sizes seem to have been achieved with the fortuitous assistance of some contamination at the tip of the needle. These bubble sizes are similar to those achieved by Neppiras [1969] who used a similar technique, but included a mechanical vibrator to dislodge the bubbles from the needle. In this application, a vibration would have resulted in blurring of the photographs of the bubbles. Another technique for producing bubbles which has been used by several other workers is electrolysis [Daniels et al. 1980, Monjaret et al. 1975]. Both techniques produce bubbles of comparable size; the limiting factor appears to be the surface tension of the needle or electrode. As the bubble grows, it remains attached to the needle, until buoyancy overcomes the surface tension.

The experiment was contained in a glass tank, dimensions: 7.5 cm long, by 45 cm high, by 30 cm wide. The tank was filled with tap water which was allowed to stand for a few days until bubbles clinging to the surfaces disappeared. Figure 4.3 is a photograph of the tank, bubble generator, microscope, and transducers.
Figure 4.3 Part of the experimental equipment, showing the tank, bubble generator, microscope, and transducers.
2. Transmitter and Receiver

The components comprising the RF section of the apparatus are shown schematically in Figure 4.1 and listed in Appendix D, which details manufacturers and model numbers. For the signal source, a frequency synthesizer was employed, which produced a stable, sinusoidal waveform of low distortion, (all harmonics less than -40 dB with respect to the fundamental component), with precise frequency control and adjustable output level. This signal was passed to the input of an RF switch, which was gated on and off by a function generator. In the off state, the signal was attenuated by 90 dB. The function generator produced a -1.5 V level to turn the switch off and a +1.5 V variable width pulse to turn the switch on. The pulse repetition rate produced by the function generator could be set by the internal oscillator, but, in that case, the pulse duration was a function of repetition rate. To achieve low enough rates, while maintaining narrow pulses, it was found best to trigger the function generator by another pulse generator at the repetition rate desired, or for single shots, to trigger it manually. A variable coaxial attenuator and an RF power amplifier followed the RF switch. The power amplifier provided 50 dB of gain, (maximum power: 100 W continuous, bandwidth: 20kHz to 10 MHz). The attenuator provided gain control and attenuated the relatively high level signal from the RF switch (0.2 V RMS), which was necessary to minimize the effect of switching transients. The output from the power amplifier was connected to the projector transducer and was monitored with an oscilloscope.

Signals scattered back from a bubble, or bubbles, were detected by the receiving transducer or by a hydrophone. The signal from the receiving transducer was fed to an amplifier. The particular unit depended on the frequency and type of experiment. For the 70.5 kHz experiments, the amplifier was a video differential type, with a 300 kHz bandwidth, maximum gain of 80 dB, and adjustable low and high cutoff frequencies. For 245 kHz and higher frequencies, the amplifier consisted of one or two 30 dB RF stages, with nominal range extending from 500 kHz to 30 MHz. In fact, tests showed that the lower frequency limit for flat response was close to 200 kHz. In the first case, frequency information above 300 kHz, and in the second case, below 200 kHz, was lost. With the hydrophone, a special 200 kHz video preamplifier was used. An attenuator was included after the amplifier(s), to adjust the signal to within ± 0.5 V prior to analogue-to-digital conversion.

3. Pulse Capture System

Spectrum analysis of transient signals with frequency components greater than 100 kHz is difficult because the conventional instruments covering this frequency region, swept-tuned analysers, require repetitive signals and are therefore not applicable. Digital spectrum analysers, which perform Fourier transforms on captured signals, are suitable for spectral analysis of transients, but most are limited to frequencies below 100 kHz. For higher frequencies, spectrum analysis of single events can be accomplished by acquiring the signal with a
transient waveform digitizer and sending the data to a computer. A system was developed [Eatock, Allin, and Ferrari, 1981] to capture scattered ultrasonic pulses and transmit the data to the minicomputer for spectral analysis. In the event that the minicomputer was not available, the data could be stored on magnetic tape cartridges included with a video terminal.

a. Description of the system Figure 4.4 shows a schematic of the pulse digitizer, and Figure 4.5 shows most of the system components. Essentially, the system consisted of a fast analogue-to-digital (A/D) converter; a logic analyser, used as a high speed memory for the data from the A/D converter; and a microprocessor, to interrogate the logic analyser memory and pass the information to a video terminal and thence to a minicomputer. The modular 8-bit A/D converter had a maximum conversion rate of 11 MHz, which was determined by the frequency of an external clock. The clock consisted of a frequency synthesizer which triggered a pulse generator (Fig. 4.6). The synthesizer provided a stable square wave with a rise time of approximately 10 ns. Its frequency was adjustable over the range of operation of the A/D converter with 0.001 Hz resolution. The pulse generator converted the square wave into a pulse train, with a pulse rise time of 10 ns and a pulse width of 35 ns, appropriate for triggering the A/D converter. The clock could be operated using the pulse generator's own oscillator, but setting the clock frequency was then much less convenient, and a counter was required to monitor the clock frequency. A coaxial switch was provided to turn off the clock to the A/D converter. This was necessary during transfer of data from the
Fig. 4.5 Most of the components of the single-pulse digitizer. From left to right: two function generators, one to provide ECL levels for the logic analyser external clock, and one to gate an RF switch, shown resting on top, which provided the pulsed signal to drive the transmitting transducer; a black box containing A/D converter, microprocessor, and interface circuits; power supplies; logic analyser; and terminal. The frequency synthesizer and counter are not shown.
logic analyser memory to the terminal, otherwise noise from the free running A/D converter would cause the data to shift in memory and interfere with the transfer.

The role of the logic analyser was to capture the data from the A/D converter in its high speed memory. The particular analyser used (a Tektronix 7D01 plug-in module in a 7600 series oscilloscope mainframe) provided the necessary access to its memory through an internal connector. In the experiments, a signal obtained by delaying the gating pulse for the transmit switch (Fig. 4.1) was used to trigger the logic analyser to store 508 sequential 8-bit samples, either before, around, or following the trigger pulse. In practice, centre-triggering was most convenient. By setting the delay time equal to the return acoustic transit time to the target of interest, a pulse scattered by that target was captured. The delay was achieved by using the gating pulse to also trigger a pulse generator with an adjustable delay circuit, and then using the output of the pulse generator to trigger the logic analyser. This trigger signal was also routed to the least significant bit of the logic analyser input to provide a time reference for the data. As a consequence, the data had 7-bit resolution, which imposes a limit on the maximum signal to noise ratio achievable. For 7-bit resolution this limit would be approximately 40 dB [Oppenheim and Schafer, p. 413]. In practice this ratio was improved by averaging, and the limiting factor in resolving harmonics proved to be the sensitivity of the transducers, discussed in the next section. The logic analyser display afforded a convenient visual check that the contents of its memory were useful. For example, the

![Figure 4.6 Components comprising the clock for the A/D converter.](image-url)
The appearance of the trigger pulse at the centre of the screen indicated that the data were positioned in memory according to the correct time sequence.

The microprocessor used standard transistor-transistor logic (TTL) while, to achieve high speed, the logic analyser used emitter coupled logic (ECL). Hence it was necessary to provide logic translators at the interface between the two devices. The ECL to TTL translation, for data and status signals from the logic analyser, was accomplished using integrated circuit translators. For the control pulse from the microprocessor, used to step through the logic analyser memory, more current was required than could be obtained from integrated circuit TTL-to-ECL translators. The translation was accomplished by using the control pulse to trigger a function generator capable of driving low impedances, and the dc offset of the function generator was set to provide ECL levels. In operation, transmission of data to the terminal was done by flipping a switch added to the front of the logic analyser, which changed its mode to permit reading the memory, and by releasing a reset switch connected to the microprocessor to start the program running.

The microprocessor transmitted the data to a video terminal for temporary storage on a magnetic tape cartridge included with the terminal. The data were then transmitted to a minicomputer for plotting and spectral analysis.

b. Performance tests

As the purpose of the experiments was to search for harmonic distortion of ultrasonic fields caused by bubbles, it was considered most important that the digitizing system should introduce negligible harmonic distortion. Two performance tests were carried out with the instrumentation to verify that signal distortion would be at an acceptably low level for the experiments. In the first test, a portion of a sine wave was captured for several A/D conversion rates from 1 to 10 MHz with the frequency of the sine wave set to 0.01 times the conversion frequency. After transmitting the data to the minicomputer, the waveform was plotted and its periodogram was calculated and plotted.

Figure 4.7 shows the results for a 20 kHz sine wave sampled at a clock frequency of 2 MHz, and Figure 4.8 shows the result for a 100 kHz sine wave sampled at 10 MHz. At 2 MHz there is little distortion, whereas at 10 MHz a significant amount of distortion has become apparent, which, however, is not harmonically related to the signal. The second harmonic is still at least 23 dB below the fundamental, and this is not limited by the A/D conversion, but by the truncation of the waveform.

A second test, which consisted of sampling a 10 kHz ramp, was carried out to check the linearity of the A/D converter. Figure 4.9 shows the result for a 1 MHz conversion rate. Some clipping is apparent at the extreme values of the ramp, near the range limits of the A/D converter, but the slope appears to be constant.
Figure 4.7 (a) An example of a 20 kHz sine wave sampled at 2 MHz. (b) The periodogram of the sine wave, calculated using a 5% cosine taper.

Figure 4.8 (a) An example of a 100 kHz sine wave sampled at 10 MHz. (b) The periodogram of the sine wave, calculated using a 5% cosine taper.
4. Transducers

The transducers used in these experiments were piezoelectric ceramic discs mounted in brass or aluminum holders. In addition, two calibrated hydrophones were available, with bandwidths from 0.1 Hz to 70 and 200 kHz respectively.

The disc transducers were designed for CW operation, since CW experiments were originally planned. They were assembled in groups of four, two resonant at a fundamental frequency and two resonant at roughly twice that frequency. The precision of this matching was limited by the tolerance specified by the manufacturer for the thickness of the discs - roughly ten per cent. The piezoelectric material was lead zirconate titanate (Vernitron PZT 5A or equivalent). Since the thickness mode resonance was used, the discs were clamped on the edges and air backed. The lowest frequency transducers consisted of two discs bonded together with conductive epoxy, because sufficiently thick single discs could not be obtained. The discs were fitted into a nylon sleeve, which was in turn held by a brass or aluminum cylinder. Hollow cylindrical handles, 25 cm long, provided protective conduits for the leads. Point solder connections were made to the front and back of the discs. Figure 4.10 shows some of the transducers and Table 4.1 lists some of their specifications. The fourth column in the table refers to the theoretical location of the last maximum in the near field pressure distribution for a piston source. This marks the transition to the far field.
Although designed for CW operation, the transducers performed fairly well in pulse mode. Their air-backed design minimizes damping and maximizes sensitivity, which is best for CW operation, but it also increases the Q and the response time of the transducers, and thus limits the ability to achieve narrow pulses. This proved to be a problem only at 70.5 kHz, where pulse lengths shorter than 400 μs (28 cycles) would have been desirable because echoes from reflecting surfaces returned to the receiving transducer before the pulse transmission was completed. In Section IV. A., it was remarked that transducers tuned specifically to the second harmonic should not be used to detect the scattered ultrasound, because these transducers tend to behave nonlinearly. Therefore, the transducer which was the companion of the transmitter, was usually selected as the receiver. The fact that the receiving transducer did not have a flat frequency response gave rise to the problem of how to interpret the received spectrum. The following procedure was adopted. An aluminum rod was placed where the bubbles had been observed to rise, and pulses scattered by the rod were acquired. (The aluminum rod was assumed to represent a linear scatterer, and in fact, the spectra obtained using the rod as a reflector did not differ in shape from those obtained by transmitting

### TABLE 4.1 Transducer Specifications

<table>
<thead>
<tr>
<th>operating frequency (kHz)</th>
<th>thickness (cm)</th>
<th>diameter (cm)</th>
<th>position of last max (cm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>70.5</td>
<td>2.54</td>
<td>6.34</td>
<td>4.4</td>
</tr>
<tr>
<td>245</td>
<td>0.91</td>
<td>4.19</td>
<td>7.3</td>
</tr>
<tr>
<td>506</td>
<td>0.392</td>
<td>4.18</td>
<td>4.2</td>
</tr>
<tr>
<td>1690</td>
<td>0.127</td>
<td>2.5</td>
<td>18.4</td>
</tr>
</tbody>
</table>
Figure 4.10 Some of the transducers used in the experiments, from left to right: a disc resonant at 506 kHz (thickness mode), a 245 kHz disc, the 0 to 200 kHz miniature hydrophone (B&K 8103), a 1.69 MHz disc, and a 70.5 kHz disc.
a pulse of ultrasound directly between the two transducers.) Spectra acquired by scattering from bubbles were then compared visually with those obtained by scattering from the rod.

The 70.5 kHz experiments proved to be the most problematical. The width of the pulse applied to the transducer was 300 \( \mu \)s, but the signal transmitted was effectively 400 \( \mu \)s wide, and the rise and fall of the pulse was much more gradual. Figure 4.11 shows this signal as observed by a hydrophone, (bandwidth 200 kHz) placed about 7 cm in front of the transmitting transducer. The return transit time to this position was only 95 \( \mu \)s so that the scattered signal arrived before the pulse finished. Therefore, only the signal present in the first 100 \( \mu \)s immediately following the end of the transmitted pulse was captured. An example of a signal scattered by the aluminum rod is shown in Figure 4.12 along with the average spectrum obtained by averaging the periodograms from ten such records. The transmitted spectrum, as observed by the 200 kHz hydrophone, is shown in Figure 4.13. Beside the fundamental peak, another peak can be seen at 30 kHz, which corresponds to the radial resonance of the disc. There is also a second harmonic peak 28 dB below the fundamental.

The amplitude of the pulse, measured at the location of the bubbles, for a driving pulse of amplitude 32 V, was 0.05 bars. Estimates of the pressures produced by the higher frequency transducers, for which no direct method of measurement was available, were based on the relative efficiency of the transducer pairs, determined by transmitting a pulse of ultrasound through water, and comparing the output
Figure 4.11 An oscilloscope photograph showing, top, the RF pulse applied to the 70.5 kHz projector and, bottom, the shape of the transmitted pulse as observed by a hydrophone 7 cm in front of the projector.
Figure 4.12 (a) Part of a pulse, with 70.5 kHz centre frequency, scattered by an aluminum pole 7 cm in front of the projector.
(b) The spectrum obtained by averaging periodograms for ten such records.

Figure 4.13 The average of seven periodograms for 70.5 kHz pulses as observed with a hydrophone, bandwidth 200 kHz, placed 7 cm in front of the projector.
voltage amplitude with the input.

The sensitivity of the receiver transducer to the second and third harmonics was examined by using the hydrophone, for which a typical transmission response was available, as a projector. The hydrophone was driven with a signal at 70.5 kHz, having known amounts of second or third harmonic distortion. The distortion was generated by superimposing the signal from a second source on top of the 70.5 kHz signal from the first source. A coaxial attenuator was used to vary the distortion level between -50 and 0 dB with respect to the fundamental, and signal distortion was checked with a spectrum analyser. The distorted waveform was transmitted by the hydrophone to the receiving transducer. The detected signal was then digitized and passed to a computer where the periodogram was computed. The received second or third harmonic levels were compared with the transmitted levels, taking into account the transmission response of the projector. This experiment showed that the 70.5 kHz transducer, used as a receiver, reduced the second harmonic component with respect to the fundamental by 22 dB. It was also shown that the transmitted second harmonic component must be greater than -10 dB with respect to the fundamental to be detected with this transducer; otherwise it would be buried in the noise. Likewise, the third harmonic component of the signal was reduced by about 5 dB, and only third harmonic signals greater than -25 dB relative to the fundamental would be detectable.

Such tests were not possible at higher frequencies, because no calibrated projector was available. For the frequencies 245 kHz, 506 kHz, and 1.69 MHz, the pulses scattered by the aluminum pole, along with average periodograms for ten such pulses, are shown in Figures 4.14(a), 4.14(b), 4.15, and 4.16, respectively. For the 285 kHz transducers, spectra were also obtained for pulses of 3 and 6 dB higher amplitude (Figure 4.14(c) and 4.14(d)). (The receiver gain was reduced for the higher amplitude pulses to keep the signal within the range of the A/D converter.) The magnitude of the prominent third harmonic did not change relative to the fundamental, indicating linear behaviour for the transducers and the rest of the system.

In the results which follow, the average periodograms for scattering by bubbles should be compared with those in Figures 4.12, 4.14, 4.15 and 4.16. The calculated radial velocity periodograms should not be compared directly with the experimental results because they represent radial velocity, not pressure as measured by the transducers, and because they are not coloured by transducer response. The predicted levels of second and third harmonics of radiated pressure are given in separate tables.

C. Results

1. Ultrasonic Frequency 70.5 kHz

The lowest ultrasonic frequency practically attainable with the available transducers was 70.5 kHz. Operation at this frequency was problematical because long pulse widths (about 400 μs) were necessary, which reduced the effectiveness of range gating for avoidance of
Figure 4.14  
(a) A 245 kHz pulse scattered by an aluminum pole.
(b) The average periodogram for ten records.
(c) The average periodogram for four pulses transmitted with 3 dB greater amplitude. (The receiver gain was reduced 3 dB.)
(d) The average periodogram for four pulses transmitted with 6 dB greater amplitude than in (b). (The receiver gain was reduced 6 dB.)

Figure 4.15  
(a) A 506 kHz pulse scattered by an aluminum pole.
(b) The average periodogram for ten records.
interference from reflections (Sections IV. A. and IV. B. 4.). As a result, the bubble signal was superimposed on a background signal which had an amplitude that was about the same as that of the bubble signal. In interpreting the results, it was assumed that this background signal was the result of linear scattering. The effect would then be to increase artificially the level of the fundamental component with respect to the harmonics which might be generated by the bubbles.

The shape of the transmitted pulse has already been displayed in Figure 4.11(a). The first results were obtained for bubbles of average radius $148 \pm 16 \mu m$ (Section IV. B. 1.). Figure 4.17 shows a periodogram obtained by averaging the periodograms for 6 records of 70.5 kHz pulses scattered by these bubbles. This spectrum is not significantly different from that obtained with the aluminum rod, representing a linear scatter (Figure 4.12(b)). The peak at 240 kHz in Figure 4.17 frequently occurred, whether or not bubbles were present, indicating that it was spurious.

This experimental result was compared with a numerically derived result for a bubble of radius 150 $\mu m$, excited by a pulse of amplitude 0.07 bars. To simulate the response of the transducer, the excitation pulse was given a 100 $\%$ cosine taper. This pulse and its periodogram are shown in Figure 4.18. The time and frequency domain representations of the bubble's radial velocity response are shown in Figure 4.19. Table 4.2 summarizes the harmonic levels predicted for this and subsequent cases. As discussed in Section III. B., the radial
Figure 4.17 The average periodogram for six records of 70.5 kHz pulses scattered by bubbles 150 μm in radius.

Figure 4.18 (a) The excitation pulse used in the 70.5 kHz numerical simulation. A 100% cosine taper was applied, to simulate the response of the transducer. (b) The periodogram of the excitation pulse.
velocity was chosen for plotting because the pressure developed by a small spherical source is proportional to it. In this table, the harmonic levels from the bubble velocity periodogram were corrected to account for the dependence of pressure on frequency. As was noted in the previous section, the second harmonic level would have to be greater than about -10 dB, and the third harmonic level greater than about -25 dB, relative to the fundamental, to be detectable above the noise level. Therefore, the harmonics predicted here would not be observed. The table also includes the calculated resonance frequency, to show the separation between it and the driving frequency. The value of the polytropic gas constant, $\kappa$, and the damping constant, $\delta$, were obtained from Prosperetti's tables [1977(b)]. For all sizes studied, the viscous component of the damping was small. For bubbles of radius 70 and 55 \( \mu \text{m} \), the thermal component was slightly larger than the acoustic component, while for larger bubbles, the acoustic component dominated.

Experimental results were also acquired for bubbles of mean radius 174 ± 19, 99 ± 12, and 56 ± 8 \( \mu \text{m} \). The results for the 56 \( \mu \text{m} \) bubbles are presented in Figure 4.20, which shows average periodograms obtained for three different driving pulse amplitudes, corresponding to approximately 0.05, 0.07, and 0.1 bars, respectively. In Figure 4.20(a) and 4.20(b), no second or third harmonic response is noticeable. The trough in Figure 4.20(a) at 240 kHz and the peak in Figure 4.20(b) at the same frequency were spurious. It appears, however, that in Figure 4.20(c) a genuine third harmonic was beginning to emerge.
In this table, $f$ is the resonance frequency of the bubbles; $\kappa$ is the polytropic exponent of gas compression; $\delta_t$ is the total damping constant; $A$ is the pulse pressure amplitude; and 2nd and 3rd refer to harmonic pressure levels relative to the fundamental.

The corresponding numerical results are summarized in Table 4.2. In most cases, these levels would not be detected with the current apparatus. The numerical results for bubbles of radius 55 $\mu$m are shown in Figure 4.21. These results show pronounced nonlinear effects. For a pulse amplitude of 0.1 bars, the numerical simulation predicts second and third harmonic levels of -22 and -41 dB relative to the fundamental. The third harmonic component in Figure 4.20(c) was -32 dB with respect to the fundamental. The actual level would be approximately -22 dB, taking into account the 5 dB reduction in sensitivity of the transducer at the third harmonic, and the increase in
the fundamental because of the background signal. This is 19 dB greater than predicted numerically for bubbles of radius 55 μm. If, however, the bubbles were assumed to be of radius 48 μm (the measurement gave 56 ± 8 μm) then the predicted third harmonic level would be -24 dB, which would constitute reasonable agreement. In addition, the pressure amplitude used in the numerical solution could be different from that felt by the bubbles by as much as 10%, because this measurement was sensitive to transducer position with respect to the measuring hydrophone.

In an attempt to improve the sensitivity of the receiver at the second harmonic, numerous other transducers were tried, including the transducer designed for operation at 1.69 MHz, and the hydrophone with 200 kHz bandwidth. Unfortunately, the first of these also had a peak in sensitivity near 70 kHz, and a minimum near 140 kHz, so that the second harmonic was reduced by about 22 dB. Because of its smaller size, this transducer was much less directional, resulting in an increased background signal level. On the other hand, the hydrophone is 2 dB more sensitive at 140 kHz than at 70 kHz, according to the manufacturer's calibration, but because of its omnidirectionality, it was also more sensitive to the background signal. Experiments with these transducers involved a range of bubble sizes from 70 to 190 μm in radius, but no significant harmonics were observed in any case. Numerical results for four of the additional bubble sizes are included in Table 4.2.

Figure 4.21 (a) A numerical solution showing the radial velocity response for a bubble of radius 55 μm, to a pulse of frequency 70.5 kHz, width 400 μs, and amplitude 0.05 bars.
(b) The periodogram of the response shown in (a).
(c) As in (b), but for a pulse of amplitude 0.07 bars.
(d) As in (b), but for a pulse of amplitude 0.1 bars.
2. Ultrasonic Frequency 245 kHz

Using an ultrasonic frequency of 245 kHz, measurements were obtained for three different bubble radii: 120 ± 15, 95 ± 12, and 87 ± 11 μm, and, for each size, the pulse amplitude was increased twice by 3 dB. The shape of the transducer pulse was shown in Figure 4.14 (a). These results were compared with numerical calculations using pulse amplitudes estimated to be 0.15, 0.22, and 0.30 bars, employing the method discussed in Section IV. B. 4.

Figure 4.22 shows a single pulse scattered by bubbles of radius 87 μm, and the average of four periodograms for each of the three amplitudes. The experimental results show no significant differences between the periodogram of the pulse scattered by bubbles and that scattered by an aluminum rod. For comparison, the numerical results are presented in Figure 4.23. The shape of the excitation pulse used in the numerical simulation and its periodogram are similar to those for 70.5 kHz shown in Figure 4.18, being changed only in scale. The harmonic levels apparent in the numerical results are included in Table 4.3. Although strikingly evident in the numerical plots, these levels are too low to be detected experimentally. For this and subsequent cases, the theoretical damping constant is dominated by the acoustic component. Here it is two to three orders of magnitude greater than the thermal and viscous components.

Similar results were obtained for bubbles of radius 95 and 120 μm, except that as bubbles become larger, the amplitude of the radial...
velocity decreases, and harmonic levels decrease.

### Table 4.3 Numerical Simulation of Experiments (2)

<table>
<thead>
<tr>
<th>( f ) (kHz)</th>
<th>( R_b ) (( \mu )m)</th>
<th>( f_r ) (kHz)</th>
<th>( \kappa )</th>
<th>( \xi_r )</th>
<th>( A ) (bar)</th>
<th>2nd (dB)</th>
<th>3rd (dB)</th>
</tr>
</thead>
<tbody>
<tr>
<td>245</td>
<td>120</td>
<td>27.2</td>
<td>1.37</td>
<td>0.124</td>
<td>0.15</td>
<td>-67</td>
<td></td>
</tr>
<tr>
<td>245</td>
<td>120</td>
<td>27.2</td>
<td>1.37</td>
<td>0.124</td>
<td>0.30</td>
<td>-61</td>
<td></td>
</tr>
<tr>
<td>245</td>
<td>95</td>
<td>34.1</td>
<td>1.35</td>
<td>0.098</td>
<td>0.15</td>
<td>-63</td>
<td></td>
</tr>
<tr>
<td>245</td>
<td>95</td>
<td>34.1</td>
<td>1.35</td>
<td>0.098</td>
<td>0.30</td>
<td>-57</td>
<td></td>
</tr>
<tr>
<td>245</td>
<td>87</td>
<td>37.3</td>
<td>1.35</td>
<td>0.091</td>
<td>0.15</td>
<td>-61</td>
<td></td>
</tr>
<tr>
<td>245</td>
<td>87</td>
<td>37.3</td>
<td>1.35</td>
<td>0.091</td>
<td>0.30</td>
<td>-55</td>
<td></td>
</tr>
</tbody>
</table>

In this table, \( f_r \) is the resonance frequency of the bubbles; \( \kappa \) is the polytropic exponent of gas compression; \( \xi_r \) is the total damping constant; \( A \) is the pulse pressure amplitude; and 2nd and 3rd refer to harmonic pressure levels relative to the fundamental.

3. Ultrasonic Frequency 506 kHz

Observations of bubble scattering for pulses of ultrasound with a centre frequency of 506 kHz were made for bubbles of radius 107 ± 13 and 73 ± 10 \( \mu \)m, for three pulse amplitudes corresponding to approximately 0.5, 0.7, and 1.0 bars. Figure 4.24 depicts a single pulse scattered by bubbles of radius 107 \( \mu \)m, and three average periodograms for different pulse amplitudes. There are no significant differences between these spectra and those obtained for scattering from a metal rod (Figure 4.15(b)). The shape of the excitation pulse used in the numerical simulation, and its spectrum, are similar to those shown in

![Figure 4.23](image-url)
Figure 4.18 for 70.5 kHz, except scaled up in frequency. Figure 4.25 and Table 4.4 present the numerical results. Only the second harmonic component is apparent, and it is too low to be detected experimentally. Again, the damping constant is dominated by the acoustic component (total damping 0.22, acoustic damping 0.21). The results for the bubbles of radius 73 μm were similar.

4. Ultrasonic Frequency 1.69 MHz

Bubble scattering spectra were obtained for ultrasonic frequency 1.69 MHz, for two bubble sizes, 74 and 81 ± 11 μm. Three pulse amplitudes were used with each case corresponding to approximately 0.35, 0.5, and 0.7 bars.

The shape of the pulse, as scattered by an aluminum rod, and its spectrum were shown in Figure 4.16. Figure 4.26 presents the experimental results for bubbles of radius 74 μm. No harmonic components are visible; the peak at 900 kHz was spurious. The shape of the excitation function, used for the numerical comparison, and its spectrum are shown in Figure 4.27. Figure 4.28 presents the corresponding numerical results. Only the second harmonic is visible in the numerical solutions, and its level is very low (Table 4.4). The damping constant for these cases is again dominated by the acoustic component. The ratio of resonance to driving frequency is roughly 0.025.
Figure 4.25 (a) A numerical solution, showing the radial velocity response for a bubble of radius 107 μm, to a pulse of frequency 506 kHz, width 50 μs, and amplitude 0.5 bars.
(b) The periodogram of the response shown in (a).
(c) As in (b), but for a pulse of amplitude 0.7 bars.
(d) As in (c), but for a pulse amplitude of 1.0 bars.

Table 4.4 Numerical Simulation of Experiments (3)

<table>
<thead>
<tr>
<th>f (kHz)</th>
<th>R₀ (μm)</th>
<th>f₀ (kHz)</th>
<th>κ</th>
<th>δ</th>
<th>A (bar)</th>
<th>2nd Harmonic</th>
</tr>
</thead>
<tbody>
<tr>
<td>506</td>
<td>107</td>
<td>30.5</td>
<td>1.37</td>
<td>0.22</td>
<td>0.5</td>
<td>-75</td>
</tr>
<tr>
<td>506</td>
<td>107</td>
<td>30.5</td>
<td>1.37</td>
<td>0.22</td>
<td>0.7</td>
<td>-72</td>
</tr>
<tr>
<td>506</td>
<td>73</td>
<td>44.6</td>
<td>1.36</td>
<td>0.15</td>
<td>0.5</td>
<td>-69</td>
</tr>
<tr>
<td>506</td>
<td>73</td>
<td>44.6</td>
<td>1.36</td>
<td>0.15</td>
<td>0.7</td>
<td>-64</td>
</tr>
<tr>
<td>506</td>
<td>73</td>
<td>44.6</td>
<td>1.36</td>
<td>0.15</td>
<td>1.0</td>
<td>-61</td>
</tr>
<tr>
<td>1690</td>
<td>74</td>
<td>43.9</td>
<td>1.35</td>
<td>0.41</td>
<td>0.35</td>
<td>-58</td>
</tr>
<tr>
<td>1690</td>
<td>74</td>
<td>43.9</td>
<td>1.35</td>
<td>0.41</td>
<td>0.5</td>
<td>-81</td>
</tr>
<tr>
<td>1690</td>
<td>74</td>
<td>43.9</td>
<td>1.35</td>
<td>0.41</td>
<td>0.7</td>
<td>-79</td>
</tr>
<tr>
<td>1690</td>
<td>74</td>
<td>43.9</td>
<td>1.35</td>
<td>0.41</td>
<td>1.0</td>
<td>-76</td>
</tr>
<tr>
<td>1690</td>
<td>81</td>
<td>40.0</td>
<td>1.35</td>
<td>0.43</td>
<td>0.35</td>
<td>-83</td>
</tr>
<tr>
<td>1690</td>
<td>81</td>
<td>40.0</td>
<td>1.35</td>
<td>0.43</td>
<td>0.5</td>
<td>-81</td>
</tr>
<tr>
<td>1690</td>
<td>81</td>
<td>40.0</td>
<td>1.35</td>
<td>0.43</td>
<td>0.7</td>
<td>-80</td>
</tr>
</tbody>
</table>

In this table, f₀ is the resonance frequency of the bubbles; κ is the polytropic exponent of gas compression; δ is the total damping constant; and A is the pulse pressure amplitude. The third harmonic component was not evident.

5. Passive Detection

Bubble sounds could be detected in the frequency range from 10 to 60 kHz, as the bubbles were released from the bubble generator orifice, by using either a hydrophone or an ultrasonic transducer as a passive detector, and amplifying the resulting signal with a tunable receiver. The nature of these sounds was investigated using a miniature hydrophone and preamplifier, having a combined bandwidth of 200 kHz. On another occasion, a more sensitive hydrophone, having a built-in preamplifier, was used with a measuring amplifier. The
Figure 4.26 (a) A 1.69 MHz pulse scattered by bubbles of radius 74 µm.
(b) The average of four periodograms of records such as in (a).
(c) As in (b), but with the amplitude of the transmitted pulse increased by 3 dB.
(d) As in (b), but with the amplitude of the transmitted pulse increased by 6 dB.

Figure 4.27 (a) The excitation pulse used in the 1.69 MHz numerical solution. A 90% cosine taper was applied, to simulate the response of the transducer.
(b) The periodogram of the excitation pulse.
Figure 4.28 (a) A numerical solution, showing the radial velocity response for a bubble of radius 74 μm, to a pulse of frequency 1.69 MHz, width 30 μs, and amplitude 0.35 bars. (b) The periodogram of the response shown in (a). (c) As in (b), but for a pulse amplitude of 0.5 bars. (d) As in (b), but for a pulse amplitude of 0.7 bars. 

Bubbles ranging in radius from 75 to 350 μm were studied. For bubble sizes less than 160 μm, the signals were noisy, and only the fundamental components of the bubble oscillations were evident. For bubbles of about 200 μm radius, the spectra were rich in harmonic content, while for the largest bubbles (350 μm), the harmonic levels were again lower. Figures 4.29, 4.30, and 4.31 illustrate some of the experimental results, and Table 4.5 summarizes the harmonic levels observed.

This process was simulated numerically by specifying zero driving force and an initial departure from equilibrium radius. The assumption of spherical geometry was maintained. To abandon that assumption would have required an entirely new and more difficult analysis and program. Bubbles observed forming at the ends of needles do not seem to depart greatly from spherical shape. Values of the polytropic gas constant and the damping constant for free oscillations were obtained from Chapman and Plesset [1971]. The numerical results for freely oscillating bubbles with radii close to those measured are presented in Figures 4.32 and 4.33, and in Table 4.6.
Figure 4.29 (a) Sound, produced by freely oscillating gas bubbles, of radius $160 \pm 40 \, \mu m$, acquired using the 200 kHz hydrophone and digitizing system.
(b) The periodogram of the record shown in (a).
(c) The average of ten periodograms for bubbles of radius $160 \, \mu m$.

Figure 4.30 (a) A periodogram of sounds produced by freely oscillating bubbles of radius $180 \pm 40 \, \mu m$.
(b) As in (a), but averaged for 100 records.
The experimentally derived spectra are more complex than the numerically derived results. This may be partly due to the cumulative contributions from many bubbles. Although the numerical results predict that the bubble oscillations damp out long before the next bubble would form at the end of the needle, the reverberations from tank surfaces would persist and interfere.

<table>
<thead>
<tr>
<th>R (μm)</th>
<th>f_0 (kHz)</th>
<th>2nd Harmonic (dB)</th>
<th>3rd Harmonic (dB)</th>
<th>4th Harmonic (dB)</th>
<th>records averaged</th>
</tr>
</thead>
<tbody>
<tr>
<td>100 ± 25</td>
<td>27.5</td>
<td>-21.5</td>
<td>-22.5</td>
<td>-30</td>
<td>10</td>
</tr>
<tr>
<td>150 ± 19</td>
<td>23</td>
<td>-16</td>
<td>-24.5</td>
<td>-34.5</td>
<td>100</td>
</tr>
<tr>
<td>180 ± 40</td>
<td>26</td>
<td>-30</td>
<td>-40</td>
<td>100</td>
<td></td>
</tr>
<tr>
<td>210 ± 45</td>
<td>16</td>
<td>-34.5</td>
<td>-36.5</td>
<td>10</td>
<td></td>
</tr>
<tr>
<td>350 ± 70</td>
<td>10.5</td>
<td>-24</td>
<td>-26</td>
<td>10</td>
<td></td>
</tr>
</tbody>
</table>

In general, the frequency of the free oscillations agrees with the theoretically predicted resonance frequency, and the harmonic content corresponds to what would be expected for a departure from equilibrium radius less than 10 per cent.
Figure 4.32 (a) A numerical result showing the response of a bubble with an initial radial displacement of + 10% of its equilibrium radius, 160 μm.
(b) The periodogram of the response shown in (a).
(c) As in (b), but for an initial radial displacement of + 20%.

Figure 4.33 (a) A numerical result showing the response of a bubble with an initial radial displacement of + 10% of its equilibrium radius, 200 μm.
(b) As in (a), but for an initial displacement of 50%.
(c) As in (a), but for a bubble of radius 350 μm, displaced from equilibrium by + 10%.
V. DISCUSSION

A. Discussion of the Results of the Present Study

The numerical studies have shown that the behaviour of bubbles in an ultrasonic field of low to moderate amplitude (0.01 to 1 bar) is essentially linear, unless the driving frequency matches one of the following conditions: It is within 10% of the radial resonance frequency of the bubble, or it is a low integer fraction (1/2, 1/3, ...) of the resonance frequency, or, to a lesser extent, it is a low harmonic of the resonance frequency. The maximum of the second harmonic component relative to the fundamental component occurred when the driving frequency was one half of the radial resonance frequency. All harmonics exhibited absolute maxima at the resonance frequency.

For frequencies far above the resonance frequency, the second harmonic component was less than -70 dB with respect to the fundamental. Far below resonance, the behaviour was more nonlinear, but the second harmonic was still less than -30 dB with respect to the fundamental. Figure 3.6 shows that, for an ultrasonic frequency of 1 MHz and amplitude greater than 0.017 bar, the ratio of the second harmonic component to the fundamental will be greater than -40 dB for bubbles of radius less than 4 μm. For an ultrasonic frequency of 500 kHz, Figure 3.8 shows that this range extends to 8 μm. For ultrasonic frequencies near the resonance frequency of the bubble, the harmonic content was shown to be sensitive to the magnitude of the damping.

<table>
<thead>
<tr>
<th>( R_0 ) (μm)</th>
<th>( f_0 ) (kHz)</th>
<th>( \kappa )</th>
<th>( \delta_r )</th>
<th>( \Delta R )</th>
<th>2nd (dB)</th>
<th>3rd (dB)</th>
<th>4th (dB)</th>
</tr>
</thead>
<tbody>
<tr>
<td>74</td>
<td>42.53</td>
<td>1.27</td>
<td>0.11</td>
<td>10</td>
<td>-11</td>
<td></td>
<td></td>
</tr>
<tr>
<td>84</td>
<td>37.59</td>
<td>1.28</td>
<td>0.10</td>
<td>10</td>
<td>-12</td>
<td></td>
<td></td>
</tr>
<tr>
<td>101</td>
<td>31.35</td>
<td>1.29</td>
<td>0.097</td>
<td>10</td>
<td>-11</td>
<td></td>
<td></td>
</tr>
<tr>
<td>161</td>
<td>19.78</td>
<td>1.37</td>
<td>0.085</td>
<td>10</td>
<td>-12.5</td>
<td></td>
<td></td>
</tr>
<tr>
<td>200</td>
<td>15.97</td>
<td>1.32</td>
<td>0.082</td>
<td>10</td>
<td>-11</td>
<td></td>
<td></td>
</tr>
<tr>
<td>350</td>
<td>9.19</td>
<td>1.34</td>
<td>0.068</td>
<td>10</td>
<td>-13.5</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

TABLE 4.6 Harmonic levels from freely oscillating bubbles - numerical

Acoustic Harmonic Levels re. Fundamental

- 2nd
- 3rd
- 4th

-70 dB
-30 dB
-40 dB
-20 dB
coefficient. This was also observed in the comparison with the numerical results of Lauterborn [1976], and it suggests an indirect method of determining the damping coefficient. The primary effect of variation of the polytropic constant, K, was to alter the resonance frequency, and thereby influence the nonlinearity by shifting the resonance frequency either toward, or away from, the driving frequency.

Tapered, pulsed, ultrasonic excitation was used for most cases studied. Other numerical studies of bubble oscillations have generally used continuous, untapered excitation. The transient oscillations, which form part of a bubble's response to pulsed ultrasound, appear as discrete components in the spectrum near the resonance frequency, if the driving frequency and resonance frequency are greatly separated. If the driving frequency is close to the resonance frequency, the transient oscillations produce an asymmetry in the spectrum toward the resonance frequency. This could form the basis of a noninvasive technique to estimate the size of bubbles in tissue.

In experiments, it was observed that the signal to noise ratio (SNR) for scattering from bubbles was greatest at the highest frequency used, 1.69 MHz. The SNR was progressively worse for frequencies 506, 245, and 70.5 kHz. For continuous wave ultrasound, it was possible to detect bubbles only at 1.69 MHz, and, for that case, the bubble signal appeared as a modulation of the background signal which was caused by reflections from tank surfaces. For lower frequencies, the background signal completely masked the bubble signal. At 1.69 MHz, the ultrasonic frequency was greater than twenty times the resonance frequency of the bubbles, and no nonlinear effects were observed. The Doppler effect was not useful for separating the bubble signal from the background signal, except at the highest frequency. Also, the continuous wave experiments demonstrated that a transducer which is resonant at the second harmonic of the driving frequency should not be used to detect second harmonic scattering, since nonlinear artifacts are likely to occur.

It was shown that, by using pulsed ultrasound, scattering from bubbles could be detected at all frequencies, although with some difficulty at the lowest frequency, 70.5 kHz. The capture of single pulses of ultrasound for subsequent spectral analysis was an innovation of this thesis. The 7-bit resolution of the A/D converter limited the spectral dynamic range of the instrument to about 40 dB for a single pulse. This was improved by averaging. Frequency resolution was limited in the case of ultrasonic frequency 70.5 kHz, by the necessity to keep the record length short, to minimize the interference from other scatterers.

Although the pulse capture instrument allowed detection of bubbles at all frequencies used, nonlinear effects were not observed, except possibly in one case where the ultrasonic frequency was 1.2 times the resonance frequency. On the other hand, passive detection of free bubble oscillations showed them to be rich in harmonic content. The most important limiting factor in detection of nonlinear scattering proved to be the response of the transducers, which suffered from low sensitivity at the second harmonics.
sensitive, and directional transducers are required. Flat frequency response would allow tuning to the resonance frequency, and would simplify interpretation of the acquired spectra. A second limiting factor proved to be the inability to produce, reliably, bubbles of radius less than 50 μm. Smaller bubbles would move the resonance frequency up to a more convenient ultrasonic range.

In general, the experimental results support the theoretical prediction that, for low to moderate amplitudes of pulsed ultrasound a bubble behaves as a linear scatterer, unless the frequency of the ultrasound is close to the resonance frequency for radial oscillations of the bubble.

B. Review of Previously Published Results

1. Welsby and Safar

Welsby and Safar [1969] may be credited with having first suggested the idea of using nonlinearity as a means to detect bubbles. They wrote:

Observations of changes in nonlinearity can be used generally as a means of detecting the appearance of small bubbles, whatever their cause. This gives, for example, a sensitive method of detecting bubbles in the blood or tissue of an animal.

To arrive at this conclusion, these authors considered a liquid containing a uniform distribution of stable gas nuclei or microbubbles. (The assumption of uniformly distributed gas nuclei is usual, but is still controversial. The subject of bubble nucleation is being actively researched [Yount et al. 1977].) Welsby and Safar expanded a form of the bubble equation, keeping first and second order terms, to show that, in the presence of a pressure wave, bubble volumetric displacement would contain fundamental and second harmonic components. They asserted that the microbubble population would then constitute a three-dimensional array source of distortion wavelets, which would add to a uniformly distributed background nonlinearity of the medium. By comparing the compressibilities of the gas and liquid phases of the medium, they concluded that "the presence of gas bubbles will have an appreciable influence on medium nonlinearity, long before they would have an influence on the first order properties of the medium." They predicted that this should occur for gas concentrations of approximately one in one hundred million, by volume. They also suggested that the nonlinearity should become observable as the cavitation threshold is reached, because then the bubble nuclei would grow in size, and the resonance frequency would drop towards the working frequency.

In summary, Welsby and Safar consider very small bubbles or gas nuclei, ultrasonic frequencies below the resonance frequency, and intensities approaching the cavitation threshold. While it would be desirable to be able to detect gas nuclei, the bubbles which are of
primary concern are those which are large enough to be important physiologically, but small enough to be difficult to detect with current techniques. Such bubbles range from 10 to 100 μm in radius. The resonance frequency for such bubbles would be below the ultrasonic frequency of most medical instruments, and intensities approaching threshold the cavitation must be avoided. Thus the recommendation of Welsby and Safar would not apply to the usual case.

2. Evans

Evans [1975] at Newcastle, with the cooperation of Welsby, conducted experiments to test the suggestion of Welsby and Safar [1969] for detection of decompression bubbles. Undamped transducers were driven at their resonance frequency, 160 kHz, in a through-transmission arrangement. Through-transmission was chosen on the recommendation of Welsby for the following reason. For bubbles much smaller than a wavelength, closely spaced, and far from resonance, the distortion components would add in phase in the direction of the incident wave, while, in other directions, there would be partial cancellation. (For sound sources closer together than one sixth of a wavelength, the effects would not simply be additive, but source interaction would produce a greater power [Skudrzyk, p. 368]). Through-transmission implies, however, that a larger fundamental component would have to be excluded by the receiver, which was tuned to the second harmonic.

Evans reported, "a swarm of bubbles introduced in a container of water, surveyed in this way, led to a most impressive rise in the level of the harmonic signal."

Similar observations have been made in preliminary experiments for this study, but it is difficult to make a quantitative statement about the scattering in this situation. For comparison, however, it was observed that Doppler ultrasound was successful at detecting individual bubbles of smaller size, where no nonlinear effect was noted.

Evans reported decompression trials on anaesthetized guinea-pigs. These trials were inconclusive because of "large apparently spontaneous variations in the second harmonic signal strength." Evans explained that this might have been caused by frequency drift in the transmitter, or drift of the centre frequency of the notch filter employed to reject the fundamental. To circumvent the drift problem, the notch filters were eliminated, and, instead, sensitivity to the second harmonic was enhanced by switching to a transducer resonant at 485 kHz for the transmitter, and one resonant at 970 kHz for the receiver. Evans reported, "the apparently random fluctuations which continued to influence the signal, persisted throughout, and reluctantly the technique was abandoned."

A similar arrangement was tested in the initial stages of this work with transducers resonant at 1.69 and 3.38 MHz. As noted earlier, the receiving transducer, being driven at one half its resonance
frequency, can distort the signal, and this distortion can change spontaneously, as reported by Evans.

Nevertheless, Evans closed optimistically, stating, "The study of distortion is a potentially very sensitive technique, and it might well repay further investigation."

3. Martin, Hudgens, and Wonn

As part of a contract with the U.S. Office of Naval Research, these authors published a report describing application of the nonlinear technique to detection of bubbles in divers. Their apparatus consisted of two sets of transducers in through-transmission, operated at 115 kHz. Two tuned receivers were used to detect fundamental and second harmonic levels simultaneously, and record them on a strip chart. Some preliminary studies were made for which they made the following claims:

Westinghouse laboratory tests with a 1 mW/sq.cm. ultrasonic field propagated through a water bath have detected bubbles 2 to 15 microns in diameter by this method. In addition, tests run on 35 - 50 mW/sq.cm. insonified meat parts, with electrolytically produced bubbles injected through the venous system thereof, have detected bubbles as small as 8 - 10 microns, and concentrations of less than 10e-07 void fraction (volume ratio of bubble to insonified liquid in blood vessel).

Details of the measurements were not provided.

These authors conducted a series of 18 man-dives to test their apparatus. Several body sites were monitored, but the thigh was found to be most successful. On five dives they reported detection of second harmonic excursions while the fundamental level remained quiescent. They claimed that these excursions represented the passage of single bubbles, or when the excursions were smaller, but more numerous, large numbers of smaller bubbles. Results from a strip chart were presented as examples of proof of bubble detection, but these appear to be inconclusive.

On the basis of the present work, the claims for their "in vitro" studies should be regarded with skepticism. The claims for their manned studies do not seem unreasonable when it is noted that substantial numbers of bubbles are often detected during decompression from similar dives using Doppler ultrasound [Spencer 1977]. The nonlinear technique would not then seem to be as sensitive as their "in vitro" claims suggest.

4. Moulinier

This author wrote his doctoral thesis [1978] on the subject of detection of bubbles in biological tissue by measuring the distortion level of ultrasound. Although the subject matter of his research was similar to that of this thesis, the approach used by him differed greatly from the one adopted here. Moulinier's work is organised in four parts: a review of the ultrasonic techniques available for
detection of bubbles, a discussion of a theory of distortion of the ultrasound, a detailed description of the development of an instrument designed to measure the second harmonic distortion level, and a description of "in vitro" and "in vivo" experiments to test the apparatus.

The theoretical development of Moulinier was an elaboration of that due to Welsby, so that the same conclusions were reached. In addition, the author considered how uniformly distributed distortion sources between two collinear transducers, contribute to a distortion level. His main conclusion was that the distortion ratio increases as the distance between the two transducers, with the qualification that attenuation, divergence, and imperfect reflections at the transducer faces reduce this effect.

The effect of bubbles which resonate at the working frequency of 100 kHz was considered briefly, and it was concluded that, since such bubbles would unduly influence the distortion level, the ultrasonic frequency should be chosen low enough to avoid resonance. This approach was motivated by the desire to obtain a correlation between the distortion level and the total gas content of the liquid. This might be possible if it were certain that the bubbles were being driven well below resonance. A frequency of 100 kHz would preferentially excite second harmonic oscillations in bubbles in the neighbourhood of 15 and 30 μm radius, however, and it would be difficult to rule out contributions from such bubbles. It would be better to regard the resonance phenomenon as a valuable effect for detecting bubbles, and to obtain an estimate of the gas content from a measurement of the bubble size distribution.

The largest part of Moulinier's thesis concerns the development of his instrumentation, especially the second harmonic receiver, which posed the greatest challenge. This instrument used through-transmission ultrasound of frequency 102.5 kHz. The receiving transducer was resonant at 205 kHz. The signal from the receiver was fed to two amplifiers, one for the fundamental, and the other for the second harmonic. Peak detectors and a ratio circuit provided three outputs, proportional to the signal level at the fundamental, the second harmonic, and the ratio of these two.

Preliminary tests [Guillaud and Moulinier 1978] with this instrument were conducted using bubbles formed by decompression in water and gelatin. Rapid changes in both the second harmonic and fundamental levels were observed, most likely caused by the presence of resonant bubbles. The first instrument provided a measurable distortion limit of 50 dB, which, it was concluded, did not permit observation of significant distortion in the absence of resonating bubbles. This conclusion is in accord with both the observations and calculations reported here.

In his thesis, Moulinier compared the efficacy of his distortion instrument with that of a Doppler ultrasonic bubble detector for detecting bubbles, both "in vitro" and "in vivo". For the tests "in vitro", electrolytically-produced bubbles were introduced into water
flowing through plastic tubes. Although the Doppler and distortion instruments produced bubble indications which were consistent, the Doppler technique appears to have been more sensitive. For the "in vivo" tests, a rabbit was cyclically compressed and decompressed between surface pressure and pressure equivalent to a depth of 80 msw (1 to 8 bars), producing large numbers of bubbles until death. The distortion transducers were placed across the thigh, while the Doppler transducers were implanted around the vena cava. The indications from the two techniques were again consistent.

It appears, then, that Moulinier's distortion instrument was successful at detecting bubbles. Unfortunately, it is not possible to make a quantitative statement from his results, because there is not sufficient information on the number and size of the bubbles involved. Perhaps the bubbles were quite close to resonant size; perhaps they were very many and large. Inherent in Moulinier's apparatus was the possibility of an artificial distortion, created in the second harmonic transducer, although he evidently did not observe this effect in thorough preliminary tests.

VI SUMMARY AND CONCLUSION

The most common current practice in decompression research is to use Doppler ultrasound for detection of bubbles in blood. This technique can detect only moving bubbles, however, and its sensitivity is limited by the presence of other scatterers which contribute a masking signal. To a lesser extent, ultrasonic imaging is also used. Although able to detect stationary bubbles, the imaging method is also restricted in its detection of small bubbles by the masking effect of surrounding scatterers. To avoid this clutter, the suggestion was made that, since bubbles behave in a more nonlinear fashion than the other scatterers in the blood and tissue, they could be detected preferentially by monitoring the second harmonic of the scattered ultrasound. Previous experiments with this technique proved to be inconclusive, but, in some respects, promising.

The research reported in this thesis was carried out to evaluate that proposal from a more fundamental perspective, to determine whether the concept was theoretically and experimentally sound, and to elucidate the results of previous experimental efforts.

A review of the literature of bubble acoustics and cavitation showed that information which described the nonlinearity of the response of a bubble to ultrasound at medical diagnostic amplitudes and frequencies was lacking. Nevertheless, the most common model for bubble dynamics, due originally to Rayleigh, but enhanced by many others, was amenable to numerical solution using a computer, and could
provide the information desired.

Many numerical results were obtained, with both continuous wave and pulsed ultrasound, for frequencies ranging from far above to far below resonance. As well, for two fixed frequencies, 500 kHz and 1 MHz, numerical solutions were obtained (Figures 3.6 and 3.8) for bubbles ranging in radius from 1 to 20 μm. Comparison of the numerical results with analytical and numerical results of three other authors showed generally good agreement. It was shown that for frequencies far above resonance the bubbles behaved in an essentially linear fashion, with the second harmonic component more than 70 dB below the fundamental, even for an ultrasonic amplitude of 1.2 bars. Far below resonance, the bubbles behaved more nonlinearly, but at diagnostic intensities the second harmonic component was, although significant, still about 30 dB below the fundamental. In contrast, at resonance and low integer submultiples of resonance, the second harmonic component grew to within a few dB of the fundamental. For an ultrasonic frequency of 1 MHz and an amplitude greater than 0.017 bar, harmonic distortion greater than -40 dB would occur for scattering from bubbles less than about 4 μm in radius, while for ultrasound at 500 kHz this range would extend up to about 8 μm.

Experimentally, it was found that continuous wave ultrasound was not useful for detecting individual small bubbles for frequencies much below 1 MHz because of interference from background reflections. The sensitivity of the nonlinear technique to instrument artifacts was also demonstrated. To circumvent the problem of excessive background signal, pulsed ultrasound was used. This necessitated the development of a system to capture and digitize the scattered ultrasound on a single shot basis. The pulses were then analysed for spectral content using a computer. The predicted, harmonic levels for the experiments were, in general, too low to be observed with the apparatus used, except for the case of passive detection of bubbles oscillating at their natural frequency. These resonant oscillations exhibited relatively high harmonic content. The experiments demonstrated the need for sensitive broadband transducers, both for quantitative experimental investigations, and for potential use in a practical bubble detector. As well, there remains a need for reliable production of bubbles ranging from 50 μm down to 1 μm in radius.

A comparison with the results of other experimenters is difficult because information concerning the number and size of bubbles involved in their experiments is not available. In addition, these authors generally used a method which was shown in this study to be prone to artifacts, which may account for some of their unexplained results. Nevertheless, there is agreement with many of their observations. In general, their results are understandable in light of the findings of this study.

At present, the usefulness of the nonlinear technique appears limited to the detection of groups of bubbles of resonance or one-half resonance size (Appendix A). For practical frequencies, these are generally very small - less than 10 μm in radius. For the future, as the technology of ultrasonic transducers advances, it may be possible to
measure the size of bubbles in tissue through a determination of their resonance frequency. The appearance of nonlinear effects could be used to identify that frequency. Coupled with an imaging technique to give the location and number of bubbles, the information about the bubbles in tissue would be complete.

A summary of the contributions of this thesis follows:

1. The numerical work provided a quantitative description of the nonlinearity of the response of bubbles to ultrasound at amplitudes and frequencies appropriate for bubble detection in man. The results were presented so as to be easily interpretable for the bubble detection application. In contrast with previous numerical studies of bubble dynamics, the form of the driving ultrasound was generally specified as a tapered pulse, to simulate better the practical case. More accurate values of the polytropic exponent and damping constant, based on Prosperetti's [1977b] results, were used than in previous numerical studies, and the results were shown to be sensitive to those parameters when the bubble oscillations were significantly nonlinear. The transition from essentially linear, to nonlinear behaviour was described. Strongly nonlinear behaviour was predicted when the ultrasonic frequency was close to, either the resonance frequency of the bubbles, or one half or one third of the resonance frequency. Figures 3.6 and 3.8 summarize the contribution in this regard. The increase in the nonlinearity accompanying the increased amplitude of the oscillations at resonance is not surprising. On the other hand, it is not obvious that the ratio of the second harmonic to the fundamental is maximum when the driving frequency is one half of the resonance frequency.

2. The technique of spectral analysis of single pulses of ultrasound scattered by bubbles was introduced, and demonstrated for ultrasonic frequencies from 70.5 kHz to 1.69 MHz. This technique circumvents the problem of masking by reflections from other surfaces in the tank. That problem makes the use of continuous wave ultrasound inappropriate for experiments. The single-pulse technique produces an ultrasonic snapshot of a single bubble or group of bubbles. In principle, a detailed comparison between experiment and theory is possible, but realization of that goal awaits the availability of a sufficiently sensitive, broadband, and directional transducer to use as the receiver. While the numerical results provide more immediately useful information, the experimental work was more difficult, and the technique should have a greater eventual impact.

3. The numerical and experimental results together have elucidated the results of previous researchers who experimented with, or discussed detection of the second harmonic as an indicator of the presence of bubbles. That the ultrasonic frequency must be close to the resonance frequency or, preferably, one half of the resonance frequency of the bubbles, had not been recognized. For a fixed-frequency device this limits the technique to detection of bubbles of two sizes primarily. It will therefore be useful when detection of bubbles of those particular sizes is required. The technical problems associated with building a more generally applicable bubble detector were pointed out (Appendix A), the most important of these being transducer design.
ACKNOWLEDGEMENTS

It is a pleasure to acknowledge Professor G.W. Johnston, my supervisor, for his enthusiastic encouragement and his interest in this research. Thanks are also due to Professor J.B. French and Dr. L.A. Kuehn for their support, and for encouraging the cooperation between UTIAS and DCIEM. Mr. R.Y. Nishi assisted greatly in many discussions by sharing his knowledge of scattering and underwater acoustics, and by providing helpful criticism. Thanks are also due to Professor H.S. Ribner and Dr. W.G. Richarz for their roles on the supervisory committee, and to Dr. K.E. Kisman, formerly of DCIEM, who at the outset shared his equipment and materials. Professor R.S.C. Cobbold and Dr. H.M. Merklinger reviewed the thesis and provided helpful criticism. Mssrs. Larry Allin and Lee Ferrari were instrumental in development of the pulse capture system. Mr. Stanley Macdonald constructed the transducers with consummate skill and care, and frequently assisted with the development of the experimental apparatus. The generosity of Dr. M.M. Taylor and Mr. Martin Tuori in allowing access to their minicomputer and graphics facilities is gratefully acknowledged. Thanks are also due to Mrs. Sandra Wright for assistance with the software, to Miss Dawn Gardham for assistance with the typing, and to my sister, Ruth Anne Eatock, who took time out of her busy schedule at Caltech to draw the frontispiece.

APPENDIX A. Implications for Bubble Detection in Blood and Tissue

The nonlinearity of the scattering of ultrasound by bubbles has been studied numerically and experimentally to determine the utility of this effect for the detection of bubbles in blood. In this Appendix, the implications of the results presented here are considered in light of this practical question.

The present study has shown that, for a fixed ultrasonic frequency, only a relatively small range of bubble sizes contributes significantly to nonlinear scattering of the ultrasound. The bubbles contributing are those which are close to the resonance size for that frequency, or smaller. For ultrasound of frequency 1 MHz, the bubbles which behave nonlinearly are less than 4 μm in radius; at 500 kHz, this range extends to 8 μm. Thus, at practical ultrasonic frequencies, the bubbles which could be detected would be very small.

To detect the scattering from the bubbles, the sensitivity of the receiver must be adequate, which may pose a problem in the case of very small bubbles. To illustrate this, from Nishi [1975], the ratio of scattered to incident intensity at 1 cm from a resonant bubble in a 1 MHz ultrasonic field is approximately one part in one million (-60 dB). For this case, the numerical results indicate that the second harmonic component would be about -20 dB with respect to the fundamental. If the incident intensity were 100 mW, the scattered intensity at 1 cm would be 0.1 μW, and the second harmonic power level would be 0.001 μW. This number does not include the effect of interference...
with the incident field. If an ideal transducer with a 100 per cent electromechanical conversion efficiency and an impedance of 100 ohms is assumed, then the total received voltage level would be about 3 mV. Hence, the level at the second harmonic would be about 300 μV. Factors which would reduce these numbers include: greater distance to the scatterer, an absorbing medium, a nonresonant bubble, a less-than-perfect transducer, and a lower transmitted power level. These could be countered by increasing the power level, but the value used in this example, 100 mW, is not conservative, especially for a continuous wave device. Except for the case of resonant bubbles close to the transducers, the combined effect of these factors would likely reduce the signal level by one to two orders of magnitude. This signal must generally be detected in competition with signals from other scatterers.

The nonlinear effect would be useful in the case where a large number of linear nongaseous scatterers masks the contributions from resonant bubbles. In that case, the bubbles would not have a noticeable effect on the overall scattering level, but would have an observable effect on the second harmonic component. Unless it were intended that the device be sensitive primarily to bubbles of one size, however, it would be necessary to augment the transmitted bandwidth, either by sweeping the frequency, or by transmitting a sharp pulse of ultrasound. To avoid contributions from as many unwanted scatterers as possible, it might also be necessary to use range gating of some description, for example, pulse ranging as employed in the experiments described here. Alternatively, detection of the Doppler shifted second harmonic component for moving scatterers would help to reduce the background signal level, but this technique generally requires a fixed frequency.

A system designed to detect the nonlinear effect would have special requirements. For a broadband system, e.g. swept-frequency or pulsed systems, the design of the receiving transducer would present a problem, because there is a trade-off between bandwidth and sensitivity. Even for a fixed frequency device, the receiving transducer should not be resonant at the second harmonic, since that would, in essence, degrade the distortion specification of the receiver, nor should it be resonant at the fundamental frequency, because that would limit the sensitivity at the second harmonic too much. Thus, the transducers would require high sensitivity and flat response over an octave. Such transducers are not yet available, however, for frequencies above a few hundred kHz. After the transducer, the rest of the receiver must have a low noise figure and low distortion specifications.

The transmitter would also have special requirements. For a fixed frequency device, the transmitter would need only produce a spectrally pure signal, which would not be a difficult requirement to meet; a standard resonant transducer would suffice. For a swept frequency device, however, a broad bandwidth projector would be required. Ideally, it should have flat transmitting response or, more realistically, a response with constant slope versus frequency. For a pulsed device, a broad bandwidth transducer would also be required, but it would be necessary to compromise between very sharp pulses which
excite a broader range of bubble sizes, and more gradual pulses which have lower signal levels at the harmonics. Except in imaging systems, spatial resolution is not a primary consideration for bubble detection, so pulses could be quite long (e.g. 13.5 µs would give 1 cm resolution), thus allowing the use of lower frequency ultrasound and more gradual pulses. For pulses of a single frequency, a damped disc transducer would suffice; but, if pulsed and swept-frequency operation were combined, then the transducer would again prove to be a limitation.

In summary, because of the narrow band nature of nonlinear scattering by bubbles, using this effect as the basis for building a practical device to detect bubbles poses demanding technical problems. There are basic trade-offs between bandwidth and sensitivity, and between ultrasonic frequency and pulse width. The requirements for the transducers are not met by the currently available technology.

After reviewing the results obtained in this work, and the others available in the literature, the prospect of using the nonlinearity of ultrasonic scattering by bubbles to improve detection of bubbles in biological tissue does not, at present, appear bright. Bubbles are, for practical purposes, linear scatterers, except when the frequency of the incident wave is close to their resonance frequency. Unless a practical method for sweeping the ultrasonic frequency becomes available, the target bubble population would be restricted to a narrow range. Nevertheless, if bubble resonance could be detected, then a bonus would be achieved, for that would constitute a noninvasive method, independent of instrument sensitivity, for estimating bubble size. Such information is not currently available in decompression research.

The present study suggests a method of sizing bubbles, which does not rely on matching the resonance frequency exactly, nor on producing a nonlinear effect. When pulsed ultrasound is used, transient bubble oscillations are excited which appear in the spectrum in the neighbourhood of the resonance frequency. Thus, pulse spectrum analysis could be used to obtain an estimate of the resonance frequency, and, hence, the bubble size, provided that the following conditions are met: the pulses must be sharp enough to excite the transient, the receiver chain must have flat frequency response, the receiver must be sufficiently sensitive, and the spectrum must be acquired with adequate resolution.

The observations on freely oscillating bubbles suggest experimentation with sensitive ultrasonic receivers to determine whether bubble formation could be detected passively "in vivo". If successful, this technique, analogous to acoustic emission spectroscopy, would permit the natural frequency of oscillations to be measured, and hence provide an estimate of bubble size. The success of such a technique would depend, however, on the process by which bubbles are released into the blood. If, as is frequently speculated, the bubbles originally grow in crevices, then upon their release they may well oscillate in a manner similar to their behaviour when released from the end of a needle.
An expression for the acoustic radiation damping of the radial oscillations of a gas bubble was contributed by Smith [1935]. The effect of the viscosity of the liquid on the growth or collapse of a bubble was discussed by Poritsky [1952]. Devin [1959] discussed damping resulting from heat transfer, and derived an expression for the thermal damping coefficient, applicable to resonant oscillations of the bubble. He compared the relative importance of the three damping mechanisms as a function of the resonance frequency of the bubble. The damping coefficients have also been discussed by Nishi [1975] and Prosperetti [1977(b)]. The latter author derived an expression for the thermal damping coefficient, which was applicable to a broad range of bubble sizes and driving frequencies. The derivation which follows, leading to the form of the damping coefficients used in this thesis, is basically that of Prosperetti [1977(b)]. The symbols used, however, are consistent with those which have been defined in this thesis. For details of the thermal damping term, which is too involved to reproduce here, reference should be made to Prosperetti’s [1977(b)] paper. That author also provided tables of calculated values. For a justification of the form of the viscous damping term, reference should be made to Poritsky [1952].

Consider a spherical gas bubble in a fluid, subjected to a steady monochromatic sound field. The bubble is small with respect to the wavelength of the sound in the fluid, i.e. $kR_0 \ll 1$. The equation of continuity of pressure across the gas-liquid interface is

$$ p_g - p_L = 2\sigma/R + 4\mu R/R. $$

In this equation, the vapour pressure and the viscosity of the gas have been neglected. In the linear approximation, the pressure in the liquid is the superposition of the hydrostatic pressure, and the pressures radiated by the bubble,

$$ p_L = p_{L0}(1 + e^{i\omega t}) + p_{rad}, $$

$$ e \ll 1. $$

The radiated pressure is related to the velocity potential by

$$ p_{rad} = \rho \frac{\partial \phi}{\partial t}. $$

For a small spherical source, $\phi$ may be expressed as

$$ \phi(r,t) = 2\pi R_0^2 r^{-1}(1 + i\omega R_0/c)^{-1}\exp[-i\omega(r - R_0)/c]. $$

Therefore, at the surface of the bubble ($r = R_0$), the pressure is

$$ p_L = p_{L0}(1 + e^{i\omega t}) + \rho_0 R_0^2(1 + i\omega R_0/c)^{-1}. $$

If it is assumed that the changes in gas pressure inside the bubble are small, one may write

$$ p_g = p_{g0} + p_{L0}a(R_0, t). $$

Here $a$ is of order $e$, i.e. $|a| \ll 1$. If the bubble is initially at equilibrium, then the following condition holds,

$$ p_{g0} - p_{L0} = 2\sigma R_0^{-1}. $$

For small amplitude oscillations, one may write

$$ R = R_0(1 + x), $$

with $x \ll 1$. If Equationns (56), (57), (58), and (59) are substituted
into Equation (52), the following equation is derived

$$\frac{\rho_0 R_0^2 \ddot{x}}{1 + i w R_0/c} + 4 i \dot{x} \frac{-2 \dot{x}}{R_0} = P_{L_0} [\alpha(R_0,t) - c e^{i w t}].$$

(60)

The pressure inside the bubble is assumed to behave according to a polytropic gas law. Prosperetti [1977(b)] represented heat dissipation by an effective thermal viscosity, $\mu_{th}$. He wrote

$$p_g = p_0 R_0^3 (\frac{R_0}{R})^{3 \kappa} - 4 \mu_{th} \ddot{x}.$$

Then Equation (57) becomes

$$P_{L_0} a(R_0,t) \frac{p_0 R_0^2}{1 + i w R_0/c} = 3 \kappa p_g \ddot{x} - 4 \mu_{th} \ddot{x}.$$

(61)

Substitution of Equation (63) into Equation (60) yields

$$\ddot{x} + \frac{4(\mu + \mu_{th})}{p_0 R_0^2} \ddot{x} + \frac{3 \kappa p_g}{p_0 R_0^2} \ddot{x} = \frac{-\varepsilon P_{L_0}}{p_0 R_0^2} e^{i w t}$$

(64)

The first term may be rationalized, and, since $\omega R_0/c$ is small, it may be expressed as a binomial expansion.

$$\ddot{x} = \frac{\ddot{x}(1 - i w R_0/c)}{1 + (\omega R_0/c)^2} + \frac{\ddot{x} i w R_0/c}{1 + (\omega R_0/c)^2} \cdot$$

$$\ddot{x} + \frac{\ddot{x} i w R_0/c}{1 + (\omega R_0/c)^2} = \frac{\ddot{x}[1 - (\omega R_0/c)^2 + (\omega R_0/c)^4 - \ldots]}{1 + (\omega R_0/c)^2} - \frac{\ddot{x} i w R_0/c}{1 + (\omega R_0/c)^2}$$

$$\ddot{x} = \ddot{x} - \frac{\ddot{x} (\omega R_0/c)^2 [1 - (\omega R_0/c)^2]}{1 + (\omega R_0/c)^2} - \frac{\ddot{x} i w R_0/c}{1 + (\omega R_0/c)^2}$$

$$\ddot{x} = \ddot{x} - \frac{\ddot{x} (\omega R_0/c)^2}{1 + (\omega R_0/c)^2} - \frac{\ddot{x} i w R_0/c}{1 + (\omega R_0/c)^2}.$$

(65)

If $x$ is assumed to be harmonic then $\ddot{x}$ in the third term in Equation (65) may be replaced by $-i \omega \ddot{x}$ while in the second term it may be replaced by $-\omega^2 x$. With these substitutions Equation (64) may be written

$$\ddot{x} + \left[ \frac{\mu + \mu_{th}}{\rho_0 R_0^2} + \frac{\mu_{th}}{1 + (\omega R_0/c)^2} \right] \ddot{x} + \frac{3 \kappa p_g}{\rho_0 R_0^2} \ddot{x} = \frac{-\varepsilon P_{L_0}}{\rho_0 R_0^2} e^{i w t}$$

(66)

Comparison of the coefficient of $\dot{x}$ in Equation (66) with the coefficient of $\ddot{x}$ in Equation (40), p. 30, shows

$$\mu = 4 \mu/(\rho_0 R_0^2 \omega),$$

(67)

$$\mu_{th} = 4 \mu_{th}/(\rho_0 R_0^2 \omega),$$

(68)

$$\mu_{ac} = (\omega R_0/c)/[1 + (\omega R_0/c)^2].$$

Prosperetti [1977(b)] expressed $\mu_{th}$ in terms of a dimensionless parameter $B$, defined by

$$B = 4 \mu_{th} (\frac{\omega}{\omega_0})^2.$$

(69)

Thus $\delta_{th}$ may be written

$$\delta_{th} = B (\frac{\omega}{\omega_0})^2.$$

(70)

Substitution of Equation (67) alone for $\delta$ in Equation (40) yields for the damping term the quantity $4 \mu_{ac}/R_0^2$. This differs from Lauterborn's exact expression $4 \mu \ddot{x} / R$, but for small amplitude oscillations this difference is negligible since both forms reduce to $4 \mu \ddot{x}$.

While Lauterborn's expression is correct for the viscous damping term, the error introduced by expressing this term as in Equation (67) is much less than that of neglecting the thermal and acoustic components for most cases studied in this thesis.
Equation (66) also provides an expression for the resonance frequency, \( w_0' \):

\[
\frac{w_0^2}{3K_0^2} = \frac{3w_0}{2a} + \frac{(wR_0/c)^2}{1 + (wR_0/c)^2} \omega_0^2. \tag{72}
\]

For free, or small bubbles (\( KR_0 \ll 1 \)), the third term may be neglected, and for bubbles of radius larger than 100 \( \mu \text{m} \), the second term is less than six percent of the first.

\[\text{APPENDIX C. Glossary of Symbols}\]

- \( A \): amplitude of the ultrasound
- \( B \): thermal damping coefficient of Prosperetti
- \( c \): speed of sound
- \( c_P \): heat capacity of the gas at constant pressure
- \( c_v \): heat capacity of the gas at constant volume
- \( D_G \): thermal diffusivity of the gas
- \( f \): excitation frequency
- \( f_r \): resonance frequency of the bubble
- \( k \): wave number, \( 2\pi/\lambda \)
- \( I_{R}(\omega) \): discrete periodogram
- \( I_{I} \): intensity of the incident ultrasound
- \( I_{S} \): intensity of the scattered ultrasound
- \( L_G \): thermal penetration depth in the gas
- \( M \): molecular weight of the gas
- \( P \): acoustic pressure
- \( P_G \): gas pressure in the bubble
- \( P_{G0} \): gas pressure in the bubble at rest
- \( P_L \): pressure in the liquid
- \( P_{L0} \): hydrostatic pressure
- \( P_m \): amplitude of the incident acoustic wave
- \( P_t \): Blake's threshold for uncontrolled bubble growth
- \( P_v \): vapour pressure
- \( P_m \): pressure in the liquid at a very great distance from the bubble
- \( r \): radial coordinate
instantaneous bubble radius
initial radius of the bubble
critical radius for uncontrolled bubble growth
equilibrium radius of the bubble
the universal gas constant
time for bubble collapse
ultrasonic pulse width
parameter specifying the taper of the ultrasonic pulse
absolute temperature
temperature at a very great distance from the bubble
radial velocity
instantaneous radial velocity of the bubble surface
magnitude of the fundamental component of the bubble's radial velocity
magnitude of the second harmonic component of the bubble's radial velocity
magnitude of the third harmonic component of the bubble's radial velocity
instantaneous volume change of the bubble
second harmonic component of the bubble's volume perturbations
sequence of N real values
\( \gamma = \frac{c_p}{c_v} \)
acoustic damping coefficient
thermal damping coefficient
viscous damping coefficient
total damping coefficient
total bubble resonance damping coefficient
polar coordinate
polytropic gas constant (effective, \( 1 \leq \kappa \leq 1.4 \))
wavelength of sound in the gas
coefficient of shear viscosity
effective coefficient of viscosity of Prosperetti
subharmonic threshold of Prosperetti
density of the liquid
surface tension coefficient
velocity potential
angular frequency
resonance angular frequency of the bubble
APPENDIX D. List of Equipment

1. Amplifier(s)  
   Anzac, Model AM110

2. Analogue-to-digital converter  
   Computer Labs,  
   Model 0811-1-Bin

3. Coaxial switch  
   Texscan, Model SW20

4. Counter  
   Monsanto, Model 8510

5. Frequency synthesizers (2)  
   (1) Clock and ramp source  
      Hewlett Packard, Model 3325A
   (2) Signal source  
      Comatron, Model 1013 or  
      Rockland, Model 5100

6. Function generators (2)  
   Wavetek, Model 164

7. Hydrophones  
   Bruel & Kjaer,  
   Models 8103 and 8102

8. Instrumentation Tape Recorder  
   Hewlett Packard, Model 3968A

9. Logic analyser  
   Tektronix, Model 7D01 plug-in with Model 7603 or  
   7623A mainframe

10. Measuring Amplifier  
    Bruel & Kjaer, Model 2607

11. Microprocessor  
    Motorola, M6800

12. Minicomputer  
    Digital Equipment Corp.,  
    Model PDP 11/34

13. Plotter  
    Versatec, Model 1200

14. Power Amplifier  
    ENI, Model 240L

15. Power Supplies  
    Hewlett Packard,  
    Models 62378 and 62278  
    Analog Devices, Model 925

16. Preamplifier  
    Bruel & Kjaer, Model 2650

17. Preamplifier  
    Princeton Applied Research,  
    Model 113

18. Pulse generators  
    Berkley Nucleonics Corp.,  
    Model 8010

19. Receiver  
    Watkins-Johnson,

20. Regulator  
    Tescom, Model 44-1610-24

21. RF switch  
    Watkins-Johnson, Model S7

22. Signal Analyser  
    Hewlett Packard, Model 5420A

23. Spectrum Analyser  
    Hewlett Packard, Model 3585A

24. Surgical Microscope  
    Carl Zeiss, Model OPMT.1F

25. Terminal  
    Hewlett Packard, Model 2648A

26. Translators, ECL to TTL  
    Motorola, MC10125L

27. Vector Impedance Meters  
    Hewlett Packard,  
    Models 4800A and 4815A

28. Vertical Pipette Puller  
    David Kopf, Model 7000

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The magnitude of the nonlinear effect in the scattering of ultrasound by nitrogen bubbles in water is examined for ultrasonic frequencies and amplitudes typical of those used in diagnostic medical instruments. The research is directed towards determining whether this effect could be used to detect bubbles in blood or tissue, for application in decompression research. The theory of bubble dynamics is reviewed. Numerical solutions of the modified Rayleigh equation, including the effects of acoustic, thermal, and viscous damping, and the dependence of the polytropic gas constant on frequency, are presented. A system for the spectral analysis of a single ultrasonic pulse, scattered by bubbles, is described, and the results of experiments, intended to complement the numerical investigations, are reported. For conditions typical of diagnostic ultrasound, it is shown that nonlinear scattering is significant (second harmonic greater than -40 dB with respect to the fundamental) only for the population of bubbles which are close to, or smaller than, the resonance size, for the ultrasonic frequency in use. Therefore, the use of nonlinear scattering for the detection of bubbles is limited to cases where the ultrasonic frequency is close to the resonance frequency of the bubbles, or requires variation of the ultrasonic frequency.