Theoretical characterisation of the radial and translational motion of coated microbubbles under acoustic excitation

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Abstract. Ultrasound contrast agents, in the form of coated microbubbles, are a powerful tool in current diagnostic imaging. Given their sensitive dynamic response they also have the potential to be used for quantitative measurements of the properties of the surrounding tissue (e.g. percentage perfusion or blood pressure). For this potential to be realised, however, the theoretical descriptions of bubble behaviour, in particular the constitutive equations for the microbubble shell, need to be improved and a method needs to be developed for the accurate characterisation of individual bubbles. In this paper the first steps are taken towards deriving a complete model for the coupled radial and translational motion of a coated bubble. It is then shown that with this model the bubble can be characterised by a unique set of parameters describing the bubble shell corresponding to its viscous and elastic response. This uniqueness will enable the model to be used to interpret experimental data and quantify these parameters for which accurate values are currently lacking but which are critical to predicting bubble response and hence enabling advanced diagnostic applications.

1. Introduction
Microbubbles are currently the most effective type of ultrasound contrast agent available. They consist of a gas core stabilised by a surfactant or polymer shell (figure 1.1) and have been in clinical use for more than two decades [1]. They scatter ultrasound much more effectively than red blood cells on account of the large contrast in density and compressibility between them and the surrounding blood. Most importantly, even at moderate ultrasound pressures, microbubbles undergo nonlinear volumetric oscillations in response to ultrasound excitation. Consequently, the scattered, or more correctly re-radiated signal, contains components other than those in the incident ultrasound beam. At typical
diagnostic pressures these are harmonics and/or fractional harmonics of the incident frequency. This allows the blood vessels carrying microbubbles to be differentiated from the surrounding tissue, which makes contrast enhanced ultrasound a very useful modality for imaging tissue perfusion [2].

For a given ultrasound field, the signal emitted will be different for different sizes of bubble, different coating materials and for different environments, for example changes in local viscosity, temperature or pressure. This means that microbubbles have the potential to be used for quantitative interrogation of the surrounding tissue (e.g. percentage perfusion or blood pressure), in addition to image contrast enhancement. To achieve this, however, accurate characterisation of the bubble is needed, in particular to determine the normally unknown shell properties.

This paper is concerned with the development of theoretical models for bubble characterisation under acoustic excitation, and proving that the bubble can be described by a unique set of shell properties.

![Figure 1.1](image.png)

*Figure 1.1* Diagram showing the different layers of a microbubble, the gas core has a radius typically between 0.5-8 µm and the shell thickness is between 1-100 nm.

### 2. Existing Models

#### 2.1. Radial model

Under ultrasound excitation, the spatially and temporally varying pressure in the surrounding medium causes microbubbles to oscillate volumetrically on account of their compressibility. Bubbles may also undergo translational motion due to the steady pressure gradient produced by the propagating wave. In a standing wave a bubble will travel to the nearest node or antinode depending on its size and the acoustic frequency.

Many equations neglect the translational motion and instead only describe the radial dynamics. These equations are usually derivatives of the Rayleigh-Plesset or, more correctly, the Rayleigh-Plesset-Noltingk-Neppiras-Poritsky (RPNNP) equation[3]:

\[
\ddot{R} + \frac{3}{2} \dot{R}^2 = \frac{1}{\rho_L} \left( p_0 + \frac{2\sigma}{R} - p_v \right) \left( \frac{R_0}{R} \right)^{3\gamma} + p_v - \frac{2\sigma}{R} - \frac{4\mu_L \dot{R}}{R} - p_0 + p_{ac}
\]

(2.1)

In equation (2.1) \( R \) is the bubble radius with initial stationary value \( R_0 \), \( \dot{R} \) is the velocity of the bubble wall, \( \ddot{R} \) is the acceleration of the bubble wall, \( p_i \) is vapour pressure, \( \gamma \) is the polytropic constant, \( \sigma \) is the surface tension, \( \mu_L \) is the viscosity of the liquid, \( p_{ac} \) is the pressure due to the acoustic wave, \( \rho_L \) is the density of the liquid and \( p_0 \) is the hydrostatic pressure of the liquid.

However the RPNNP equation only describes the motion of an uncoated bubble. Church [4] was the first author to publish a model for an ultrasound contrast agent with a finite thickness viscoelastic shell on the microbubble surface. Church’s model does not include any thermal or radiative dissipation effects, but the paper did indicate how they could be incorporated.

Hoff et al. [5] took the limit of Church’s model for an infinitely thin shell to obtain the governing equation.
that is, in fact, mathematically equivalent to the equation derived by de Jong et al.\[6-8\]. In this equation $G_s$ represents the shear modulus, $d_s$ the thickness of the shell, $\mu_s$ the viscosity of the shell, and $p_c(R) = p_0 \frac{(\frac{R_0}{R})^3}{R}$ is the pressure due to the gas inside the bubble.

Glazman [9] and Morgan et al. [10] built on previous studies of oceanic bubbles, to develop models that consider the vibrations of surfactant coated microbubbles, which treated the shell as an absorbed surface layer instead of a discrete shell. Although these treatments are essentially the same as [6], they use different functional relationships to describe the effective elasticity and viscosity of the shell. However there was an error in Glazman’s derivation in [9] which was propagated by Morgan but later corrected by Marmottant et al. [11] who produced a model that described the effect of the shell through an area dependent interfacial tension with a constant surface viscosity.

Stride [12] developed a model, equation (2.3), in which interfacial tension and viscosity are modelled as continuous functions of surface molecular concentration, $\Gamma$, obeying the experimentally derived functions of a power law and exponential relationship respectively. These variable terms replace the more conventional elastic terms seen in previous models and yield the following equation for an encapsulated bubble.

$$R\ddot{R} + \frac{3}{2} \dot{R}^2 = \frac{1}{\rho_L} \left( -p_0 + p_{ac} + p_G(R) - \frac{4\dot{R}}{R} \mu_L - \frac{12\mu_5 d_s R_0^2 \dot{R}}{R^4} - \frac{12G_5 d_s R_0^2}{R^3} \left( 1 - \frac{R_0}{R} \right) \right)$$  \hspace{1cm} (2.2)

Where $\Gamma_0$ is the initial molecular concentration, $\sigma_0$ is surface tension at equilibrium, $K$ is the Power law constant, $\gamma$ is the power law exponent, $\zeta$ the surface viscosity exponent and $\eta_{so} \epsilon^{(R^2-R_0^2)}$ is the surface viscosity constant.

2.2 Coupled Translational and Radial Models

Interest in bubble translational motion initially started with the investigation by King [13] into the effect of acoustic radiation pressure on a sphere in a standing wave. Watanabe and Kukita [14] solved the radial and translational equations simultaneously for an uncoated bubble of mass $m_b$ and volume $V(t)$, using the RPNNP for the radial displacement $R(t)$ alongside the following equation for the bubble’s translation from the origin $x(t)$: using the RPNNP for the radial and equation (2.4) for the translational motion.

$$\ddot{x} = \frac{1}{m_b} \left( -V \frac{dp_{ac}}{dx} - \frac{1}{2} \rho_L \frac{d}{dt} \left( V \frac{du_r}{dx} \right) - \frac{1}{2} \rho_L \frac{d}{dt} u_r A C_d \right)$$  \hspace{1cm} (2.4)

Where first term on the right hand side is the primary Bjerknes force, the second is the added mass and the third is the drag term. It is noteworthy that the governing equation for translational displacement is dependent on the relative velocity of the liquid $u_r = u_b - u_L$, where $u_b$ is the liquid velocity, leading to a drag term proportional to $A = \pi R^2$ (the so-called projected cross-sectional area of the bubble) and the following drag coefficient.
where $Re$ is the Reynolds number defined by

$$Re = \frac{2\rho_L u_r R}{\mu} \quad (2.6)$$

Doinikov [15] solved the radial and translational motion problem initially for an uncoated bubble using a Lagrangian approach to derive coupled equations.

$$R\ddot{R} + \frac{3}{2} R^2 = \frac{1}{\rho_L} \left( p_0 + \frac{2\sigma}{R_0^3} \left( \frac{R_0}{R} \right)^3 \right) - 2\frac{\sigma}{R} - p_0 + p_{ac} - \frac{4\mu_L \dot{R}}{R} + \frac{x^2}{4} \quad (2.7)$$

$$\ddot{x} + \frac{3R\dot{x}}{R} = \frac{3F_{ex}}{2\pi\rho_L R^3} \quad (2.8)$$

Where $F_{ex}$ represents the external forces (including the viscous drag and primary Bjerknes forces).

This derivation ignores the mass of the bubble and the relative velocity is treated inconsistently, being included in the viscous drag term but not in the kinetic energy of the liquid. A new term $\frac{x^2}{4}$ was however included in the radial equation which coupled equations (2.7) and (2.8) together. This term is also referred to as the slip velocity [16] and had been noted previously [15], but ignored because it is often negligibly small; however this is not always the case and should therefore be included for accuracy.

Later Doinikov and Dayton [17], re-derived the coupled equations using the same method as above, but for a shelled bubble using the Church equation (2.2) to describe the radial dynamics including the coupling term along with

$$\ddot{x} = \frac{1}{m_b + \frac{2}{3} \pi R_2^3 \rho_L} \left( - \frac{4\pi R_2^3}{3} \frac{dp_{ac}(x, t)}{dx} - 6\pi\mu_L R \dot{x} - 2\pi\rho_L \left( R_2^3 \dot{R} \dot{x} \right) \right) \quad (2.9)$$

to describe the translational motion, where $R_2$ is the outside radius of the bubble including the shell. We note here that equation (2.9) can be shown to be equivalent to that of equation (2.4) if the relative motion between the liquid and the bubble is ignored.

Finally Dayton et al. [18] subsequently published a model which also included some inconsistent terms in particular accounting for the added mass due to the oscillations of the bubble twice.

### 3. Derivation of Model

In reviewing the previous equations a number of inconsistencies were discovered. As above, these included the frame of reference used and the treatment of the relative velocity between the bubble and the liquid. In [15], the relative velocity is included in the drag force but not in the added mass or slip velocity in the radial term; whilst the model in [17] neglects the relative velocity of the bubble with respect to the liquid completely. Therefore a new version is derived in this section, following a discussion of the underlying assumptions.

#### 3.1 Assumptions

The assumptions made in the derivation of the model are as follows:

1. Spherical symmetry is maintained during microbubble oscillation.
2. External forces such as gravity and buoyancy can be ignored as they are very small in comparison to other forces experienced by the bubble.
3. There are no changes in the bubble properties during oscillation, i.e. equilibrium radius, shell and liquid properties remain constant (unless otherwise stated). This includes the assumption
that the bubble remains intact and there is negligible gas diffusion over the period of oscillation [19].

4. Thermal damping and radiation are taken to be negligible on account of the small amplitudes of oscillation being considered.

5. Due to the gas having very low density and viscosity compared with that of the surrounding liquid, the inertial and viscous contributions of the gas can be taken to be negligible.

6. The acoustic wavelength of the ultrasound is much larger than the radius of the bubble. Hence pressure variations across the bubble can be regarded as negligible.

7. Vapour inside the bubble is negligible; i.e. it is filled purely with gas. This is because the majority of microbubbles are manufactured under gas saturated conditions [20] and it is assumed that the shell would inhibit inwards diffusion after bubble formation.

8. The thermal diffusivity of the filling gas must remain constant during oscillations for the above assumptions to be consistent. Therefore it is also assumed to exhibit ideal and polytropic behaviour.

9. The surrounding fluid is taken to be infinite, initially static, homogeneous, incompressible and Newtonian.

3.2 Derivation

Following the approach adopted by Doinikov [15, 17] and starting from the Lagrangian equation

\[
\frac{d}{dt} \frac{dL}{dq_i} - \frac{dL}{dq_i} = -\frac{dF}{dq_i} \quad (3.1)
\]

Where \( L \) is the Lagrangian function defined by \( L = T - U \), \( T \) is the kinetic energy, \( U \) is the potential energy and \( q_i \) and \( \dot{q}_i \) are the generalised coordinates and velocities respectively. The \( F \) is the dissipative function which will take account of the dissipative effects of the liquid viscosity.

The total kinetic energy function in the frame of reference of the bubble centre is given by

\[
T = 2\pi \rho_L R^3 \left( \dot{R}^2 + \frac{u_r^2}{6} \right) \quad (3.2)
\]

and hence includes the relative velocity of the liquid, \( u_r = u_b - u_L \) where the bubble velocity \( u_b = \dot{x} \) and the liquid velocity \( u_L \), can be described by

\[
u_L = \frac{p_a}{\rho_L c_s} \cos(\omega t) \cos(kx) \quad (3.3)
\]

with the angular frequency \( \omega = 2\pi f \), the wavenumber \( k = \frac{2\pi f}{c_s} \), where \( p_a \) is the pressure amplitude, \( c_s \) is the speed of sound and \( f \) is the frequency of the acoustic wave. It should be noted that equation (3.2) does not include the kinetic energy of the bubble as in [17]. This is because in the frame of reference used this term does not exist.

The total potential energy is given by the sum of the potential energy of the gas within the bubble, the potential energy of the surface tension and the potential energy of the external pressure, leading to the following expression given the initial volume of the bubble \( V_0 \)

\[
U = \frac{p_0 V_0}{\gamma - 1} \left( \frac{R_0}{R} \right)^{3(\gamma - 1)} + 4\pi R^2 \sigma + \frac{4\pi}{3} R^3 [p_0 + p_{ac}] \quad (3.4)
\]

Equation (3.4) does not contain the viscous forces and the dissipative function of the liquid is of the form
\[ F = \int_{V_L} f_L dV \quad (3.5) \]

where \( V_L \) is the volume occupied by the liquid, and \( f_L \) is the density of the dissipative function. Defining \( \xi_L \) as the bulk viscosity coefficient and \( v_{ij} \) as the rate-of-strain tensor of the liquid, the integrand of equation (3.5) can be written

\[ f_L = \mu_L \left( v_{ij} - \frac{1}{2} \delta_{ij} v_{kk} \right)^2 + \frac{1}{2} \xi_L v_{kk}^2 \quad (3.6) \]

Moreover, since the liquid is incompressible, \( v_{kk} = \Delta v = 0 \), equation (3.6) becomes

\[ f_L = \mu_L (v_{ij})^2 \quad (3.7) \]

and by integrating \((v_{ij})^2\) over \( V_L \), the volume of the liquid, it is found that

\[ F_L = 8\pi \mu_L R \dot{R}^2 + 3\pi \mu_L R u_r^2 \quad (3.8) \]

Substituting equations (3.2),( 3.4) and( 3.8) into equation (3.1) for generalised coordinate \( q = R \) and \( x \) respectively then yields

\[ R \ddot{R} + \frac{3}{2} \dot{R}^2 = \frac{1}{\rho_L} \left( \frac{p_0 + 2\sigma + p_{ac}}{R} \right)^3 \left( \frac{R}{R} \right)^3 - \frac{2\sigma}{R} - p_0 - p_{ac} - \frac{4\mu_L \ddot{R}}{R} \right) + \frac{u_r^2}{4} \quad (3.9) \]

\[ \ddot{x} = \frac{1}{\left( \frac{2}{3} \pi \rho_L R^3 \right)} \left( -2\pi \rho_L R^2 u_r + \frac{2}{3} \pi \rho_L R^3 \dot{u}_L - \frac{4}{3} \pi R^3 \frac{dp_{ac}(x, t)}{dx} - 6\pi \mu_L R u_r \right. \\
\left. + \frac{\pi \rho L R^3}{3} \frac{d u_r^2}{dx} \right) \quad (3.10) \]

As can be seen this gives the RPNNP equation with the slip velocity included and an equation for the translational motion that is the same as that derived in[15] apart from the relative motion being used to describe the velocity of the bubble in the flow as well as the inclusion of the final \(\frac{d^2 u_r^2}{dx^2}\) term. [17] would also produce the same equation if the relative velocity were treated correctly and the frame of reference consistently applied.

In most applications, the \(\frac{d^2 u_r^2}{dx^2}\) term in equation(3.10) is negligibly small and makes no difference to the bubble’s behaviour. For equation (3.10) it should also be noted that the derivation assumes Stokes’ law as the drag term, which only holds for Reynolds number \( Re \ll 1 \).

The model needs to include a shell for the purposes of characterising a coated bubble. It was shown by Doinikov and Dayton [17] that the translational term and coupling term are unaltered by the presence of a shell as described by Church’s model[4]. As Hoff’s model [5] represents the thin shell limit of Church’s equation, the translational equation above can still be used, yielding the following equation for the radial dynamics

\[ R \ddot{R} + 3 \dot{R}^2 = \frac{1}{\rho_L} \left( p_0 + p_{ac} + p_G(R) - \frac{4R}{R} \dot{u}_L - \frac{12\mu_s d s R_0^2 \ddot{R}}{R^4} - \frac{12G_s d s R_0^2}{R^3} \left( 1 - \frac{R_0}{R} \right) \right) + \frac{u_r^2}{4} \quad (3.11) \]

Equation (3.11) was verified against the analytical solution of the Hoff model [5]. In figure 3.1 it is shown that difference between using the slip velocity and neglecting it with an example of when this term starts to become important. This graph was produced in MatLabRb2011a using ode45 with the
values in Table 1 and run for 50 acoustic cycles. The shell parameters were based on those of SonoVue found in [21].

Table 1. Showing the parameters used to produce the figure 3.1.

| Parameter | Value |
|-----------|-------|
| $R_0$     | 1 $\mu$m |
| $p_0$     | 100kPa |
| $\mu_s$  | 2.5Pa.s |
| $\mu_L$  | 8.9x10^{-3}Pa.s |
| $\Sigma$ | 7.2x10^{-3}N.m^{-1} |
| $\rho_L$ | 998 kg.m^{-3} |
| $G_s$     | 122x10^6 Pa |
| $d_s$     | 1.5nm |
| $c_s$     | 1500m.s^{-1} |
| $\rho_g$  | 1.83kg m^{-3} |
| $\Gamma$ | 1.4 |

Figure 3.1 Comparison between using the set of equations (3.10) and (3.11) represented by the blue dotted line, against equation (2.2) and (3.10) represented by the red line which neglects the coupling term in the radial equation. Where (a) is the radial, (c) is the translational motion of the bubble with $p_{ac}=10$kPa and frequency 0.7MHz, (b) is the radial and (d) the translational motion of the bubble with $p_{ac}=1.1$MPa and frequency 4.9MHz.
4. Uniqueness of the Equation

In order for characterisation of the shell to be useful it is essential that a unique set of shell parameters can be determined through fitting experimental data to a theoretical model. Assume for instance that the variation in radius \( R(t) \) of an encapsulated bubble driven by a known external ultrasound pressure field \( p_{ac}(t) \) can be accurately measured during an experiment. If everything about the bubble and the surrounding medium is known aside from the properties of the shell then one can assume the behaviour of \( R(t) \) to be governed by a model such as that given in (2.2) and then attempt to find values of the shell’s shear modulus \( G_s \), the shell thickness, \( d_s \), and the shell viscosity, \( \mu_s \), that fit the experimental \( R(t) \) profile accurately. However, the question remains over whether such parameter values are unique for the bubble that was measured experimentally.

To answer the uniqueness question, equation (2.2) can be written in the following way

\[
\rho_L \left( R \ddot{R} + \frac{3}{2} \dot{R}^2 \right) + (-p_0(R) + p_0 - p) + \frac{4}{R} \mu_L \left( -\frac{R}{R} u_f^2 \right) = -\frac{12 \mu_s d_s R_s^2 \dot{R}}{R^4} - \frac{12 G_s d_s^2 R_s^2}{R^3} \left( 1 - \frac{R_0}{R} \right)
\]

(4.1)

where only terms containing the unknown shell parameters are retained on the right-hand side. The three parameters \( \mu_s, d_s, G_s \) can then be reduced to just two unknowns, \( G_s d_s = v \) and \( \mu_s d_s = u \) as they only appear as these two products, noting that \( d_s \) can be measured independently. For this proof it shall be assumed that these parameter values are constant over an oscillation and their uniqueness will be proved by contradiction. Hence, we initially assume that values \( v \) and \( u \) are not unique and thus there must exist at least two sets of values \( v_1, u_1 \) and \( v_2, u_2 \) that, when substituted into (4.1), identically replicate the experimental measurement of \( R(t) \). Redefining the left hand side of equation (4.1) to be equal to a function \( F(\ddot{R}, \dot{R}, R) \), it must therefore be the case that

\[
F(\ddot{R}, \dot{R}, R) = -\frac{12 R_0^2 \ddot{R}}{R^5} u_1 - \frac{12 R_0^2}{R^4} \left( 1 - \frac{R_0}{R} \right) v_1
\]

(4.2a)

\[
F(\ddot{R}, \dot{R}, R) = -\frac{12 R_0^2 \ddot{R}}{R^5} u_2 - \frac{12 R_0^2}{R^4} \left( 1 - \frac{R_0}{R} \right) v_2
\]

(4.2b)

Eliminating \( F(\ddot{R}, \dot{R}, R) \) from these equations and rearranging then yields the ordinary first-order differential equation for \( R(t) \),

\[
\dot{R} + \frac{R(v_1 - v_2)}{(u_1 - u_2)} = \frac{R_0(v_1 - v_2)}{(u_1 - u_2)}
\]

(4.3)

Letting \( \frac{(v_1 - v_2)}{(u_1 - u_2)} = \beta \) and solving subsequently reveals the general solution to be

\[
R(t) = R_0 + \frac{C}{\beta} e^{-\beta t}
\]

(4.4)

where \( C \) is an unknown constant, which can be set by the initial condition. Equation (4.4) can be interpreted in the following way: if the varying bubble radius \( R(t) \) in the experiment does not exhibit exponential decay to the value \( R_0 \) then it must be the case that \( u_1 = u_2 \) and \( v_1 = v_2 \). As a consequence, any data-fitted parameter values must be unique (so long as \( d_s \) can be measured separately).
Following similar logic it can also be proven that if values of parameters $\eta_{so}$ and $K\Gamma_0$ are fitted to experimental data of $R(t)$ in equation (2.3) with power-law exponent $\gamma=0$ and surface viscosity exponent $\varepsilon=0$, then those fitted values are unique. In this case the governing equation is given by:

$$\rho_l \left( R \dddot{R} + \frac{3}{2} \dot{R}^2 \right) + p_0 - p_a - p_0(R) + \frac{4}{R} \mu \frac{4\dot{R}}{R^3} \eta_{so} = -\frac{4\dot{R}}{R^3} \eta_{so} - K\Gamma_0 \frac{3}{R^2} \left( 1 - \frac{R_0^2}{R^2} \right)$$

(4.5)

Hence, as before by taking $\alpha_1 = \eta_{so}$, $\beta_1 = K\Gamma_0$ and $\alpha_2 = \eta_{so}$, $\beta_2 = K\Gamma_0$ as two sets of parameter values that accurately fit the experimental data, these two sets can be substituted into (4.5) and, on eliminating the common left-hand side of the equation in each case, yields

$$R \frac{dR}{dt} + \Omega R^2 = \Omega R_0^2$$

for $\Omega = \frac{3(\beta_1 - \beta_2)}{4(\alpha_1 - \alpha_2)}$. The general solution to this differential equation is then $R(t) = \sqrt{R_0^2 + 2Ce^{-2\Omega t}}$ and hence unless $R_2^2(t)$ exponentially decays in this manner to $R_0^2$ in the experimental data then the fitted parameter values must be unique.

5. Conclusions

Previous descriptions of the coupled radial and translational motion of a bubble have contained inconsistent treatments of the relative velocity of the bubble and been inconsistent with the frames of reference used. A derivation to include these has been presented here and these effects have been shown to be non-negligible, particularly at high pressures.

It has been proven that a unique set of parameters describing a microbubble coating can be derived by fitting experimental data to the theoretical model. This will enable it to be used effectively for bubble characterisation.

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