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Viscous streaming from surface waves on the wall of acoustically-driven gas bubbles

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Abstract

Microstreaming which is induced when a single bubble is driven acoustically in the regime of parametric generation of Faraday waves is studied. The greater wall displacement amplitudes for n > 1 modes mean that their effect on the flux of species in the liquid can be much greater than that of the breathing mode. It has been shown that the acoustical streaming from a bubble undergoing axi-symmetric surface wave oscillations is enhanced in comparison with lateral oscillations. This study was initiated by the observations of bubble oscillation by electrochemical sensing technique. Now we can quantitatively evaluate the magnitude of the measured currents and its relation to the optically detected amplitudes of bubble oscillations. © 2006 Elsevier Masson SAS. All rights reserved.

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1. Introduction

This study deals with the microstreaming which is induced when a single bubble is driven acoustically in the regime of parametric generation of Faraday waves. Unlike the monopole breathing mode, the higher order surface waves generate little far-field acoustic emission. However the greater wall displacement amplitudes for n > 1 modes mean that their effect on the flux of species in the liquid can be much greater than that of the breathing mode. This large enhancement to mass transfer above the threshold of generation of Faraday waves has been detected and investigated in the experiments [1–3].

The study of mass transfer in water waves has a long history. It was shown by Stokes in a classical memoir [4] that in an irrotational water wave the fluid particles posses, in addition to their orbital motion, a steady second-order drift velocity. Longuet-Higgins [5] provided a general theory for this, taking into account viscosity. The bubble (cavitation) microstreaming was first observed by Elder [6], who noted steady streaming in the neighbourhood of a small bubble attached to a vibrating piston. Elder found that the microstreaming is most pronounced for bubbles undergoing breathing mode (pulsation) resonance. In some of Elder's experiments, surface modes were excited at the bubble surface. These significantly changed the streaming pattern. A consistent theoretical investigation of the streaming in the neighbourhood of the bubble which performs translational harmonic oscillations has been done by Davidson and Riley [7].

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Axi-symmetric patterns of streaming around a single sonoluminescing bubble have been observed by Lepoint et al. [8], a situation theoretically described by Longuet-Higgins [9] who found analytically the drift velocities induced by a spherical bubble undergoing small radial and transverse oscillations.

The experiments of Marmottant and Hilgenfeldt [10,11], in which a microbubble fixed on substrate performs gentle linear oscillations that were sufficient to rupture lipid membranes, makes acoustics on the micrometer scale a tool for cell-wall permeation. Bubble induced microstreaming has been proposed as a possible mechanism for membrane permeation.

This study undertakes an extension of the methods of Davidson and Riley [7] and Longuet-Higgins [9] to include surface modes of order n > 1. A complete solution of the problem would seem to required a lengthy time integration of the full Navier–Stokes equations. We restrict our analysis by accounting for axi-symmetric modes only. In this way we can introduce stream function and thus use the advantages of the approach of Longuet-Higgins [9]. This provides the knowledge of how electrochemical current in experiments [1–3] relates to convective flow.

2. Definition of the problem

For a liquid of kinematic viscosity ν , there are five important length scales in this problem: the bubble equilibrium radius R_0 , the displacement amplitude of the breathing (monopole) mode ξ_0 , the amplitude of the parametrically generated distortion mode ξ_n , the wavelength of the surface mode $(2\pi R_0/n)$ and the viscous length $\delta = (2\nu/\omega)^{1/2}$ (noting that this is the viscous length corresponding to the frequency $(\omega/2)$ of the parametrically generated distortion mode). From these scales we can construct four independent parameters $\varepsilon_1 = (\xi_0/R_0)$, $\varepsilon_2 = (\xi_n/R_0)$, $\beta = (\delta/R_0)$ and (1/n). We shall be concerned entirely with the situation $\varepsilon_1 \ll 1$, $\varepsilon_2 \ll 1$, $\beta \ll 1$, $1 \ll n$.

In discussed experiments [1–3] the images of surface waves are clearly seen on the bubble interface. For the bubble with equilibrium radius $R_0 \sim 2.5$ mm, driven into oscillation by acoustical field with the frequency $f = (\omega/2\pi) \sim 1.3$ kHz, the image indicates the distortion mode (n = 15) excited with amplitude $\xi_n \sim 0.1$ mm and the wavelength $(2\pi R_0/n) \sim 1$ mm. The breathing mode is of course also excited, but cannot be seen in the image. The thickness of the oscillatory shear layer (Stokes layer) near the surface of the bubble $\delta = \sqrt{2\nu/(2\pi f)} \approx 1.6 \times 10^{-2}$ mm (for an assumed water viscosity of $\nu = 0.01$ cm²/s).

Consider a clean uncoated bubble of radius R_0 driven acoustically in an unbounded fluid of uniform density ρ_0 . We shall use the radial coordinates $(r, \vartheta, \bar{\alpha})$ and assume the flow to be axi-symmetric. At any point in the liquid (described in this axi-symmetric case by radial distance r and the azimuth angle ϑ), we can write the equation of the bubble surface as $r = R_0 + \xi(\vartheta, t)$.

The nonlinear response of the gas bubble to a pumping wave with frequency ω results in parametrically-generated shape oscillations above a well-defined threshold. According to [12], the increment of instability of the *n*th distortion mode $(n \gg 1)$ has the following form

$$\lambda = -\gamma_n + \sqrt{\left(\frac{P_m}{2\omega_0 \rho_0 R_0}\right)^2 \frac{Q_n^2}{[(\omega_0 - \omega)^2 + \gamma_0^2]} - \left(\sigma_n - \frac{\omega}{2}\right)^2},$$
(2.1)

where $\omega_0 = \sqrt{3\gamma p_0/\rho_0 R_0^2}$ is the frequency of the fundamental (breathing) mode; γ is the polytropic exponent, ρ_0 and p_0 are the equilibrium density and pressure; the bubble is driven by a pumping wave of amplitude P_m and angular frequency ω ; $\sigma_n = \sqrt{(\sigma/\rho_0 R_0^3)(n-1)(n+1)(n+2)}$ is the natural frequency of the nth distortion mode; γ_0 is the sum of radiation damping, viscous damping and damping due to thermal diffusion for the breathing mode; γ_n is the viscous damping of the nth distortion mode, as estimated by a linear analysis; $Q_n = (4n-1)\sigma_n[64\pi(n+1)R_0]^{-1}$ is the coefficient of parametric coupling between the distortion and breathing modes. The condition under which λ vanishes determines the threshold of instability $P_m = P_{th}(\omega, n)$ for the distortion modes:

$$P_{th}^{2}(\omega, n) = \left(\frac{2\omega_{0}\rho_{0}R_{0}}{Q_{n}}\right)^{2} \left[\gamma_{n}^{2} + \left(\sigma_{n} - \frac{\omega}{2}\right)^{2}\right] \left[(\omega_{0} - \omega)^{2} + \gamma_{0}^{2}\right]. \tag{2.2}$$

For the millimeter sized bubbles and the kilohertz frequency range corresponding with the conditions of the experiments [1–3], the parametric resonance $\omega_0(R_0) \approx 2\sigma_n(R_0)$ between breathing and distortion modes can be satisfied

assuming n = 11-15 modes excitation. The minimum driving acoustic pressure amplitude (2.2) which will excite a mode of order n is low in these conditions (circa 30–50 Pa).

The bubble was restricted against buoyant rise by the glass rod placed above it in the experiments [1–3]. The presence of the rod will be accounted for only by excluding the dipole mode of bubble oscillations, so we suppose the centre of the sphere remains fixed and only the breathing (monopole) and distortion (n > 1) modes can be excited on the bubble wall. Movie images (available at http://www.isvr.soton.ac.uk/fdag/Echem_ Faraday.htm) indicate that whilst this is true for the low driving amplitudes, at higher amplitudes translation may take place.

3. Equations and boundary conditions

From the equation of continuity in terms of the liquid velocity $\mathbf{u} = u_r \mathbf{e}_r + u_{\vartheta} \mathbf{e}_{\vartheta} + u_{\alpha} \mathbf{e}_{\alpha}$

$$\operatorname{div}(\mathbf{u}) = 0, \qquad \frac{1}{r^2} \frac{\partial}{\partial r} (r^2 u_r) + \frac{1}{r \sin \vartheta} \frac{\partial}{\partial \vartheta} (\sin \vartheta u_\vartheta) = 0, \tag{3.1}$$

we can introduce an axi-symmetric stream function ψ , in terms of which the components of radial and tangential velocity are given by

$$u_r = \frac{1}{r^2 \sin \vartheta} \frac{\partial \psi}{\partial \vartheta} = -\frac{1}{r^2} \frac{\partial \psi}{\partial \mu}, \quad u_\vartheta = -\frac{1}{r \sin \vartheta} \frac{\partial \psi}{\partial r} = -\frac{1}{r(1-\mu^2)^{1/2}} \frac{\partial \psi}{\partial r}, \quad \mu = \cos \vartheta. \tag{3.2}$$

The Navier-Stokes equation in radial coordinates takes the form

$$\frac{\partial u_r}{\partial t} + \left(u_r \frac{\partial}{\partial r} + \frac{u_{\vartheta}}{r} \frac{\partial}{\partial \vartheta}\right) u_r - \frac{u_{\vartheta}^2}{r} = -\frac{1}{\rho_0} \frac{\partial p}{\partial r} + \nu \left[\Delta u_r - \frac{2u_r}{r^2} - \frac{2}{r^2 \sin^2 \vartheta} \frac{\partial (u_{\vartheta} \sin \vartheta)}{\partial \vartheta}\right], \\
\frac{\partial u_{\vartheta}}{\partial t} + \left(u_r \frac{\partial}{\partial r} + \frac{u_{\vartheta}}{r} \frac{\partial}{\partial \vartheta}\right) u_{\vartheta} + \frac{u_r u_{\vartheta}}{r} = -\frac{1}{\rho_0 r} \frac{\partial p}{\partial \vartheta} + \nu \left[\Delta u_{\vartheta} - \frac{u_{\vartheta}}{r^2 \sin^2 \vartheta} + \frac{2}{r^2} \frac{\partial u_r}{\partial \vartheta}\right].$$
(3.3)

Then if $\overline{\omega}$ denotes the vorticity

$$\overline{\mathbf{\omega}} = \left[\nabla \times \mathbf{u}\right] = \frac{1}{r \sin \vartheta} \left[\frac{\partial}{\partial \vartheta} \left(u_{\alpha} \sin \vartheta \right) - \frac{\partial u_{\vartheta}}{\partial \bar{\alpha}} \right] \mathbf{e}_{r} + \frac{1}{r} \left[\frac{1}{\sin \vartheta} \frac{\partial u_{r}}{\partial \bar{\alpha}} - \frac{\partial (r u_{\alpha})}{\partial r} \right] \mathbf{e}_{\vartheta} + \frac{1}{r} \left[\frac{\partial (r u_{\vartheta})}{\partial r} - \frac{\partial u_{r}}{\partial \vartheta} \right] \mathbf{e}_{\alpha} = \frac{1}{r} \left[\frac{\partial (r u_{\vartheta})}{\partial r} - \frac{\partial u_{r}}{\partial \vartheta} \right] \mathbf{e}_{\alpha}, \tag{3.4}$$

it is convenient (following to Longuet-Higgins [9]), to define the 'total' vorticity $\Omega = \bar{\omega}_{\alpha} r \sin \vartheta$ which is proportional to the line integral of $\bar{\omega}_{\alpha}$ around a circle of latitude. From (A3.4) and (A3.2) we find

$$\Omega = \sin \vartheta \left[\frac{\partial (r u_{\vartheta})}{\partial r} - \frac{\partial u_r}{\partial \vartheta} \right] = -\frac{\partial^2 \psi}{\partial r^2} - \frac{1 - \mu^2}{r^2} \frac{\partial^2 \psi}{\partial \mu^2} = -D^2 \psi, \tag{3.5}$$

where D^2 denotes the linear operator $D^2 \equiv \partial^2/\partial r^2 + (1-\mu^2)r^{-2}\partial^2/\partial \mu^2$. The vorticity equation then becomes

$$\frac{\partial \Omega}{\partial t} + \frac{1}{r^2} \left[\frac{\partial (\psi, \Omega)}{\partial (r, \mu)} + 2\Omega \Lambda \psi \right] = \nu D^2 \Omega, \tag{3.6}$$

where

$$\frac{\partial(\psi,\Omega)}{\partial(r,\mu)} \equiv \frac{\partial\psi}{\partial r}\frac{\partial\Omega}{\partial\mu} - \frac{\partial\psi}{\partial\mu}\frac{\partial\Omega}{\partial r}, \qquad \Lambda \equiv \frac{\mu}{1-\mu^2}\frac{\partial}{\partial r} + \frac{1}{r}\frac{\partial}{\partial\mu}$$

(see [9])

The main equations (3.3) and (3.6) should be supplied with dynamical and kinematic boundary conditions. The kinematic boundary condition is that, on the distorted sphere, the normal velocity should be continuous

$$\left[\frac{\partial}{\partial t} + (\mathbf{u}, \nabla)\right] (r - R)_{r = R_0 + \xi} = u_r - \dot{\xi} - \frac{u_{\vartheta}}{r} \frac{\partial \xi}{\partial \vartheta} \bigg|_{r = R_0 + \xi} = 0$$
(3.7a)

which in terms of the stream function has the form

$$\dot{\xi} = u_r - \frac{u_{\vartheta}}{R_0 + \xi} \frac{\partial \xi}{\partial \vartheta} = -\frac{1}{(R_0 + \xi)^2} \left(\frac{\partial \psi}{\partial \mu} + \frac{\partial \xi}{\partial \mu} \frac{\partial \psi}{\partial r} \right) \bigg|_{r = R_0 + \xi}.$$
(3.7b)

The dynamical conditions are the balance of the normal and tangential components of stress: $\sigma_{ik} = -p\delta_{ik} + \eta_d(\partial u_i/\partial x_k + \partial u_k/\partial x_i)$. The difference in the normal component of stress in liquid σ_{nn}^l and gas σ_{nn}^g is equal to surface tension

$$\sigma_{nn}^{l} = \sigma_{nn}^{g} + \sigma(\nabla, \mathbf{n}), \tag{3.8}$$

where $\sigma_{nn}^l = n_i \sigma_{ik}^l n_k = -p + 2\eta_d n_i(\mathbf{n}, \nabla) u_i$, p – is the pressure in liquid, σ is the coefficient of surface tension, η_d is the liquid dynamical viscosity, and \mathbf{n} is the unit vector normal to the surface $r = R_0 + \xi(\vartheta, t)$, such that

$$\mathbf{n} = \left\{ \mathbf{e}_r - (R_0 + \xi)^{-1} \left(\frac{\partial \xi}{\partial \vartheta} \right) \mathbf{e}_{\vartheta} \right\} \left(1 + (R_0 + \xi)^{-2} \left(\frac{\partial \xi}{\partial \vartheta} \right)^2 \right)^{-1/2}.$$

We shall ignore gas viscosity so that $\sigma_{nn}^g = n_i \sigma_{ik}^g n_k = -p_g$, where p_g is the pressure in the bubble, and we adopt a polytropic law

$$p_g = p_0 \left(\frac{V_0}{V}\right)^{\gamma}, \quad V = \frac{2\pi}{3} \int_{-1}^{1} d\mu \left[R_0 + \xi(\mu)\right]^3,$$

where V, V_0 are the instantaneous and equilibrium bubble volume. Thus the first dynamical boundary condition takes the form

$$p - 2\eta_d n_i(\mathbf{n}, \nabla) u_i = p_0 \left(\frac{V_0}{V}\right)^{\gamma} - \sigma(\nabla, \mathbf{n}). \tag{3.9a}$$

The explicit form of the normal stress in the radial coordinates is

$$\sigma_{nn}^{l} = \left[1 + \frac{1}{r^{2}} \left(\frac{\partial \xi}{\partial \vartheta}\right)^{2}\right]^{-1} \left\{\sigma_{rr} - \frac{1}{r} \left(\frac{\partial \xi}{\partial \vartheta}\right) (\sigma_{r\vartheta} + \sigma_{\vartheta r}) + \frac{1}{r^{2}} \left(\frac{\partial \xi}{\partial \vartheta}\right)^{2} \sigma_{\vartheta\vartheta}\right\} \bigg|_{r=R_{0}+\xi}, \tag{3.9b}$$

where

$$\sigma_{rr} = -p + 2\eta_d \left(\frac{\partial u_r}{\partial r} \right), \qquad \sigma_{r\vartheta} = \sigma_{\vartheta r} = \eta_d \left[r^{-1} \frac{\partial u_r}{\partial \vartheta} + \frac{\partial u_{\vartheta}}{\partial r} - r^{-1} u_{\vartheta} \right]$$

and

$$\sigma_{\vartheta\vartheta} = -p + 2\eta_d r^{-1} \left(\frac{\partial u_{\vartheta}}{\partial \vartheta} + u_r \right)$$

[13]. Substituting these expressions into (3.9b) gives:

$$\begin{split} \sigma_{nn}^{l} &= -p + 2\eta_{d} \left\{ \frac{\partial u_{r}}{\partial r} - \frac{1}{r} \left(\frac{\partial \xi}{\partial \vartheta} \right) \left[\frac{\partial u_{\vartheta}}{\partial r} + \frac{1}{r} \frac{\partial u_{r}}{\partial \vartheta} - \frac{u_{\vartheta}}{r^{2}} \right] + \frac{1}{r^{2}} \left(\frac{\partial \xi}{\partial \vartheta} \right)^{2} \\ &\times \left[\frac{1}{r} \frac{\partial u_{\vartheta}}{\partial \vartheta} + \frac{u_{r}}{r} \right] \right\}_{r=R_{0}+\xi} \left[1 + \frac{1}{(R_{0}+\xi)^{2}} \left(\frac{\partial \xi}{\partial \vartheta} \right)^{2} \right]^{-1}. \end{split}$$

Finally, expressed in terms of the stream function, the condition (3.9a) takes the form:

$$p - 2\eta_{d} \left[(R_{0} + \xi)^{2} + (1 - \mu^{2}) \left(\frac{\partial \xi}{\partial \mu} \right)^{2} \right]^{-1} \left\{ \left[-\frac{\partial^{2} \psi}{\partial r \partial \mu} + \frac{2}{R_{0} + \xi} \frac{\partial \psi}{\partial \mu} \right] + \left(\frac{\partial \xi}{\partial \mu} \right) \left[-\frac{\partial^{2} \psi}{\partial r^{2}} + \frac{1 - \mu^{2}}{(R_{0} + \xi)^{2}} \frac{\partial^{2} \psi}{\partial \mu^{2}} \right] \right\} + \frac{2}{r} \frac{\partial \psi}{\partial r} + \frac{1}{(R_{0} + \xi)^{2}} \left(\frac{\partial \xi}{\partial \mu} \right)^{2} \left[(1 - \mu^{2}) \frac{\partial^{2} \psi}{\partial r \partial \mu} + \mu \frac{\partial \psi}{\partial r} - \frac{\sqrt{1 - \mu^{2}}}{R_{0} + \xi} \frac{\partial \psi}{\partial \mu} \right] \right\}_{r=R_{0} + \xi}$$

$$= p_{0} \left(\frac{V_{0}}{V} \right)^{\gamma} - \sigma(\nabla, \mathbf{n}). \tag{3.10}$$

The dynamical boundary condition that the tangential stress $\sigma_{\tau n}^l = \tau_i \sigma_{ik}^l n_k$ vanishes at $r = R_0 + \xi$ leads to

$$\sigma_{\tau n}^{l} = \eta_{d} \left[1 + \frac{1}{(R_{0} + \xi)^{2}} \left(\frac{\partial \xi}{\partial \vartheta} \right)^{2} \right]^{-1} \left\{ \frac{1}{r} \left(\frac{\partial \xi}{\partial \vartheta} \right) (\sigma_{rr} - \sigma_{\vartheta\vartheta}) + \left[1 - \frac{1}{r^{2}} \left(\frac{\partial \xi}{\partial \vartheta} \right)^{2} \right] \sigma_{r\vartheta} \right\}_{r=R_{0} + \xi} = 0,$$

where $\tau = \{(R_0 + \xi)^{-1}(\partial \xi/\partial \vartheta)\mathbf{e}_r + \mathbf{e}_{\vartheta}\}[1 + (R_0 + \xi)^{-2}(\partial \xi/\partial \vartheta)^2]^{-1/2}$, $(\tau, \mathbf{n}) = 0$. On substituting explicit expressions for the components of stress in the radial coordinates we get

$$\sigma_{\tau n}^{l} = \eta_{d} \left[1 + \frac{1}{(R_{0} + \xi)^{2}} \left(\frac{\partial \xi}{\partial \vartheta} \right)^{2} \right]^{-1} \left\{ \left[\frac{1}{r} \frac{\partial u_{r}}{\partial \vartheta} + \frac{\partial u_{\vartheta}}{\partial r} - \frac{u_{r}}{r} \right] + \frac{2}{r} \left(\frac{\partial \xi}{\partial \vartheta} \right) \left[\frac{\partial u_{r}}{\partial r} - \frac{1}{r} \frac{\partial u_{\vartheta}}{\partial \vartheta} - \frac{u_{r}}{r} \right] \right\} - \frac{1}{r^{2}} \left(\frac{\partial \xi}{\partial \vartheta} \right)^{2} \left[\frac{1}{r} \frac{\partial u_{r}}{\partial \vartheta} + \frac{\partial u_{\vartheta}}{\partial r} - \frac{u_{r}}{r} \right] \right\}_{r=R_{0}+\xi} = 0.$$
(3.11)

Expressed in terms of the stream function (3.11) takes the form:

$$\left[1 - \frac{1 - \mu^2}{(R_0 + \xi)^2} \left(\frac{\partial \xi}{\partial \mu}\right)^2\right] \left[\frac{\partial^2 \psi}{\partial r^2} - \frac{1 - \mu^2}{(R_0 + \xi)^2} \frac{\partial^2 \psi}{\partial \mu^2} - \frac{2}{R_0 + \xi} \frac{\partial \psi}{\partial r}\right]_{r=R_0 + \xi} + \frac{2}{(R_0 + \xi)^2} \frac{\partial \xi}{\partial \mu} \left[-2(1 - \mu^2) \frac{\partial^2 \psi}{\partial r \partial \mu} - \mu \frac{\partial \psi}{\partial r} + \frac{3(1 - \mu^2)}{R_0 + \xi} \frac{\partial \psi}{\partial \mu}\right]_{r=R_0 + \xi} = 0.$$
(3.12)

Thus we obtain the governing equations in the forms which account for the symmetry of the problem, and which are suitable for finding approximate solutions.

4. Perturbation solution. The first approximation

We seek a perturbation solution of Eqs. (3.6) and (3.3) subject to conditions (3.7), (3.10), (3.12) in the form

$$\psi(r,\mu,t,\varepsilon_{1},\varepsilon_{2},\beta,n) = \varepsilon_{1}\psi_{10}(r,\mu,t,\beta,n) + \varepsilon_{2}\psi_{01}(r,\mu,t,\beta,n) + \varepsilon_{1}^{2}\psi_{20}(r,\mu,t,\beta,n) + \varepsilon_{2}^{2}\psi_{02}(r,\mu,t,\beta,n) + \varepsilon_{1}\varepsilon_{2}\psi_{11}(r,\mu,t,\beta,n) + \cdots$$

$$(4.1)$$

Since, from general consideration, we expect that $\psi_{10}(r, \mu, t, \beta, n)$ describes the flow induced by the breathing mode and $\psi_{01}(r, \mu, t, \beta, n)$ describes that which is generated by the surface mode of order n which corresponds to the Faraday wave, then

$$\psi_{10}(r,\mu,t,\beta,n) = \psi_{10}(r,\mu,\beta,n) \exp(-\mathrm{i}\omega t), \qquad \psi_{01}(r,\mu,t,\beta,n) = \psi_{01}(r,\mu,\beta,n) \exp\left(-\frac{\mathrm{i}\omega t}{2}\right).$$

If we substitute expansion (4.1) into Eq. (3.6) and retain only terms of order ε_2 , we obtain for the stream function of the distortion mode

$$-i\left(\frac{\omega}{2\nu}\right)\Omega_{01} = D^2\Omega_{01} \quad \left(\Omega_{01} \equiv -D^2\psi_{01}\right). \tag{4.2}$$

The same procedure for Navier-Stokes equations, giving for example the radial component:

$$\frac{1}{r^2} \frac{\partial}{\partial \mu} \left[-i \left(\frac{\omega}{2} \right) \psi_{01} - \nu \left(\frac{\partial^2 \psi_{01}}{\partial r^2} + \frac{1 - \mu^2}{r^2} \frac{\partial^2 \psi_{01}}{\partial \mu^2} \right) \right] = \frac{1}{\rho_0} \frac{\partial p_{01}}{\partial r},\tag{4.3}$$

and thus determining the pressure p_{01} in terms of the stream function ψ_{01} .

There are now two ways to approximate the boundary conditions. Either we may take radial coordinates (r, ϑ) fixed in space, in which case the boundary conditions (3.7), (3.10), (3.12) have to be satisfied on a variable surface. This approach seems at first to restrict us to the normal displacements that are small compared to the boundary-layer thickness, or use introduced dimensionless parameters: $\varepsilon_1 \ll \beta$, $\varepsilon_2 \ll \beta$. Alternatively we may choose moving coordinates. Following Longuet-Higgins [5] a coordinate system may be chosen attached to the moving boundary, where s_a is the arc measured along the boundary, and \bar{n} is the distance measured inwards along a normal. With this choice there is no limitation on the relationship between mode amplitudes and the boundary-layer thickness. However, the use of moving coordinates presents great technical complications in the averaging process. The streaming arises from terms which are of second order in the velocity and hence of order ε_2^2 . On taking mean values with respect to the time for the stream function in the fixed coordinate point, we have the Eulerian mean flow. Although such difficulties can be overcome, as it was demonstrated by Longuet-Higgins for water waves, he did demonstrate that the two approaches yield in the end the same formulae for the Lagrangian streaming. Thus following Longuet-Higgins

[9] we shall adopt the first approach, which is simpler one. In this approach the boundary conditions to be satisfied on the moving surface, $r = R_0 + \xi(\mu, t)$, are replaced by equivalent conditions to be satisfied on $r = R_0$, by use a Taylor expansion in powers of $\xi(\mu, t)$.

The boundary conditions of the first approximation now take the form

$$\left(-\frac{\mathrm{i}\omega}{2}\right)\xi_{1n} = -\frac{1}{R_0^2} \left(\frac{\partial\psi_{01}}{\partial\mu}\right)_{r=R_0},\tag{4.4}$$

$$p_{01}|_{r=R_0} - 2\eta_d \left[-\frac{1}{R_0^2} \left(\frac{\partial^2 \psi_{01}}{\partial r \partial \mu} \right)_{r=R_0} + \frac{2}{R_0^3} \left(\frac{\partial \psi_{01}}{\partial \mu} \right)_{r=R_0} \right] = \frac{\sigma}{R_0^2} (2 + \nabla_s^2) \xi_{1n}, \tag{4.5}$$

$$\left(\frac{\partial^{2} \psi_{01}}{\partial r^{2}}\right)_{r=R_{0}} - \frac{1-\mu^{2}}{R_{0}^{2}} \left(\frac{\partial^{2} \psi_{01}}{\partial \mu^{2}}\right)_{r=R_{0}} - \frac{2}{R_{0}} \left(\frac{\partial \psi_{01}}{\partial r}\right)_{r=R_{0}} = 0.$$
(4.6)

Here ∇_s^2 is the surface Laplacian. Axial symmetry means that $\nabla_s^2 \equiv \partial/\partial\mu(1-\mu^2)\partial/\partial\mu$.

Now we present the solution of Eq. (4.2). Introducing $i(\omega/2\nu) = i\delta^{-2} \equiv \alpha^2$, $\alpha = \delta^{-1} \exp(i\pi/4) = \delta^{-1}(1+i)/\sqrt{2}$ we have used the fact of separation of variables

$$\Omega_{01} = \frac{C_1}{R_0^2} \frac{[(\alpha r) h_n^{(1)}(\alpha r)]}{[(\alpha R_0) h_n^{(1)}(\alpha R_0)]} (1 - \mu^2)^{1/2} P_n^1(\mu)$$
(4.7)

where Riccati-Bessel functions $zh_n^{(1)}(z)$ has the form [14]

$$h_n^{(1)}(\alpha r) = \sqrt{\frac{\pi}{2\alpha r}} H_{n+1/2}^{(1)}(\alpha r) = -\mathrm{i}^{(n+1)} \frac{1}{\alpha r} \exp(\mathrm{i}\alpha r) \sum_{k=0}^{n} \left(n + \frac{1}{2}, k\right) \frac{1}{(-2\mathrm{i}\alpha r)^k},$$

and $P_n(\mu)$ are Legendre polynomials $P_n^1(\mu) = (1 - \mu^2)^{1/2} d/\mu [P_n(\mu)]$; C_1 is a constant. On obtaining an explicit solution for the vorticity we can evaluate the stream function

$$D^{2}\psi_{01} = -\Omega_{01} = -\frac{C_{1}}{R_{0}^{2}} \frac{\left[(\alpha r) h_{n}^{(1)}(\alpha r) \right]}{\left[(\alpha R_{0}) h_{n}^{(1)}(\alpha R_{0}) \right]} (1 - \mu^{2})^{1/2} P_{n}^{1}(\mu). \tag{4.8}$$

The general form of the solution is the sum of the partial solution of inhomogeneous equation, that is

$$\psi_{01}^{\text{partial}} = \frac{C_1}{(\alpha R_0)^2} \left(1 - \mu^2\right)^{1/2} P_n^1(\mu) \frac{[(\alpha r) h_n^{(1)}(\alpha r)]}{[(\alpha R_0) h_n^{(1)}(\alpha R_0)]}$$
(4.9)

and general solution of homogeneous one, but as $D^2\psi_{01}=0$ is the condition of potentiality we immediately obtain

$$\psi_{01}^{\text{potential}} = A_0 \mu + \sum_{l=1}^{\infty} A_l(t) \left(\frac{R_0}{r}\right)^l \left(1 - \mu^2\right)^{1/2} P_l^1(\mu). \tag{4.10}$$

To satisfy kinematic boundary condition we expand ξ in terms of spherical harmonics Y_{10} :

$$\xi = \sum_{l=0}^{\infty} \xi_l Y_{l0}(\mu), \quad Y_{l0}(\theta, \alpha) = \left[\frac{2l+1}{4\pi}\right]^{1/2} P_l(\mu), \quad \xi_0 = \bar{\xi} \sqrt{4\pi}, \quad \int_{0}^{2\pi} d\alpha \int_{0}^{\pi} \sin\theta \, d\theta \, Y_{lm}^* Y_{lm} = 1$$

and retain only terms corresponding to the breathing mode (l = 0) and the parametrically generated distortion mode (l = n). From Eq. (4.4) we immediately obtain for the distortion mode

$$\left(-\frac{\mathrm{i}\omega}{2}\right)\xi_{1n}\sqrt{\frac{2n+1}{4\pi}} = \frac{n(n+1)}{R_0^2} \left[A_n + \frac{C_1}{(\alpha R_0)^2}\right],\tag{4.11}$$

where we used that $\partial/\partial\mu[(1-\mu^2)^{1/2}P_n^1(\mu)] = -n(n+1)P_n(\mu)$.

It follows from (4.3) that the pressure is expressed in terms of the potential part of the stream function only

$$p_{01}(r,\mu) = \left(\frac{i\omega}{2}\right) \rho_0 \frac{nA_n}{R_0} \left(\frac{R_0}{r}\right)^{n+1} P_n(\mu). \tag{4.12}$$

The dynamical conditions (4.5) and (4.6) take the form

$$\left(-\frac{\mathrm{i}\omega}{2}\right) \frac{nA_n}{R_0} - \frac{\sigma}{\rho_0 R_0^2} \left[n(n+1) - 2\right] \sqrt{\frac{2n+1}{4\pi}} \,\xi_{1n} - 2\nu \left[-\frac{n(n+1)(n+2)}{R_0^3} A_n + \frac{n(n+1)}{(\alpha R_0)^2 R_0^2} C_1 \left\{ \frac{\partial/\partial R_0 \left[(\alpha R_0) h_n^{(1)}(\alpha R_0)\right]}{\left[(\alpha R_0) h_n^{(1)}(\alpha R_0)\right]} - \frac{2}{R_0} \right\} \right] = 0, \tag{4.13}$$

$$A_n(2n(n+2)) + C_1 \left\{ -1 + \frac{2n(n+1)}{(\alpha R_0)^2} - \frac{2}{\alpha^2 R_0} \frac{\partial/\partial R_0[(\alpha R_0) h_n^{(1)}(\alpha R_0)]}{[(\alpha R_0) h_n^{(1)}(\alpha R_0)]} \right\} = 0.$$
 (4.14)

In the absence of parametric driving, which will appear only in the second order ($\sim \varepsilon_1 \varepsilon_2$), these equations describe natural oscillation of the distortion mode, attenuated due to dissipative processes. The natural frequency and damping are determined by equating to zero the determinant of the linear system (4.11), (4.13), (4.14).

We can carry out some simplifications by exploiting the smallness of the viscous length $\delta = (2\nu/\omega)^{1/2}$ compared with bubble radius R_0 ($\beta \ll 1$); and the even stronger inequality $\beta \ll 1/n \ll 1$ which expresses the smallness of the viscous length relative to the distortion mode wavelength. These conditions were realized in the experiments described in the body of this paper. Thus retaining only two main terms in expansion over β , (βn) we obtain

$$(\alpha r)h_n^{(1)}(\alpha r) \simeq (-i)^{n+1} \exp(i\alpha r) \left[1 + i\frac{n(n+1)}{2\alpha r} + \cdots \right],$$

$$C_1 = \left(2n(n+2) \right) A_n \left[1 - 2\sqrt{2}(1-i)\beta \right], \tag{4.15}$$

$$A_n = \left(-\frac{\mathrm{i}\omega}{2}\right) \xi_{1n} \sqrt{\frac{2n+1}{4\pi}} \frac{R_0^2}{n(n+1)}.$$
 (4.16)

Thus expressions for the first order vorticity (4.7) and the stream function (4.9), (4.10) become

$$\Omega_{01} = \left(\frac{\mathrm{i}\omega}{2}\right) \xi_{1n} \sqrt{\frac{2n+1}{4\pi}} \frac{2(n+2)}{n+1} \exp\left[\mathrm{i}\alpha(r-R_0)\right] \left[1 - \mathrm{i}\frac{n(n+1)}{2\alpha r R_0} (r-R_0) - \frac{2}{\alpha R_0}\right] (1-\mu^2)^{1/2} P_n^1(\mu),$$

$$\psi_{01} = \left(-\frac{\mathrm{i}\omega}{2}\right) \xi_{1n} \sqrt{\frac{2n+1}{4\pi}} \frac{R_0^2}{n(n+1)} \left\{ \left(\frac{R_0}{r}\right)^n + 2n(n+2) \left[1 - \mathrm{i}\frac{n(n+1)}{2\alpha r R_0} (r - R_0) - \frac{2}{\alpha R_0} \right] \right.$$

$$\times (\alpha R_0)^{-2} \exp\left[\mathrm{i}\alpha (r - R_0)\right] \left\{ (1 - \mu^2)^{1/2} P_n^1(\mu). \right. \tag{4.17}$$

Because of the factor $\exp[i\alpha(r-R_0)] = \exp[(-1+i)(r-R_0)\delta^{-1}]$, the first order vorticity is confined to a thin oscillatory layer next to the surface of the bubble, with thickness of order δ .

5. The second approximation. Mean streaming

The streaming arises from terms which are second order in the velocity and hence of order $\sim \varepsilon_1^2$, $\varepsilon_1 \varepsilon_2$, ε_2^2 . On taking mean values with respect to the time in Eq. (3.6) we obtain for the stream function of the Eulerian mean flow $\bar{\psi} \approx \varepsilon_2^2 \bar{\psi}_{02}$

$$\nu D^4 \bar{\psi} = \frac{1}{r^2} \left\{ \left[\frac{\partial \psi_{01}}{\partial r} \frac{\partial D^2 \psi_{01}}{\partial \mu} - \frac{\partial \psi_{01}}{\partial \mu} \frac{\partial D^2 \psi_{01}}{\partial r} \right] + 2 \overline{D^2 \psi_{01} \Lambda \psi_{01}} \right\}. \tag{5.1}$$

Note, that we follow to the notations of the paper [9]. In evaluating the right-hand side of (5.1) the mean product of any two terms such as $K \exp(\mathrm{i}\omega t/2)$ and $Q \exp(\mathrm{i}\omega t/2)$ can be written as either $(1/2)KQ^*$ or $(1/2)K^*Q$, where an asterisk denotes the complex conjugate, and only the real part of the expression is accounted. The cross terms proportional to $\varepsilon_1\varepsilon_2$ vanish as they oscillate with the frequency $(\omega/2)$ and we ignore contribution of the breathing mode $\sim \varepsilon_1^2$ due to the smallness of its amplitude with respect to the distortion one $\varepsilon_1 \ll \varepsilon_2$.

We need not analyze second order pressure, so after excluding terms of second order stream function from the averaged Navier-Stokes equation we get an equation for the static pressure distribution which is balanced by square of

the velocity according to Bernoulli's principle [13]. By the same reasoning, the dynamical boundary condition (3.10) defines the static compression in the bubble, which is not the subject of the present paper. Thus we consider only the kinematic and dynamical conditions (3.7) and (3.12) which have the following form in the second approximation

$$\left[\frac{1}{R_{0}^{2}}\frac{\partial\bar{\psi}}{\partial\mu} + \frac{1}{R_{0}^{2}}\frac{\overline{\partial\xi_{1n}}}{\partial\mu}\frac{\partial\psi_{01}}{\partialr} - \overline{\left(\frac{2}{R_{0}}\frac{\partial\psi_{01}}{\partial\mu} - \frac{\partial^{2}\psi_{01}}{\partialr\partial\mu}\right)\frac{\xi_{1n}}{R_{0}^{2}}}\right]_{r=R_{0}} = 0, \tag{5.2}$$

$$\left[\frac{\partial^{2}\bar{\psi}}{\partial r^{2}} - \frac{1-\mu^{2}}{R_{0}^{2}}\frac{\partial^{2}\bar{\psi}}{\partial\mu^{2}} - \frac{2}{R_{0}}\frac{\partial\bar{\psi}}{\partial r}\right]_{r=R_{0}} + \left[\overline{\frac{\partial^{3}\psi_{01}}{\partial r^{3}}}\xi_{1n} + \frac{2(1-\mu^{2})}{R_{0}^{2}}\overline{\frac{\partial^{2}\psi_{01}}{\partial\mu^{2}}}\xi_{1n} - \frac{1-\mu^{2}}{R_{0}^{2}}\overline{\frac{\partial^{3}\psi_{01}}{\partialr\partial\mu^{2}}}\xi_{1n} + \frac{2(1-\mu^{2})}{R_{0}^{2}}\overline{\frac{\partial^{2}\psi_{01}}{\partial\mu^{2}}}\xi_{1n} + \frac{2}{R_{0}^{2}}\overline{\frac{\partial\psi_{01}}{\partialr}}\xi_{1n} - \frac{2}{R_{0}}\overline{\frac{\partial^{2}\psi_{01}}{\partialr^{2}}}\xi_{1n}\right]_{r=R_{0}}$$

$$-\frac{2}{R_{0}^{2}}\left(\overline{\frac{\partial\xi_{1n}}{\partial\mu}}\right)\left[2(1-\mu^{2})\frac{\partial^{2}\psi_{01}}{\partial r\partial\mu} + \mu\frac{\partial\psi_{01}}{\partial r} - \frac{3(1-\mu^{2})}{R_{0}}\frac{\partial\psi_{01}}{\partial\mu}\right]_{r=R_{0}} = 0.$$
(5.2)

To find a solution of Eq. (5.1) which satisfies to boundary conditions (5.2) and (5.3), we shall follow to the approach of Longuet-Higgins [9]. The explicit form of the right-hand side of Eq. (5.1) is

$$\begin{split} & \left[\left(\frac{\omega}{2} \right) R_0^2 \beta^2 \right] D^4 \bar{\psi} = \frac{1}{r^2} \left\{ \left[-\frac{A_n^* C_1}{2R_0^2} \left(\frac{R_0}{r} \right)^2 \frac{(n/r)[(\alpha r)h_n^{(1)}(\alpha r)] + (\partial/\partial r)[(\alpha r)h_n^{(1)}(\alpha r)]}{(\alpha R_0)h_n^{(1)}(\alpha R_0)} \right. \\ & \left. + \frac{C_1^* C_1}{2R_0^2 (\alpha^* R_0)^2} \frac{[(\alpha r)h_n^{(1)}(\alpha r)](\partial/\partial r)[(\alpha^* r)h_n^{(1)*}(\alpha r)] - [(\alpha^* r)h_n^{(1)*}(\alpha r)](\partial/\partial r)[(\alpha r)h_n^{(1)}(\alpha r)]}{[(\alpha R_0)h_n^{(1)}(\alpha R_0)][(\alpha^* R_0)h_n^{(1)*}(\alpha R_0)]} \right] \\ & \times (-)n(n+1) \left(1 - \mu^2 \right) P_n(\mu) \frac{\mathrm{d} P_n(\mu)}{\mathrm{d} \mu} \\ & + \left[\frac{A_n^* C_1}{R_0^2} \left(\frac{R_0}{r} \right)^n \frac{(-)(n/r)[(\alpha r)h_n^{(1)}(\alpha r)]}{(\alpha R_0)h_n^{(1)}(\alpha R_0)} + \frac{C_1^* C_1}{R_0^2 (\alpha^* R_0)^2} \frac{[(\alpha r)h_n^{(1)}(\alpha r)](\partial/\partial r)[(\alpha^* r)h_n^{(1)*}(\alpha r)]}{[(\alpha R_0)h_n^{(1)}(\alpha R_0)][(\alpha^* R_0)h_n^{(1)*}(\alpha R_0)]} \right] \\ & \times \mu \left(1 - \mu^2 \right) \left(\frac{\mathrm{d} P_n(\mu)}{\mathrm{d} \mu} \right)^2 \\ & + \left[\frac{A_n^* C_1}{R_0^2} \left(\frac{R_0}{r} \right)^n \frac{(1/r)[(\alpha r)h_n^{(1)}(\alpha r)]}{[(\alpha R_0)h_n^{(1)}(\alpha R_0)]} + \frac{C_1^* C_1}{R_0^2 (\alpha^* R_0)^2} \frac{(1/r)[(\alpha r)h_n^{(1)}(\alpha r)][(\alpha^* r)h_n^{(1)*}(\alpha r)]}{[(\alpha R_0)h_n^{(1)}(\alpha R_0)][(\alpha^* R_0)h_n^{(1)*}(\alpha R_0)]} \right] \\ & \times (-)n(n+1) \left(1 - \mu^2 \right) P_n(\mu) \frac{\mathrm{d} P_n(\mu)}{\mathrm{d} \mu} \right\}. \end{split} \tag{5.4}$$

Consider first the streaming inside the boundary layer. We write $r = R_0 + \delta \eta$ so that in the boundary layer $(r - R_0)$ is of order βR_0 and η is of order unity. Retaining only the terms of two main order in β we get

$$\frac{d^{4}\bar{\psi}}{d\eta^{4}} = -\frac{\omega}{2} \frac{2n+1}{4\pi} \frac{n+2}{n+1} |\xi_{1n}|^{2} R_{0} \exp\left(-\frac{1-i}{\sqrt{2}}\eta\right) \left\{ \left[\frac{1-i}{\sqrt{2}} (\beta - \beta^{2} \eta(n+2)) - \frac{1+i}{\sqrt{2}} \beta^{2} (n-4) \right] (1-\mu^{2}) P_{n}(\mu) \frac{dP_{n}(\mu)}{d\mu} + \frac{2\beta^{2}}{n+1} \mu (1-\mu^{2}) \left(\frac{dP_{n}(\mu)}{d\mu} \right)^{2} \right\}.$$
(5.5)

Because the two equations

$$\frac{d^4y}{d\eta^4} = \exp(-\lambda_a \eta)$$
 and $\frac{d^4y}{d\eta^4} = (\lambda_a \eta) \exp(-\lambda_a \eta)$

have particular integrals $y = \lambda_a^{-4} \exp(-\lambda_a \eta)$ and $y = \lambda_a^{-4} (4 + \lambda_a \eta) \exp(-\lambda_a \eta)$, respectively, we can integrate Eq. (5.5) exactly to give

$$\bar{\psi} = \frac{\omega}{2} \frac{2n+1}{4\pi} \frac{n+2}{n+1} |\xi_{1n}|^2 R_0 \exp\left(-\frac{1-i}{\sqrt{2}}\eta\right)
\times \left\{ \left[\frac{1-i}{\sqrt{2}} \left(\beta + \beta^2 \eta(n+2)\right) + 3\frac{1+i}{\sqrt{2}} \beta^2 (n-4) \right] \left(1-\mu^2\right) P_n \frac{dP_n}{d\mu} + \frac{2\beta^2}{n+1} \mu \left(1-\mu^2\right) \left(\frac{dP_n}{d\mu}\right)^2 \right\}
+ L(\mu) + M(\mu)\eta + N(\mu)\eta^2 + Z(\mu)\eta^3$$
(5.6)

where L, M, N, Z are the functions of the angular variable to be determined from the boundary conditions (5.2), (5.3). Instead to applying these conditions directly to $\bar{\psi}$ we follow Longuet-Higgins [9] and shall first derive boundary conditions for the Lagrangian mean stream function ψ_L . This is obtained by adding to $\bar{\psi}$ the stream function ψ_S associated with Stokes drift

$$\psi_{L} = \bar{\psi} + \psi_{S},$$

$$\psi_{S} = \frac{1}{r^{2}} \int \frac{\partial \psi_{01}}{\partial r} dt \frac{\partial \psi_{01}}{\partial \mu}.$$
(5.7)

The Lagrangian mean stream function ψ_L satisfies the same two boundary conditions at the mean position $(r = R_0)$ of the bubble surface as it would be if this surface was stationary.

$$\frac{1}{R_0^2} \frac{\partial \psi_{\rm L}}{\partial \mu} \bigg|_{r=R_0} = 0,\tag{5.8}$$

$$\left[\frac{\partial^2 \psi_{\rm L}}{\partial r^2} - \frac{1 - \mu^2}{R_0^2} \frac{\partial^2 \psi_{\rm L}}{\partial \mu^2} - \frac{2}{R_0} \frac{\partial \psi_{\rm L}}{\partial r}\right]_{r=R_0} = 0. \tag{5.9}$$

This result, obtained by Longuet-Higgins for the dipole and breathing modes of bubble oscillation, is true for the axisymmetric distortion modes. We omitted the cumbersome calculations in evaluation of the right-hand sides of Eqs. (5.8), (5.9) as we followed to the approach derived by Longuet-Higgins and the result is as expected.

The explicit expression for the contribution of the Stokes drift in Eq. (5.8) is

$$\frac{1}{R_0^2} \frac{\partial \psi_s}{\partial \mu} \Big|_{r=R_0} = \frac{1}{R_0^4} \frac{\partial}{\partial \mu} \left(\int \frac{\partial \psi_{01}}{\partial r} dr \frac{\partial \psi_{01}}{\partial \mu} \right)_{r=R_0} = \frac{1}{R_0^4 (i\omega/2)} \frac{1}{2} \frac{\partial}{\partial \mu} \left(\frac{\partial \psi_{01}^*}{\partial r} \frac{\partial \psi_{01}}{\partial \mu} \right)_{r=R_0} \\
= \frac{\omega}{2} |\xi_{1n}|^2 \frac{2n+1}{4\pi} \frac{n+2}{(n+1)R_0} \frac{\beta}{\sqrt{2}} \frac{\partial}{\partial \mu} \left[(1-\mu^2) P_n(\mu) \frac{dP_n(\mu)}{d\mu} \right]. \tag{5.10}$$

Thus, in the main order in β

$$\frac{1}{R_0^2} \frac{\partial \psi_L}{\partial \mu} \Big|_{r=R_0} = \left(\frac{1}{R_0^2} \frac{\partial \psi_S}{\partial \mu} + \frac{1}{R_0^2} \frac{\partial \bar{\psi}}{\partial \mu} \right)_{r=R_0} = \frac{\omega}{2} |\xi_{1n}|^2 \frac{2n+1}{4\pi} \frac{n+2}{(n+1)R_0} \frac{\beta}{\sqrt{2}} \\
\times \frac{\partial}{\partial \mu} \left((1-\mu^2) P_n \frac{dP_n}{d\mu} \right) + \frac{\omega}{2} |\xi_{1n}|^2 \frac{2n+1}{4\pi} \frac{n+2}{(n+1)R_0} \frac{\beta}{\sqrt{2}} \frac{\partial}{\partial \mu} \left((1-\mu^2) P_n \frac{dP_n}{d\mu} \right) \\
+ \frac{1}{R_0^2} \frac{\partial}{\partial \mu} L(\mu) = 0, \tag{5.11}$$

giving

$$L(\mu) = Const - \sqrt{2}\beta \left(\frac{\omega}{2}\right) |\xi_{1n}|^2 R_0 \frac{2n+1}{4\pi} \frac{n+2}{(n+1)} (1-\mu^2) P_n \frac{dP_n}{d\mu}.$$
 (5.12)

An explicit form for the Stokes stream function ψ_S in the boundary layer is found from Eqs. (5.7) and (4.17) specifically

$$\psi_{S} = \frac{\omega}{2} |\xi_{1n}|^{2} R_{0} \frac{2n+1}{4\pi} \frac{n+2}{n+1} \exp\left(-\frac{\eta}{\sqrt{2}}\right) \times \left\{\beta \cos\left(\frac{\eta}{\sqrt{2}} + \frac{\pi}{4}\right) \left(1 - \beta \eta (n+2)\right) + \beta^{2} (n+3) \sin\left(\frac{\eta}{\sqrt{2}}\right)\right\} \left(1 - \mu^{2}\right) P_{n} \frac{dP_{n}}{d\mu}.$$
(5.13)

When this expression is added to $\bar{\psi}$ (Eq. (5.6)) to obtain ψ_L we find in the boundary layer

$$\psi_{L} = \frac{\omega}{2} |\xi_{1n}|^{2} R_{0} \frac{2n+1}{4\pi} \frac{n+2}{n+1} \exp\left(-\frac{\eta}{\sqrt{2}}\right) \left\{ \left[\cos\left(\frac{\eta}{\sqrt{2}}\right) (\beta\sqrt{2}+3\beta^{2}(n-4)) + \sin\left(\frac{\eta}{\sqrt{2}}\right) \beta^{2} ((n+3)+\sqrt{2}(n+2)\eta) \right] (1-\mu^{2}) P_{n} \frac{dP_{n}}{d\mu} + \beta^{2} \frac{2}{n+1} \cos\left(\frac{\eta}{\sqrt{2}}\right) \mu (1-\mu^{2}) \left(\frac{dP_{n}}{d\mu}\right)^{2} \right\} + L(\mu) + M(\mu)\eta + N(\mu)\eta^{2}.$$
(5.14)

Expanding the boundary condition (5.9) in two main order in β gives:

$$\psi_{L} = \beta \left(\tilde{\psi}_{1L} + L_{1}(\mu) \right) + \beta^{2} \tilde{\psi}_{2L} + \left(M_{0} + \beta M_{1} + \beta^{2} M_{2} \right) \eta + \left(N_{0} + \beta N_{1} + \beta^{2} N_{2} \right) \eta^{2},
\left[\frac{1}{\beta^{2} R_{0}^{2}} \frac{\partial^{2} \psi_{L}}{\partial \eta^{2}} - \frac{2}{\beta R_{0}^{2}} \frac{\partial \psi_{L}}{\partial \eta} - \frac{1 - \mu^{2}}{R_{0}^{2}} \frac{\partial^{2} \psi_{L}}{\partial \mu^{2}} \right]_{\eta=0} \approx \frac{N_{0}}{\beta^{2} R_{0}^{2}} + \frac{1}{\beta R_{0}^{2}} \left(\frac{\partial^{2} \tilde{\psi}_{1L}}{\partial \eta^{2}} - 2M_{0} + 2N_{1} \right)_{\eta=0}
+ \frac{1}{R_{0}^{2}} \left(\frac{\partial^{2} \tilde{\psi}_{2L}}{\partial \eta^{2}} - 2\frac{\partial \tilde{\psi}_{1L}}{\partial \eta} - 2M_{1} + 2N_{2} \right)_{\eta=0} = 0. \quad (5.15)$$

Now since $(\partial^2 \tilde{\psi}_{1L}/\partial \eta^2)_{\eta=0} = 0$ then

$$N_0 = 0$$
, $M_0 - N_1 = 0$,

$$M_{1} - N_{2} = \frac{1}{2} \left(\frac{\partial^{2} \tilde{\psi}_{2L}}{\partial \eta^{2}} - 2 \frac{\partial \tilde{\psi}_{1L}}{\partial \eta} \right)_{n=0} = \frac{\omega}{2} |\xi_{1n}|^{2} R_{0} \frac{2n+1}{4\pi} \frac{(n+2)(n+3)}{2(n+1)} (1-\mu^{2}) P_{n} \frac{dP_{n}}{d\mu}.$$
 (5.16)

Thus the boundary conditions (5.8), (5.9) define function L and yield to the relations (5.16) for functions M and N. Now we consider outer solution. When $\eta \gg 1$ the right-hand side of Eq. (5.1) is exponentially small and $\bar{\psi}$ should satisfy

$$D^4 \bar{\psi} = 0. ag{5.17}$$

Assuming the same form of angular dependence of the outer solution as the inner one

$$\bar{\psi}(r,\mu) = \bar{\psi}(r)\left(1 - \mu^2\right)P_n\frac{\mathrm{d}P_n}{\mathrm{d}\mu},\tag{5.18}$$

we simplify this dependence by use the asymptotic of Legendre polynomials for $(n \gg 1)$

$$P_n(\cos\vartheta) \approx \sqrt{\frac{2}{\pi n}} \frac{\sin[(n+1/2)\vartheta + \pi/4]}{\sqrt{\sin\vartheta}}, \quad \vartheta n \gg 1, \ (\pi - \vartheta)n \gg 1,$$

$$P_n(\cos\vartheta) \approx J_0\left(\left(n + \frac{1}{2}\right)\vartheta\right), \quad \vartheta \ll 1.$$

Thus for $\vartheta n \gg 1$, $(\pi - \vartheta)n \gg 1$

$$(1 - \mu^2) P_n \frac{\mathrm{d}P_n}{\mathrm{d}\mu} \approx \sin^2 \vartheta \frac{2}{\pi n} (-) \frac{n + 1/2}{\sin \vartheta} \frac{\sin[(n + 1/2)\vartheta + \pi/4] \cos[(n + 1/2)\vartheta + \pi/4]}{\sin \vartheta}$$
$$\approx -\frac{1}{\pi} \cos[(2n + 1)\vartheta]. \tag{5.19}$$

Satisfying this form (5.18), (5.19) of the solution the equation of (5.17) we obtain

$$\bar{\psi}(r) = E\left(\frac{R_0}{r}\right)^{k_1} + F_1\left(\frac{R_0}{r}\right)^{k_2},$$

$$\left[k(k+1) - (2n+1)(2n+2)\right] \left[(k+2)(k+3) - (2n+1)(2n+2)\right] = 0, \quad k_1 = 2n-1, \quad k_2 = 2n+1.$$
(5.20)

Consider the domain of some viscous length from the bubble wall where one can ignore exponential terms such that

$$\bar{\psi}(r,\mu) = \left(L + M\eta + N\eta^2 + \cdots\right)\left(1 - \mu^2\right)P_n\left(\frac{\mathrm{d}P_n}{\mathrm{d}\mu}\right). \tag{5.21}$$

Sewing together the outer solution (5.20) expanded in power of β

$$\bar{\psi}(r) \approx (E+F) - [(2n-1)E + (2n+1)F_1]\beta\eta + [(2n-1)2nE + (2n+1)(2n+2)F_1]\frac{(\beta\eta)^2}{2} + \cdots$$

with the inner solution (5.21), we obtain

$$L = E + F_1, \quad M = -[(2n-1)E + (2n+1)F_1]\beta, \quad N = [(2n-1)nE + (2n+1)(n+1)F_1]\beta^2.$$

Since the Faraday waves on bubble wall with $n \gg 1$ look very much like the capillary waves on the flat surface because their wavelengths are much smaller then the radius of curvature, we can take advantage of the well-known fact that second order vorticity induced by surface waves does not depend on viscosity outside the boundary layer [9,15]. This means that the outer solution (5.20) and therefore E and F_1 should not depend on β . This immediately gives

$$E + F_1 = L_0 = 0$$
, $M_0 = 0$, $M_1 = -[(2n-1)E + (2n+1)F_1] = -2F_1$, $N_0 = 0$, $N_1 = 0$
 $N_2 = [(2n-1)nE + (2n+1)(n+1)F_1] = (4n+1)F_1$. (5.22)

Now using Eq. (5.16) we have

$$M_{1} - N_{2} = \frac{\omega}{2} |\xi_{1n}|^{2} R_{0} \frac{2n+1}{4\pi} \frac{(n+2)(n+3)}{2(n+1)} = -2F_{1} - (4n+1)F_{1} = -(4n+3)F_{1},$$

$$F_{1} = -\frac{\omega}{2} |\xi_{1n}|^{2} R_{0} \frac{2n+1}{4\pi} \frac{(n+2)(n+3)}{2(n+1)(4n+3)},$$

$$M_{1} = \frac{\omega}{2} |\xi_{1n}|^{2} R_{0} \frac{2n+1}{4\pi} \frac{(n+2)(n+3)}{(n+1)(4n+3)},$$

$$N_{2} = -\frac{\omega}{2} |\xi_{1n}|^{2} R_{0} \frac{2n+1}{4\pi} \frac{(n+2)(4n+1)(n+3)}{2(n+1)(4n+3)}.$$
(5.23)

Substituting in (5.20) we have the lowest order Lagrangian stream function, coinciding with Eulerian one in the outer region

$$\bar{\psi}(r,\mu) \approx \psi_{L}(r,\mu) = \frac{(\omega/2)|\xi_{1n}|^{2}R_{0}}{\pi} \frac{2n+1}{4\pi} \frac{(n+2)(n+3)}{2(n+1)(4n+3)} \times \left[-\left(\frac{R_{0}}{r}\right)^{(2n-1)} + \left(\frac{R_{0}}{r}\right)^{(2n+1)} \right] \cos[(2n+1)\vartheta].$$
(5.24)

6. Discussion

Consider the radial and tangential velocities corresponding to the Lagrangian stream function (5.23). Using Eqs. (3.2) we find

$$\bar{u}_r = \frac{(\omega/2)|\xi_{1n}|^2}{2\pi R_0} \frac{2n+1}{4\pi} \frac{(n+2)(2n+1)(n+3)}{(n+1)(4n+3)} \left(\frac{R_0}{r}\right)^{(2n-1)} \left[1 - \left(\frac{R_0}{r}\right)^2\right] \frac{\sin[(2n+1)\vartheta]}{\sin\vartheta},$$

$$\bar{u}_\vartheta = \frac{(\omega/2)|\xi_{1n}|^2}{2\pi R_0} \frac{2n+1}{4\pi} \frac{(n+2)(2n-1)(n+3)}{(n+1)(4n+3)} \left(\frac{R_0}{r}\right)^{(2n+1)} \left[1 - \frac{2n+1}{2n-1} \left(\frac{R_0}{r}\right)^2\right] \frac{\cos[(2n+1)\vartheta]}{\sin\vartheta}.$$
(6.1)

Comparing these expressions with the streaming velocities around a spherical bubble executing both lateral and radial oscillations (which are nearly an order magnitude greater than from purely transverse oscillations) [9]

$$\begin{split} \bar{u}_r &= \frac{\omega |\xi_0| |\xi_1|}{R_0} \sin \phi \left(-\frac{R_0}{r} + \frac{1}{2} \frac{R_0^3}{r^3} + \frac{1}{2} \frac{R_0^6}{r^6} \right) \cos \vartheta, \\ \bar{u}_{\vartheta} &= \frac{\omega |\xi_0| |\xi_1|}{R_0} \sin \phi \left(\frac{1}{2} \frac{R_0}{r} + \frac{1}{4} \frac{R_0^3}{r^3} + \frac{R_0^6}{r^6} \right) \sin \vartheta \end{split}$$
(6.2)

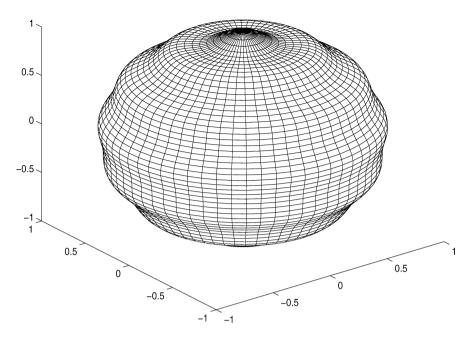


Fig. 1. The shape of axi-symmetric n = 13 (m = 0) surface mode.

we note that they are the same order in the domain $r \sim R_0$, if the amplitude of the distortion mode is comparable with the amplitude of the breathing mode ξ_0 and the dipole mode $\xi_1(|\xi_n| \sim |\xi_0|, |\xi_1|)$. But in reality, for the experiments [1–3] with parametrically generated Faraday waves the amplitude of the surface modes is more than an order magnitude greater than the amplitude of the radial oscillations. Thus streaming is dominated by the surface wave contribution in the region of the liquid from the bubble wall out to distances equivalent to several wavelengths of that surface wave. At greater distances the contribution of the streaming due to radial and lateral oscillation will dominate. In the region for which $r = R_0 + (\pi R_0/n)x$, $x \sim 1$

$$\left(\frac{R_0}{r}\right)^{(2n+1)} \approx \left(1 - \frac{\pi x}{n}\right)^{(2n+1)} \approx \exp(-2\pi x) = \exp\left(-\frac{2\pi}{R_0}(r - R_0)\right),$$

Faraday waves on the bubble wall for $(\pi R_0/n) \ll R_0$ resemble the capillary waves on a flat surface.

To evaluate the characteristic flow in [1–3] the values of relevant parameters are chosen to correspond with the conditions of the experiment, specifically $R_0 = 1.56$ mm, $\xi = 0.05$ mm, f = 2000 Hz, n = 13. For this case the shape of the n = 13 axi-symmetric mode is shown in Fig. 1. Axi-symmetric patterns of streaming around a single bubble based on the equations labeled 6.1 are shown in Fig. 2(a), (b). So when $r \approx R_0 + \lambda_n = (1 + 2\pi/13)R_0$ the contribution to mass flux by convective transfer $\bar{u}_r \approx 0.1 \times \sin[(2n+1)\vartheta] \text{ mm s}^{-1}$ (see also Fig. 2(b)) will dominate over the contribution from diffusion $\sim (D/\lambda_n) \sim 3 \times 10^{-3} \text{ mm s}^{-1}$.

Acoustoelectrochemistry [1–3] incorporates the effect of acoustic fields on the investigation of chemical processes through the application and measurement of an appropriate electrical current. The bubble activity is the way in which acoustics can affect electrochemical reactions. The electrochemical current is related to rate of flux of species

$$i = n_e F S k_m C$$

where n_e is the number of electrons exchanged, F is the Faraday's constant, S is the area of the electrode, C is the concentration of the particular electroactive species, k_m is the mass transfer coefficient. The mass transfer coefficient is determined by the microstreaming induced by the bubble (6.1) for the conditions of experiments [1–3]. It is given by $k_m = (4D/\pi a)$, where D represents the diffusion coefficient and a the radius of the microelectrode, when convective transfer can be ignored.

The plots of the current against time recorded by a 25 μ m Pt microelectrode are available in the experiments [1–3] and give rise to the observed dependence of current on amplitude of bubble oscillation. The presence of microelectrode disturbs the mean flow (6.1) and the resulting velocity incorporates the microstreaming induced by the bubble and

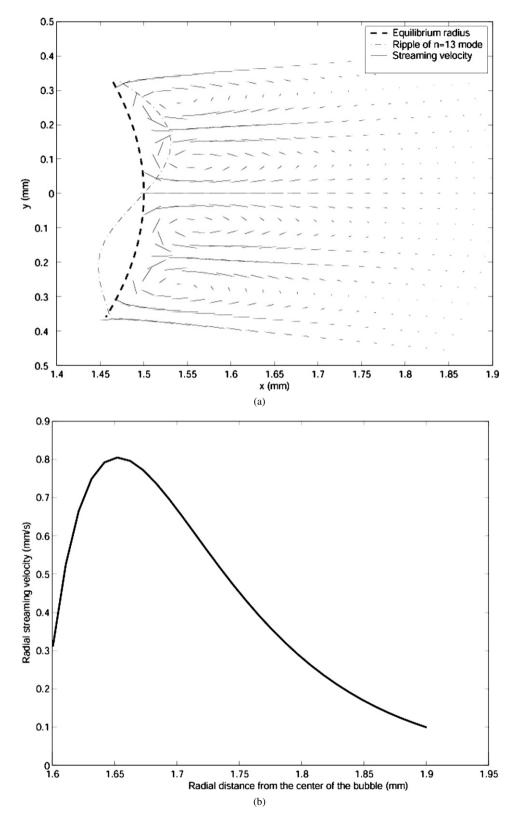


Fig. 2. (a). Streaming induced by the axi-symmetric (n=13, m=0) mode near the equator of the bubble. The calculations are based on Eq. (6.1) for $R_0=1.56$ mm, $\xi=0.05$ mm, f=2000 Hz, n=13. (b) The absolute value of the radial velocity \bar{u}_r near the equator.

the flow around the microelectrode. To analyze the problem one should consider characteristic scales expressed by Reynolds and Peclet numbers, where we assume that the characteristic velocity \bar{u} coincides on the order of magnitudes with one given by (6.1) at the distance corresponding to the microelectrode location. There can be three regimes of the flow around the microelectrode leading to $i \sim \bar{u}$, $i \sim (\bar{u})^{1/2}$, or $i \sim (\bar{u})^{1/3}$. These regimes can be simultaneously realized with decreasing distance between the microelectrode and the bubble wall. The extensional study on the electrochemical measurement of mass flux in liquids associated with surface waves on the walls of acoustically excited gas bubbles, where this matter is discussed in details, will appear as a separate publication.

Circulation is in cells bounded by the azimuth surfaces $\vartheta = \pi/2n$ (see Fig. 2(a)) that is in accordance with the Longuet-Higgins result [5] for the standing water waves where the circulation takes place within a quarter of a wavelength cell.

Microstreaming induced by the parametrically excited surface waves will be less effective for small bubbles of about 10–100 µm radius which can be potentially used in microfluidic devices [10,11]. Resonance condition: the frequency of the breathing equals to twice the frequency of the shape oscillation can be satisfied only for the lowest mode numbers n = 2, 3, as a result the amplitudes of the distortion modes will be comparable with that of the breathing mode. On the other hand, the presence of the rigid wall, to which small bubble adhering in the experiments [10,11] influences notably its dynamics [16]. In this case microstreaming can be produced by volumetric bubble oscillations only. There is an analogy to the streaming induced by shallow standing water waves [5] where the circulation is driven by the tangential velocity near the bottom. Thus there should be second cell besides that observed in [10] probably smaller in size and located near the bubble contact line. Circulation in this cell should be opposite to those observed in the region of the polar hemisphere [10].

7. Conclusions

It has been shown that the acoustical streaming from a bubble undergoing axi-symmetric surface wave oscillations is enhanced in comparison with lateral oscillations. When the bubble simultaneously pulsates in a radial sense and parametrically generates surface waves, the magnitude of the streaming induced by the surface waves is greater than the streaming induced by combined effect of lateral and radial oscillations due to greater amplitude of distortion modes.

The present analysis is strictly valid only for oscillations of the bubble that are small compared to the bubble radius. The approach used coordinates fixed in space and boundary conditions have to be satisfied on a variable surface. This seems to restrict us to the displacement amplitudes that are small compared to the boundary layer thickness. However in an investigation involving a moving stress-free surface, Longuet-Higgins discovered that the two approaches yield in the end the same formulae for the Lagrangian streaming. Thus our results for the Lagrangian streaming are not necessarily invalid when the oscillations are not small compared to the thickness of the shear layer.

Lastly we note that this study was initiated by the observations of bubble oscillation by electrochemical sensing technique. Now we can quantitatively evaluate the magnitude of the measured currents and its relation to the optically detected amplitudes of bubble oscillations.

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