Bubble population phenomena in acoustic cavitation

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Received 8 October 1994

Theoretical treatments of the dynamics of a single bubble in a pressure field have been undertaken for many decades. Although there is still scope for progress, there now exists a solid theoretical basis for the dynamics of a single bubble. This has enabled useful classifications to be established, including the distinction between stable cavitation (where a bubble pulsates for many cycles) and transient cavitation (where the bubble grows extensively over time-scales of the order of the acoustic cycle, and then undergoes an energetic collapse and subsequent rebound and then, potentially, either fragmentation, decaying oscillation or a repeat performance). Departures from sphericity, such as shape and surface oscillations and jetting, have also been characterized. However, in most practical systems involving high-energy cavitation (such as those involving sonochemical, biological and erosive effects), the bubbles do not behave as the isolated entities modelled by this single-bubble theory: the cavitational effect may be dominated by the characteristics of the entire bubble population, which may influence, and be influenced by, the sound field.

The well established concepts that have resulted from the single-bubble theory must be reinterpreted in the light of the bubble population, an appreciation of population mechanisms being necessary to apply our understanding of single-bubble theory to many practical applications of 'power' ultrasound. Even at a most basic level these single-bubble theories describe the response of the bubble to the *local* sound field at the position of the bubble, and that pressure field will be influenced by the way sound is scattered by neighbouring bubbles. The influence of the bubble population will often go further, a non-uniform sound field creating an inhomogeneous bubble distribution. Such a distribution can scatter, channel and focus ultrasonic beams, can acoustically shield regions of the sample, and elsewhere localize the cavitational activity to discrete 'hot spots'. As a result, portions of the sample may undergo intense sonochemical activity, degassing, erosion, etc., whilst other areas remain relatively unaffected. Techniques exist to control such situations where they are desirable, and to eliminate this localization where a more uniform treatment of the sample is desired.

Keywords: bubble population phenomena; acoustic cavitation

1. Introduction

In 1917, Lord Rayleigh[1] developed his pioneering analysis for the collapse of an empty spherical cavity under a static pressure. Coupling this energetic collapse phase with the explosive growth phase of a sufficiently small bubble (as expounded by Blake[2]), Noltingk and Neppiras[3,4] characterized a particular type of cavitation whereby appropriately small bubbles in sufficiently strong sound fields undergo growth to many times their original size and then subsequent rapid collapse. The growth phase is to a first approximation isothermal, and the collapse phase adiabatic, such that the bubble serves to concentrate the acoustic energy. Flynn[5] further distinguished this so-called 'transient' cavitation from the less energetic 'stable' cavitation, where the bubble pulsates about an equilibrium radius over many acoustic cycles. Flynn then analysed the energetics of transient collapse, through consideration of the mechanical work done on the cavity by the spherical convergence of the liquid, and the dissipation of energy during the collapse process[6]. Subsequent formulations have concentrated mainly on attempting to distinguish the threshold values of acoustic frequency, pressure amplitude and initial bubble radius corresponding to the transition between stable and transient cavitation[7–17]. The equations of motion of single bubbles undergoing stable cavitation in acoustic fields have been further developed[18–34] to investigate a range of phenomena, notably rectified diffusion[35–46] and bifurcation behaviour[42–48].

1350-4177/95/$09.50
SSDI 1350-4177(95)00021-6

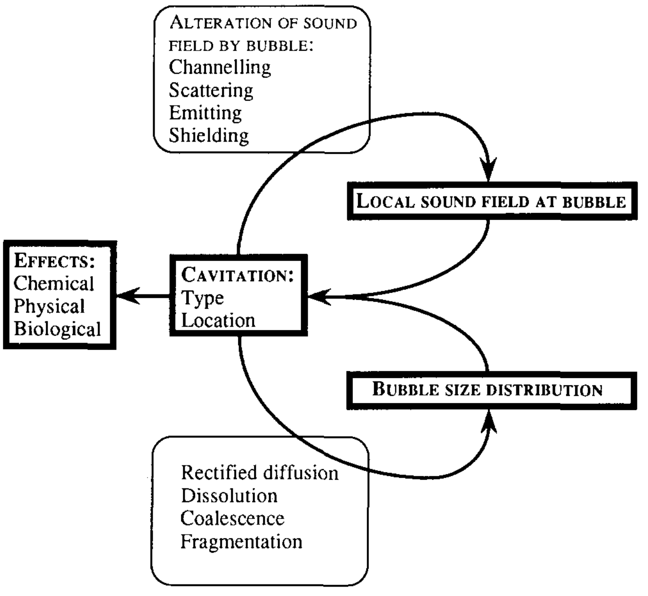

Figure 1 The chemical, physical and biological effects of cavitation depend both on the type of cavitation (e.g. transient, stable, jetting, fragmentary) and its location. Both of these factors depend strongly on the local sound field at the bubble and on the sizes of bubble present in the population. These two together, for example, characterize the transient cavitation threshold, and also where bubbles will migrate and accumulate under radiation forces. Such accumulation will in turn affect the local sound field, through the processes of channelling, scattering and shielding, and will affect the bubble size distribution through its influence on the processes of coalescence, fragmentation and rectified diffusion. In summary, therefore, the observed effect depends on the characteristics of the cavitation, which are determined by the local sound field and the bubble size distribution. However, there is feedback from the cavitation which influences these two key parameters. These interactions are the topic of this paper

The results of such work substantiate Flynn's approximate categorization. The oscillations of single bubbles in acoustic fields may be classified as stable (being approximately linear at low amplitude and non-linear at high amplitudes) or, for a sufficiently small bubble in a sufficiently intense pressure field, transient. During transient collapse, energy is focused within the bubble to generate a range of potential local effects, including a gaseous hot spot[3,49] and gas shocks[50], both of which can potentially generate free radicals and other reactive chemical species within the bubble gas[51§5.2.1], the subsequent reaction of which can generate sonoluminescence[51§5.2,52–54]. In addition, such concentration can generate effects in the medium surrounding the bubble, including spherically diverging liquid shocks resulting from bubble rebound[55,56], which may lead to erosion of solids placed within a cavitating liquid (as can the liquid jet resulting from bubble involution[57–61]). However, all these effects which characterize transient cavitation are based on the energetics of the collapse, which are high if inertial forces dominate the collapse[6], rather than on the temporal characteristics. Therefore, with the discovery of sonoluminescence associated with stable cavitation under controlled conditions[62,63], it has been suggested that the term 'inertial cavitation' would more appropriately describe the type of cavitation which gives rise to the above effects[64].

Whilst theory has tended to concentrate on the dynamics of single bubbles, in practice when cavitation causes sonochemical and erosive changes, it is generally *populations* of bubbles that are involved: single-bubble manifestations tend to occur only in controlled laboratory conditions (e.g. for sonoluminescence[62,63] and jetting[65–67]). The influence of the population may arise through *direct* action. For example, whilst the rebound shock from a single bubble is rapidly attenuated with distance such that only surfaces within about one bubble radius from the centre of collapse may be damaged by the rebound of a single bubble[68,69], the combined and cooperative shocks from a concentrated mass of bubbles (where the collapse of one may initiate the collapse of a neighbour) can cause damage at much greater distances[70,71]. Significant erosion by jet impact is usually the cumulative result of the individual involutions of many members of a bubble population. The direction of the jet may be influenced by neighbouring bubbles[65,72]. However, it is the *indirect* action of the population which is the subject of this paper. This can also be illustrated through jet impact erosion, where the daughter bubble fragments produced by the involution of a single bubble[73,74] may seed many subsequent cavitation events. This in turn illustrates how the number, size and distribution of bubbles within the population may determine the macroscopic effect observed during energetic cavitation.

The exploitation of the chemical and mechanical effects resulting from the acoustically induced cavitation of a cloud of bubbles in many common applications (such as cell killing, ultrasonic cleaning, cavitation erosion and sonochemistry) relies in action upon the relatively high-energy thermal, sonochemical, gas and liquid shock and jetting processes described above. However, the application of these effects depends on the characteristics of the bubble population, which may be influenced not only by the high-energy events but also by the relatively low-energy phenomena of bubble migration (Section 4), dissolution and exsolution (Section 3) with which this paper is concerned (*Figure 1*).

The discussion begins with a description of the way the bubble population can modify the incident sound field (Section 2). The formulations by which the behaviour of a bubble in a sound field is characterized and predicted are primarily concerned with the effect on a single bubble of the *local* pressure field. In many practical situations this local field can differ from the field one would predict in a bubble free liquid in its intensity, temporal profile, isotropy and directionality, and in the relative proportions of static, acoustic and discontinuous pressure fields (*Figure 1*). In intensely cavitating fields it will rarely be possible to characterize the local pressure field at an individual bubble; however, an appreciation of the deviations from the bubble-free pressure field, and a qualitative knowledge of the likely effects of the perturbations, may be vital in linking the predictions of single bubble theory with the observed effects of high-energy acoustic cavitation in a bubble population.

2. Equivalent bulk properties of bubbly liquids

The acoustic behaviour of a simple homogeneous liquid can to a first approximation be characterized through the specification of a number of basic parameters, notably the density, ρ, and bulk modulus, B, (or equivalently the

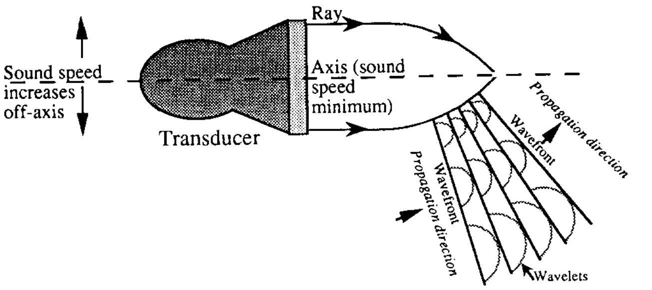

Figure 2 The use of Huygen's principal to illustrate self-focusing towards the axis of a transducer, the sound speed being a minimum there as a result of the beam intensity being a maximum on axis. The rays, which are locally perpendicular to the waveforms, bend inwards as successive wavefronts are angled in towards the beam axis; the inclination of each wavefront is governed by the envelope of the wavelets emitted by the preceding wavefront, the wavelets propagating in a given time interval a distance proportional to the local sound speed

sound speed, c, and the specific acoustic impedance, $Z = \rho c$) and the attenuation. A volume of liquid containing a uniform distribution of a large number of bubbles separated by much less than the acoustic wavelength may be treated as an equivalent fluid, the bubbles tending to reduce the density and bulk modulus (or equivalently increase the compressibility) of the 'bubbly liquid' compared with the bubble-free liquid. As a result, the specific acoustic impedance and sound speed $c = \sqrt{B/\rho}$ are also reduced. Whilst real bubble populations are seldom homogeneous, it is sometimes possible to distinguish regions of approximate uniformity to which equivalent bulk properties can be assigned. This will permit an appreciation of how an inhomogeneous bubble population, when considered to be made up of volumes of bubbly liquid having differing equivalent properties which may vary smoothly or discontinuously between regions, can interact with the sound field, and how ultrasonic beams may undergo reflection, refraction and attenuation by such populations.

In a low-amplitude sound field, bubbles behave as approximately single degree of freedom oscillators with the resonance frequency which varies inversely with bubble radius[75]. If *low*-amplitude sound of a specific frequency is passed through a bubbly liquid, the effect of the bubbles on the field will be dominated by those bubbles which have radii that are close to the bubble size that is resonant at the acoustic frequency[51§4.1.2,76].

If there is a distribution of bubble sizes within the cloud, such that $n_b^{gr}(z, R_0)\,dR_0$ is the number of bubbles per unit volume at depth z having radii between R_0 and $R_0 + dR_0$, the phase speed of sound in the bubbly liquid, c_c, is a function of both the depth and the acoustic frequency, and is related to the phase speed of sound in bubble-free liquid, c_0, by

$$c_c(z, \omega) = c_0\left(1 - (2\pi c_0^2)\int_{R_0=0}^{\infty} \frac{R_0}{\omega^2}\left\{\frac{(\omega_0/\omega)^2 - 1}{[(\omega_0/\omega)^2 - 1]^2 + d^2}\right\} \times n_b^{gr}(z, R_0)\,dR_0\right) \quad (1)$$

where ω is the circular frequency of the sound field, ω_0 the bubble resonance frequency and d the dimensionless damping coefficient of the bubble, which is dependent on R_0 and ω. Limiting forms of this equation can be found elsewhere[51§3.8.2b(i)].

The dominance of resonant bubbles indicates that the sound speed will be frequency dependent, and the addition of bubbles to a medium will cause dispersive propagation. Discontinuous variations in any of Z, ρ or c will result in the familiar phenomenon of beam reflection, with associated applications of Snell's law and definition of appropriate reflection and transmission coefficients. Continuous variations can cause refraction and the institution of wave guides[77,78]. Refraction can be simply demonstrated by applying Huygen's principle to construct schematically an off-axis ray path in an ultrasonic beam. Assume that the beam has maximum intensity at the centre, falling off monotonically as one moves off-axis. Spatial variations in intensity can lead to sound speed inhomogeneities, for example by causing the accumulation of bubbles (see Section 4). If, for example, the medium contains primarily bubbles which are smaller than resonance size, the intensity variation described above will cause an accumulation of small bubbles on the axis of the beam, with fewer bubbles off-axis. This will cause the sound speed to decrease towards the centre of the beam. The resulting ray paths can be seen through the Huygen's construction in *Figure 2*. Consider the off-axis beam shown. Perpendicular to the ray path, the local wavefront can be viewed as a continuous source of Huygen's wavelets. These wavelets propagate out from the wavefront, their envelope a short time later giving the new position of the wavefront. Those wavelets closer to the beam axis propagate less far in the time interval, since the sound speed there is less. The resultant wavefront is angled with respect to the previous wavefront, and the ray is seen to bend inwards towards the centre of the beam. This phenomenon is known as *self-focusing*[79–81]. It should be remembered that this is a demonstration scheme only: a preponderance of bubbles larger than resonance size may cause self-defocusing, as the accumulation of bubbles off-axis causes the ray path to bend in that direction. Thermal effects, if dominant over bubble effects, can also cause self-focusing (for example in water at temperatures in excess of 74 °C) and self-defocusing (at lower water temperatures)[82], since in pure water at 1 bar the sound speed is a maximum[83,84] at 74 °C. Other self-effects which may be bubble- or thermal-mediated include self-concentration[85] and self-transparency[85–87]. The addition of bubbles to a fluid can increase attenuation of an ultrasonic beam both by scattering energy out of the beam and by converting it to heat through the thermal and viscous damping processes to which bubble pulsations are subject[26,89–91].

3. Alteration of bubble radii through mass transfer

The size distribution of bubbles in a population is a key parameter in determining the acoustic effects. There is, for example, a critical size range in which a free-floating bubble must lie in order to undergo inertial cavitation[6,51§4.3.1]. However the limits of that range are dependent on the characteristics of the local field, which may be influenced by bubble-mediated changes in sound speed. This mediation is strongly dependent on how the bubble population is distributed with respect to the radius which is resonant with the sound field (note the above

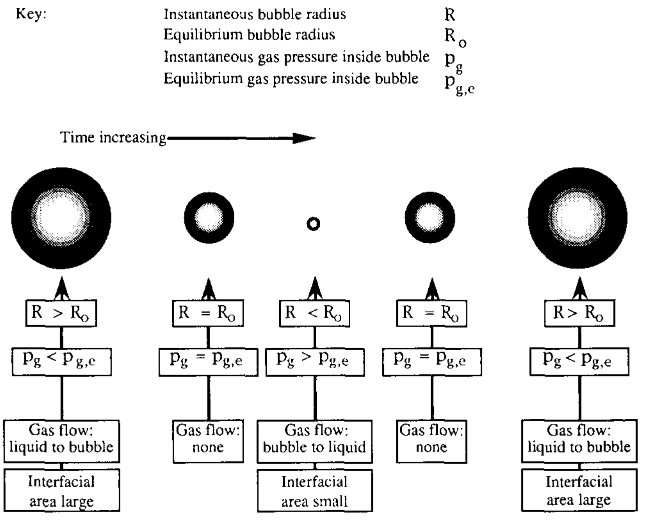

Figure 3 The 'area' effect. One oscillatory cycle of the bubble pulsation is illustrated. The pressure within a pulsating bubble and the surface area of the transfer (the bubble wall) both oscillate about an equilibrium value, causing a mass flux imbalance

example of bubble-mediated self-focusing and self-defocusing).

Many other phenomena are similarly dependent on the relationship of the size distribution to the resonance. These include radiation forces, which are described in Section 4, and can lead to coalescence. This section describes others, such as bubble fragmentation and coalescence, rectified diffusion and microstreaming, which like coalescence are not only dependent on this relationship but may also in turn affect the bubble size distribution and therefore the cavitational effects observed.

A gas bubble in a liquid, in the absence of a sound field, will slowly dissolve owing to the excess internal gas pressure[92] required to balance the presssure $2\sigma/R$ due to surface tension σ. Thus, in the absence of a sound field and any stabilizing mechanisms[51§2.1.2(b)], all bubbles gradually dissolve[93]. In the presence of a sound field, the situation is different. During stable cavitation, since evaporation and condensation take place so much more rapidly than the bubble dynamics, it is a common approximation to assume that the vapour pressure within the bubble remains constant at the equilibrium value. However, this is not so for the gas content of the bubble, a gas which will also be dissolved in the liquid. Harvey *et al.*[94], studying the formation of bubbles in animals, suggested a mechanism by which bubbles undergoing stable cavitation in a sound field can experience a steady increase in R_0, their equilibrium radius. This inwardly directed *rectified diffusion* comes about through the active pumping of gas, initially dissolved in the liquid, into the bubble, using the energy of the sound field. There are two contributory elements to a full description of the processes, an 'area effect' and a 'shell effect'.

The area effect arises through the correlation between the direction of mass flux and the area of the bubble wall. Whilst the bubble radius is less than equilibrium, the gas inside is at a greater pressure than the equilibrium value, and thus diffuses out into the liquid. Conversely, when the bubble radius is greater than R_0 (except when it is just greater than R_0, at which time there is an excess internal pressure resulting from surface tension), the internal gas pressure is less than the equilibrium value, and so gas diffuses from the liquid into the bubble interior. The net flow-rate of the gas, however, is not equal during the compressed and expanded phases of the bubble motion, because the area of the bubble wall (the transfer interface) is greater in the latter case than in the former. This process is illustrated in *Figure 3*. Therefore, over a period of time, there will be a net influx of gas to the bubble interior.

The shell effect occurs because the diffusion rate of a gas in a liquid is proportional to the concentration gradient of the dissolved gas. As the bubble pulsates, a spherical shell of liquid surrounding the bubble will change volume, and so the concentration gradient will change. Consider the bubble shown at equilibrium in *Figure 4a*. Two of the shells surrounding the bubble are illustrated schematically. When the bubble is expanded, each liquid shell contracts (*Figure 4b*). The concentration of dissolved gas in the liquid adjacent to the bubble wall is less than the equilibrium value (Henry's law), but the

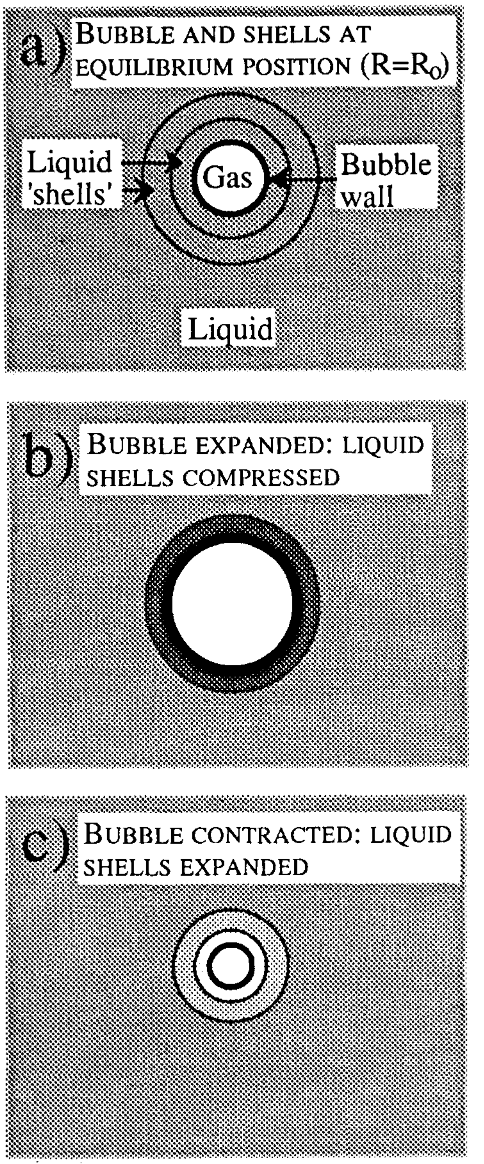

Figure 4 The 'shell' effect: the bubble and two of the liquid shells surrounding it are shown. (a) The bubble is at equilibrium size; (b) the bubble expands, and the liquid shells contract; (c) the bubble contracts, and the liquid shells dilate

shell is thinner than when the bubble is at equilibrium radius, so that the gradient across the shell is higher. Thus the rate of diffusion of gas towards and into the bubble is high. When the bubble is contracted, the liquid shells surrounding the bubble are expanded (*Figure 4c*). Although the concentration of gas near the bubble wall is higher than when the bubble is expanded (Henry's law), the increased thickness of the shell means that the concentration gradient is not as great as when the bubble is expanded. The two factors (gas concentration at the bubble wall, and shell thickness) work together when the bubble is expanded, but against one another when the bubble is contracted; on expansion, there is a large concentration gradient driving gas a short distance, and in the second case a lesser gradient drives the gas a longer distance. The former effect is dominant.

Therefore, not only is the area asymmetric in expansion and contraction, so is the diffusion rate; the two effects reinforce one another. The combined effect means that during stable cavitation in sufficiently intense fields (Crum[35] estimates for acoustic pressure amplitude $P_A \gtrsim 0.01$ MPa) the time-averaging bubble radius R_0 will increase.

Rectified diffusion of gas has far-reaching consequences: bubble nuclei can grow to provide intense cavitational activity, and small bubbles may grow to a resonant or threshold size. For example, if a bubble then fragments, the resulting small bubble nuclei may eventually grow to collapse themselves, so providing cavitation with a self-enhancing mechanism of positive feedback.

The development of the formulations used to characterize rectified diffusion is reviewed in the literature[51§4.4.3]. The transport of dissolved gas within a liquid is governed by Fick's law of mass transfer:

$$\frac{\mathrm{d}C}{\mathrm{d}t} = \frac{\partial C}{\partial t} + \mathbf{v}\cdot\nabla C = D\nabla^2 C \tag{2}$$

where D is the diffusion constant of dissolved gas within the liquid, C is the concentration of dissolved gas within the liquid and $\mathbf{v}$ is the liquid velocity at a point. To solve the mass transfer of gas between the liquid and a spherical bubble, requires solution of both Equation (2) and some suitable equation of motion for the bubble. However, such a solution is complicated by the fact that these two equations are coupled through the convective term $\mathbf{v}\cdot\nabla C$, and through the appearance of the gas pressure in the equation of motion. This pressure will vary in time as gas leaves the bubble in a manner governed by Fick's law, so that the equation of motion is dependent on Equation (2).

Solution of mass transfer between bubble and liquid, to describe either passive dissolution or rectified diffusion, therefore usually involves making approximations which reduce the coupling between Fick's law and the equation of motion.

Initial formulations[95–97] neglected the convective term in the diffusion equation. When the mass diffusion length $\sqrt{4Dt}$ of gas in the liquid[98] is small compared with the bubble radius, the convective term is not negligible. Thus, because of the time-scales involved, the approximation is justifiable with passive bubble dissolution, but not with rectified diffusion. Realizing this, Hsieh and Plesset[99] reinstated the convective time in the equation for mass transfer in a spherically symmetric frame. The threshold for rectified diffusion predicted by them is only valid at insonation frequencies far below the resonance, and predicts no additional effect due to the bubble resonance.

The key to much of the current work in rectified diffusion is the formulation for mass flux developed in 1965 by Eller and Flynn[38]. First, they found a solution for Fick's law applicable to any general oscillation $R(t)$, provided that the oscillation was in some way periodic. Their result depends on known parameters, and also on time averages of $R(t)$ and $R(t)^4$. The second part of the solution involves solving an appropriate equation of motion to calculate these time averages, which are then substituted in Eller and Flynn's solution to Fick's law. Several workers have since adopted this solution to Fick's law, completing the analyses by calculating the two time averages by applying suitable analytical or numerical techniques to a chosen equation of motion. The Eller–Flynn threshold predicts a smooth minimum in the threshold pressure for the onset of net growth by rectified diffusion, and a smooth maximum in the growth rate, both at the bubble resonance. This is a result of the pulsation amplitude being a maximum there. The results compare with the analytical solutions of Crum[35]. However, the analytical solutions cannot predict any additional effect of harmonics and subharmonics, which were subsequently shown by Church[40] to cause local minima in threshold, superimposed on the global minimum at the radius corresponding to the low-amplitude pulsation resonance (when $\omega = \omega_0$ and $R_0 = R_r$, the equilibrium bubble radius which is resonant with the acoustic frequency). In the absence of acoustic microstreaming and when there are no surface-active agents on the bubble surface (either of which could increase growth rates), good agreement with experiment may be obtained for the growth rate prediction of Church[40,41]:

$$\begin{aligned}\frac{\mathrm{d}R_0}{\mathrm{d}t} = \frac{DR_gT}{R_0p_0}C_{R_0}\left(1+\frac{4\sigma}{3R_0p_0}\right)^{-1} \\ \times\left(\langle R/R_0\rangle + R_0\sqrt{\frac{\langle (R/R_0)^4\rangle}{\pi Dt}}\right) \\ \times\left(\frac{C_\infty}{C_{R_0}} - \frac{\langle (R/R_0)\rangle}{\langle (R/R_0)^4\rangle}\right)\end{aligned} \tag{3}$$

where R_g is the gas constant ($\sim 8.314\ \mathrm{J\,K^{-1}\,mol^{-1}}$). The initial uniform concentration of gas dissolved in the liquid at $t = 0$ is given by C_∞. The concentration at the bubble wall when the wall passes through the equilibrium position (i.e. when $R = R_0$) is C_{R_0}. Church[41] observes that in equation (3), the moment from which time t is measured can be taken to be the instant the acoustic pressure exceeds the threshold for rectified diffusion[100]. Since this term appears as $\sqrt{1/t}$ in Equation (3), its effect is large at small times, but it becomes increasingly insignificant as $t \to \infty$, and so the $\sqrt{1/t}$ term was initially referred to as the 'transient term'. It may be interpreted as representing the initial depletion of gas from the shell surrounding a bubble growing by rectified diffusion (or, in the case of a dissolving bubble, the initial excess gas that has come from the bubble), $\sqrt{Dt}$ representing a diffusion thickness. Therefore, that whole term in Equation (3) may be interpreted for growing bubbles as the contribution to the growth from dissolved gas within a shell-like diffusion boundary layer just outside the bubble. The first term in that bracket might be considered physically to represent the contribution to growth from gas that resided in the liquid, outside the diffusion boundary layer. Once growth or dissolution begins, it will take a certain time to establish new equilibrium concentrations: once these are set up, there are steady-state conditions whereby there is a net gas flow into the diffusion boundary layer from the liquid reservoir outside, and from the diffusion boundary layer into the bubble. It is in the period up to the establishment of the new equilibrium that the so-called 'transient' term takes effect[41].

By employing these formulations to predict bubble behaviour, Church demonstrated that bubble histories which progress by rectified diffusion to end in transient collapse are not common in the biomedical frequency range 1–10 MHz. At 3 MHz, for an insonation pressure of 1 bar, 80% of cavities that undergo transient collapse do so because they lie above the transient cavitation threshold immediately at the start of insonation (corresponding to initial bubble radii of 1.03–1.15 μm). Only 20% of all cavities that will go transient do so by growing there through rectified diffusion (corresponding to initial bubble radii of 1.00–1.03 μm). Bubbles of initial radii 1.15–1.35 μm will grow by rectified diffusion to become trapped, stably cavitating, at a radius of 1.35 μm. Bubbles of initial radius greater than 1.35 μm will dissolve until they too become trapped at that radius. Bubbles of initial radius below 1.00 μm will dissolve away.

The likelihood of a bubble growing by rectified diffusion to transient collapse decreases with increasing frequency since the rectified diffusion threshold increases more rapidly with increasing frequency than does the transient cavitation threshold.

Church[40] similarly predicted that the contribution to violent cavitation of bubbles which grow by rectified diffusion to resonance radius R_r, and the collapse, is not common at these frequencies.

Although predictions of the acoustic pressure threshold for growth by rectified diffusion agree with observation, in general the measured growth rates are often much greater than the predicted values as a result of microstreaming, agreement only occurring when microstreaming is avoided. The reason for the increased growth rate is clear. As a bubble grows by rectified diffusion, the dissolved gas is taken from the liquid near the bubble. If there is no flow, then the rate at which the deficit is met depends on the rate at which dissolved gas can diffuse from regions farther out from the bubble. Since this is in general a slow process, the liquid outside the bubble wall will become depleted of dissolved gas. The resulting change in concentration gradient reduces the rate of further growth. However, microstreaming flows will tend to bring liquid from further out close to the bubble wall. The convection of dissolved gas reduces the depletion and increases the growth rate. Microstreaming will continually refresh the liquid at the bubble wall, giving it a dissolved gas concentration close to that found far from the bubble. The converse process is, of course, valid: if a bubble is dissolving, microstreaming will tend to remove from the region outside the bubble wall the excess dissolved gas concentration, so increasing the rate of dissolution.

Kapustina and Statnikov[101] considered the effect on the diffusion of microstreaming around a bubble that is fixed in space, whilst Davidson[102] separated the effects of rectified diffusion from the effect of acoustic microstreaming on mass transfer. As he did previously[40], Church[41] calculated the necessary time averages from computed periodic radius–time solutions to the Gilmore–Akulichev equation. These are employed in a reinterpretation of the formulation of Eller and Flynn[38], where the $\sqrt{1/t}$ term no longer represents the time since the acoustic pressure amplitude exceeded the threshold for growth, as it did in the absence of microstreaming, but is instead a function of the microstreaming velocity and the bubble radius. Referring back to the interpretation of the relevant terms in Equation (3) as representing contributions to growth from gas outside and within the diffusion boundary layer, Church[41] notes that the microstreaming flow, which brings in liquid containing the gas concentration found in the reservoir, will affect the diffusion boundary layer. A spatially non-uniform, dynamic equilibrium gas concentration is established. This has a higher average value, and is established more rapidly, than would be the case in the absence of microstreaming. Church renames the $\sqrt{1/t}$ term the 'decay' term, since it will always give a finite contribution in the presence of microstreaming, and because t is a decay time regardless of streaming. He confirmed that, when microstreaming is present, the circulation may enhance both the rate of growth by rectified diffusion and the rate of dissolution[41].

The illustration that microstreaming circulation about the bubble can have a dramatic influence on the rate of bubble growth through rectified diffusion suggests that not only is rectified diffusion important in the dynamics of bubble populations in sound fields through its ability to alter the radii of bubbles in that population, but also through the ability of one bubble to affect the growth of its neighbour. This can come about through the microstreaming flows induced by the first bubble impinging upon the second, in addition to the phenomenon whereby the local pressure field experienced by a bubble undergoing rectified diffusion will contain components of the pressure field emitted by its neighbour, and it is this local field (as stated above) which is the relevant one in the formulation of rectified diffusion.

Bubbles may affect the mass of gas contained within their neighbours, and therefore the distribution of equilibrium bubble radii within the population, through far more direct means than simply influencing the rate of rectified diffusion. It was stated above that neighbours can influence the directionality of the local sound field at another bubble. It was also illustrated how bubble involution and jetting may lead to bubble fragmentation and therefore convert a single bubble into a large number of much smaller entities (although it should be noted that diffusion of previously dissolved gas from the liquid into the bubble during the expansion phase of the bubble can, in extreme cases even during a single cycle[103], result in the net volume of gas contained within the bubble fragments being significantly larger than the gas contained within the initial bubble)[51§5.1.2]. The action of involution suggests the definition of a unique direction for the bubble jet, which may be instituted by the sound field (for example, through the direction of a jet propagation[64,104]) or by the medium (for example, through the presence of a solid boundary[105–107] or of bubbles[72,108]). The production of a jet through involution is an extreme form of shape oscillation commonly seen in bubbles. These may be oscillatory in nature, and amenable to expression through a summation of spherical harmonic perturbations[51§2.2.4c,§3.6]. A sufficiently pronounced perturbation can, at an extreme, cause bubble fragmentation[51§3.5.2]. *Figure 5* shows the disturbance of one bubble, A, which was initially undergoing stable cavitation, by the approach of a second bubble, B, which induces in A a shape oscillation resembling the superimposition of a second-order spherical harmonic perturbation. This is so extreme as to split bubble A in frame 6 into three fragments, C, D and E.

4. Alteration in the spatial distribution of the bubble population through radiation forces

In Section 2, the ability of an inhomogeneous bubble population to affect the sound field was discussed. However, a sufficiently strong sound field can in turn influence the bubble distribution, not only by altering the radii of existing bubbles (through rectified diffusion and fragmentation, as discussed in the previous section), but also by relocating individual bubbles.

In an accelerating liquid a gas bubble, being less dense than the surrounding fluid, will accelerate in the same direction as the surrounding liquid, but to a greater extent, in much the same way as a helium balloon behaves within an accelerating motor car[51§3.3.2d]. Such accelerations may be caused by an acoustic field, and being oscillatory in nature will induce oscillatory linear accelerations of the bubble. Consider for a first approximation a bubble undergoing stable cavitation in an acoustic pressure field which varies sinusoidally in time, the bubble pulsating at the single frequency inherent in the sound field. The local acceleration in the liquid will similarly vary sinusoidally, being always in one direction during the expansion phase of the bubble, and in the opposite direction during the contraction phase. The acceleration of the bubble in the direction of the fluid acceleration will be greater during the expansion phase than during the contraction since then the density of the gas contained within the bubble is less. Therefore, the net acceleration of the bubble will be in the direction of the fluid particle acceleration during the expansion phase of the bubble[5§3.3.1d]. If the bubble is assumed to be a simple harmonic oscillator, with one specific bubble size being resonant with the sound field, then bubbles having an equilibrium radius greater than that resonance radius (i.e. $R_0 > R_r$) will be pulsating in anti-phase to those bubbles which are less than the resonance radius (i.e. $R_0 < R_r$)[109]. The conclusion is therefore that in such a simple situation the bubbles of larger than resonance size will be accelerated in one direction by the sound field, and the bubbles that are smaller than the resonance size will be accelerated in the opposite direction.

In a plane standing-wave field, bubbles of less than resonance size are attracted to the pressure antinodes, and bubbles greater than resonance size travel to the pressure nodes, under the influence of the so-called primary Bjerknes force[51§4.4.1b,110,111]. Consider the pressure field $p(y, t)$ which varies with position y and time t as

$$p(y,t) = p_0 + 2\hat{P}_A \sin(ky)\cos(\omega t) \tag{4}$$

where p_0 is the static pressure, $k = \omega/c$ is the wavenumber and $\hat{P}_A$ is the acoustic pressure amplitude of the incident plane wave which, on reflection from a free or rigid boundary, sets up the standing-wave field. In this field the radius $R(t)$ of a bubble, whose resonance is ω_0, is assumed to oscillate linearly about an equilibrium radius R_0 such that

$$R(t) = R_0 - [R_{\varepsilon_0 a}\sin(ky)]\cos(\omega t - \vartheta) \tag{5}$$

where the phase factor ϑ equals zero for bubbles much smaller than resonance, and $\vartheta = \pi$ for bubbles much larger than resonance. The term $R_{\varepsilon_0 a}\sin(ky)$ represents the radial amplitude of oscillation, $R_{\varepsilon_0 a}$ being the amplitude of radial oscillation at a pressure antinode, and the $\sin(ky)$ dependence arising since the acoustic pressure amplitude experienced by the bubble varies as $\sin(ky)$ in the standing-wave field. The negative sign in equation (5) follows since a positive acoustic pressure causes a reduction in bubble volume when the two are in-phase. The amplitude of wall oscillation at the pressure antinodes [where $ky = (2n+1)\pi/2$, where n is an integer] is given by

$$R_{\varepsilon_0 a} = \frac{2\hat{P}_A}{R_0\rho}\frac{1}{\sqrt{(\omega_0^2 - \omega^2)^2 + (2\beta_{tot}\omega)^2}} \tag{6}$$

where $R_{\varepsilon_0 a} \ll R_0$, and where β_{tot} is a damping parameter associated with the bubble oscillation. If ρ and c refer to the density and sound speed in a liquid, then this bubble will experience a radiation force of magnitude

$$\bar{F}_{B1} = \frac{4\pi R_0 \hat{P}_A^2 \sin(2ky)}{\omega\rho c}\frac{(\omega_0/\omega)^2 - 1}{[(\omega_0/\omega)^2 - 1^2]^2 + (2\beta_{tot}/\omega)^2} \tag{7}$$

aligned with the y-axis. The limiting forms of Equation (7) show that for bubbles smaller than resonance ($\omega_0 > \omega$) the primary Bjerknes force is of magnitude

$$\bar{F}_{B1} = \frac{3P_A k V_0 \sin(2ky)}{2R_0}R_{\varepsilon_0 a} \qquad (\omega_0 > \omega;\quad R_0 < R_r) \tag{8}$$

and attracts bubbles up pressure gradients towards pressure antinodes. Bubbles of larger than resonance size ($\omega_0 < \omega$) will experience a force

$$\bar{F}_{B1} = -\frac{3P_A k V_0 \sin(2ky)}{2R_0}R_{\varepsilon_0 a} \qquad (\omega_0 < \omega;\quad R_0 > R_r) \tag{9}$$

directed down the pressure gradient (i.e. towards the pressure node). This migration of the various bubbles types can have an important influence on cloud cavitation. It is most often bubbles of smaller than resonance size which are associated with high-energy cavitation effects such as sonochemistry and sonoluminescence, and the formation of such regions of high-energy cavitation can cause localization of the effects to specific parts of the sound field[112,113] (*Figure 6*). Similarly, bubbles larger than resonance size may be associated with geometric shielding of the sound field[51§4.2d,114]. Disturbance of the sound field can disperse these bubble aggregations which may be associated with either very high or very low activity, and cause more unified sample treatment. Such disturbance may be achieved by sweeping the frequency to alter the modal standing wave patterns[115], by scanning or rocking

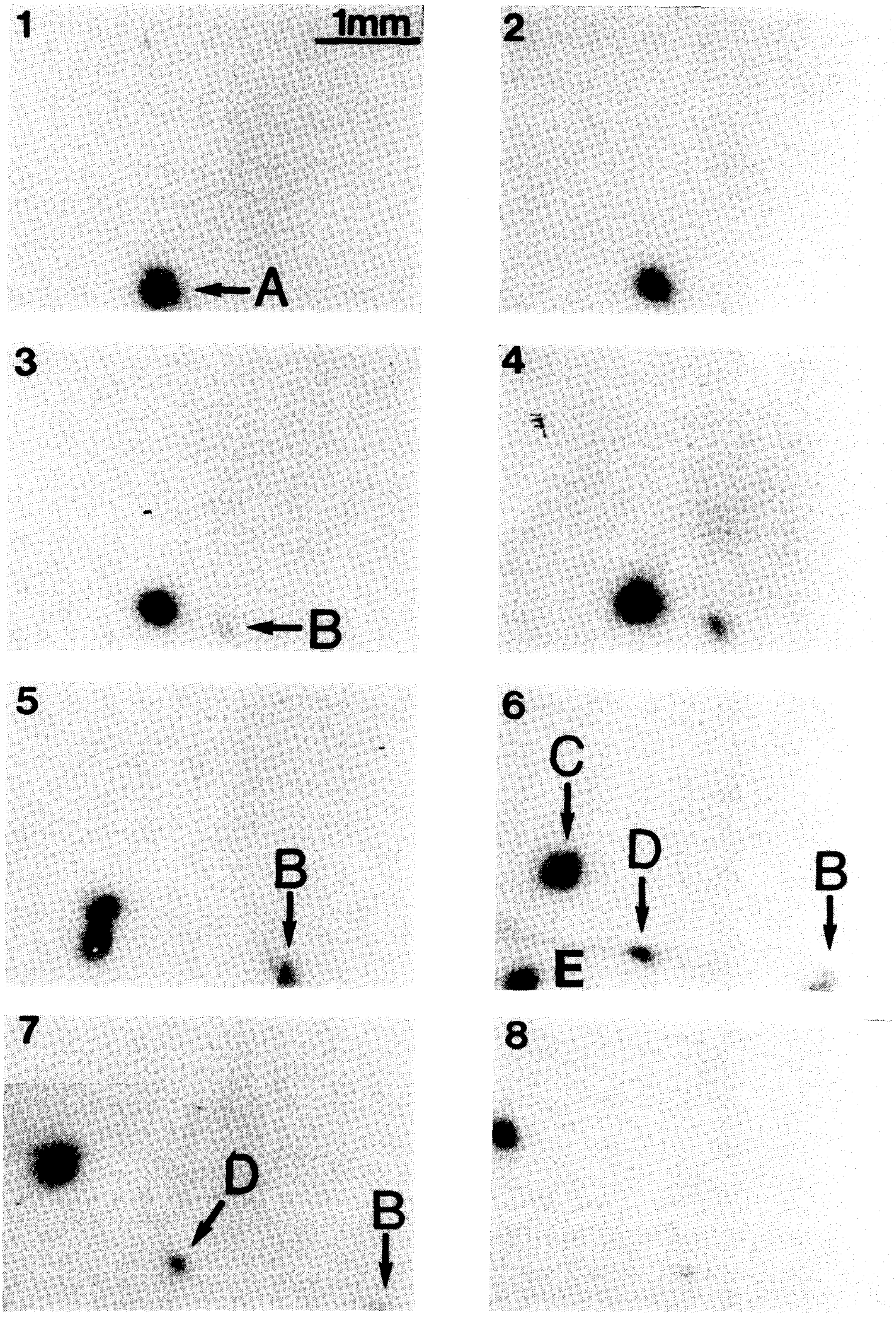

Figure 5 Eight frames selected from a film shot at 2000 frames s^{-1}. Bubble A is initially oscillating stably. The approach of a similar bubble (B) disturbs the sound field around A. Violent shape oscillations in A, reminiscent in frame 5 of a second-order spherical harmonic perturbation, cause it to break up (frame 6) into bubble fragments, labelled C, D and E. These, and B, disperse rapidly. Acoustic pressure amplitude, 2 atm; acoustic frequency, 10 kHz (since the framing rate is less than this, no information on individual bubble pulsations can be deduced from images)

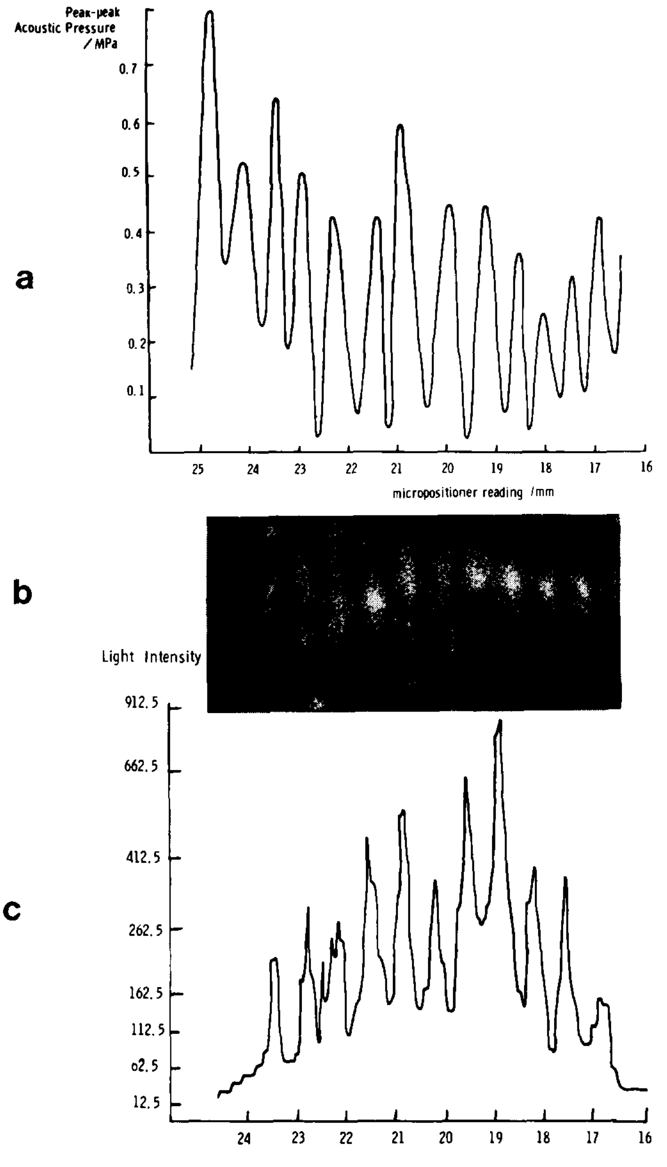

Figure 6 A partially standing wave field is set up in tap water at 22 °C by reflecting a 1 MHz physiotherapeutic ultrasound beam. (a) Peak-to-peak acoustic pressure as measured with the needle hydrophone; (b) photograph of the appearance of the corresponding sonoluminescence as seen through the intensifier; (c) light intensity (as measured in relative units); (a), (b) and (c) have a common abscissa. Nominal intensity setting on Therasonic device = 3.3 W cm^{-2}. After Leighton *et al.*[113]. Reprinted by permission from *Physics in Medicine and Biology* (1988) **33** 1239–1248; Copyright © 1988 IOP Publishing Ltd

the sound field[113], by 'tweaking' the sound field (by frequency alteration or beam rotation[116]) or by pulsing the sound field[51§5.3.1,117–121] or rotating the sample[51§5.3.2,122,123]. Alternatively, the localization of activity within the sample to specific regions may be exploited to cause the effect at one specific location[124]. It should be noted that inhomogeneities in the sample (e.g. solid particles, droplets) will also be subject to radiation forces and may accumulate near to, or far from, the localized cavitation[125–127] in an acoustic field containing pressure gradients (e.g. standing-wave, focused, modal).

Bubble localization and migration as described above (and with particular reference to rotation studies[122,123]) may occur not only in standing-wave conditions, but also in travelling-wave conditions, where similar radiation forces exist. The radiation force on a bubble, which is assumed to oscillate linearly with amplitude R_{ε_0} as

$$R = R_0 - R_{\varepsilon_0} \cos(\omega t - kz - \vartheta) \tag{10}$$

in a planar travelling wave field having a pressure p of

$$p = p_0 + P_A \cos(\omega t - kz) \tag{11}$$

is given to the same approximation by

$$\begin{aligned}\bar{F}_{\text{trav}} &= \frac{3}{2}\frac{P_A^2 V_0 k}{R_0^2 \rho \omega^2} \frac{2\beta_{\text{tot}}/\omega}{[(\omega_0/\omega)^2 - 1]^2 + (2\beta_{\text{tot}}/\omega)^2} \\ &= \frac{3}{2}\frac{P_A^2 V_0 k}{R_0^2 \rho \omega_0^2} \frac{2\beta_{\text{tot}}/\omega}{[1 - (\omega/\omega_0)^2]^2 + (2\beta_{\text{tot}}\omega/\omega_0^2)^2}\end{aligned} \tag{12}$$

where $V_0 = 4\pi R_0^3/3$ is the equilibrium bubble volume. This takes a value of

$$\bar{F}_{\text{trav}} = \frac{3}{4}\frac{P_A^2 V_0 k}{R_0^2 \rho \omega_0 \beta_{\text{tot}}} \qquad (\omega = \omega_0; \quad R_r = R_0) \tag{13}$$

for resonant bubbles in a travelling-wave field, which is numerically equal to the ratio of the mean acoustic power input to the speed of sound in the medium[128,129].

Formulations also exist for the radiation force between two pulsating bubbles,[51§4.4.1c,130–134] the so-called *secondary* or *mutual* Bjerknes force. However, the general rule that bubbles which are both less than, or both greater than, resonance size attract, whilst the force is repulsive if one bubble is greater than and the other less than resonance size, is a simplification. The results may be more complicated if the bubble population density is high or the incident sound field is strong. Such attractions can influence the size of individual bubbles by causing coalescence or, through the surface waves induced on one bubble by the proximity of its neighbour, fragmentation. *Figure 5* showed such a neighbour-induced fragmentation, whereby the original bubble undergoes an extreme shape oscillation and fragments into a number of similar-sized daughter bubbles. A second form of fragmentation[135] is shown in *Figure 7* whereby many bubbles much smaller than the mother bubble (which remains relatively intact, and is labelled A) are generated from the surface waves on the bubble wall[136]. Gas is lost from the mother bubble through such bubble generation, but is transferred to a neighbour bubble (B) as it coalesces with the fragments, which are attracted to it through a mutual Bjerknes interaction[51§4.4.1b,c]. Bubble gas content may also be influenced by radiation forces as neighbouring bubbles set up microstreaming circulations, or deplete the local liquid of dissolved gas, so influencing rectified diffusion. If the bubble is oscillating close to a boundary (e.g. solid or free surface), migrations may be influenced through secondary Bjerknes interactions with the appropriate image sources, and by disturbance to the flow. Crevices in solids may attract bubbles, the subsequent expansion of which can extend the crevice[51§5.4.1,137].

Coakley and Nyborg[128] give, for the radiation force on a particle of volume V_p and density ρ_p close to a pulsating bubble,

$$\bar{F}_{\text{part}} = -\frac{3V_p(\rho_p - \rho)}{2\rho_p + \rho}\frac{\rho\omega^2 R_{\varepsilon_0}{}^2 R_0^4}{r^5} \tag{14}$$

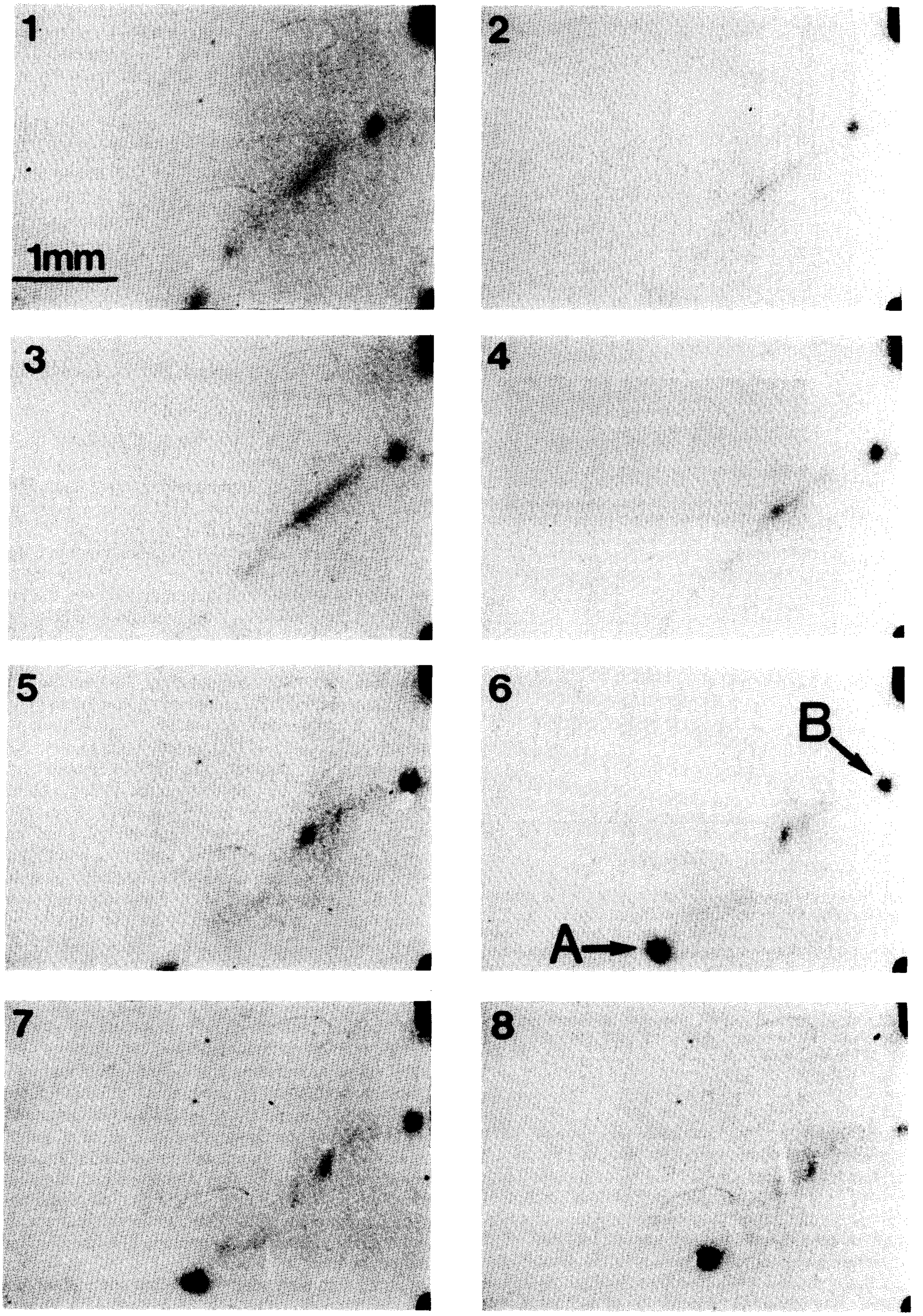

Figure 7 Eight frames selected from a film shot at 2000 frames s^{-1} showing a microstreamer of tiny bubbles propelled between two larger bubbles by radiation forces. The source is the bubble labelled A. Bubble fragments are sheared off from the surface of A; they travel in the microstreamer under the influence of the mutual Bjerknes force, to coalesce with the bubble marked B. Acoustic parameters as for *Figure 5*

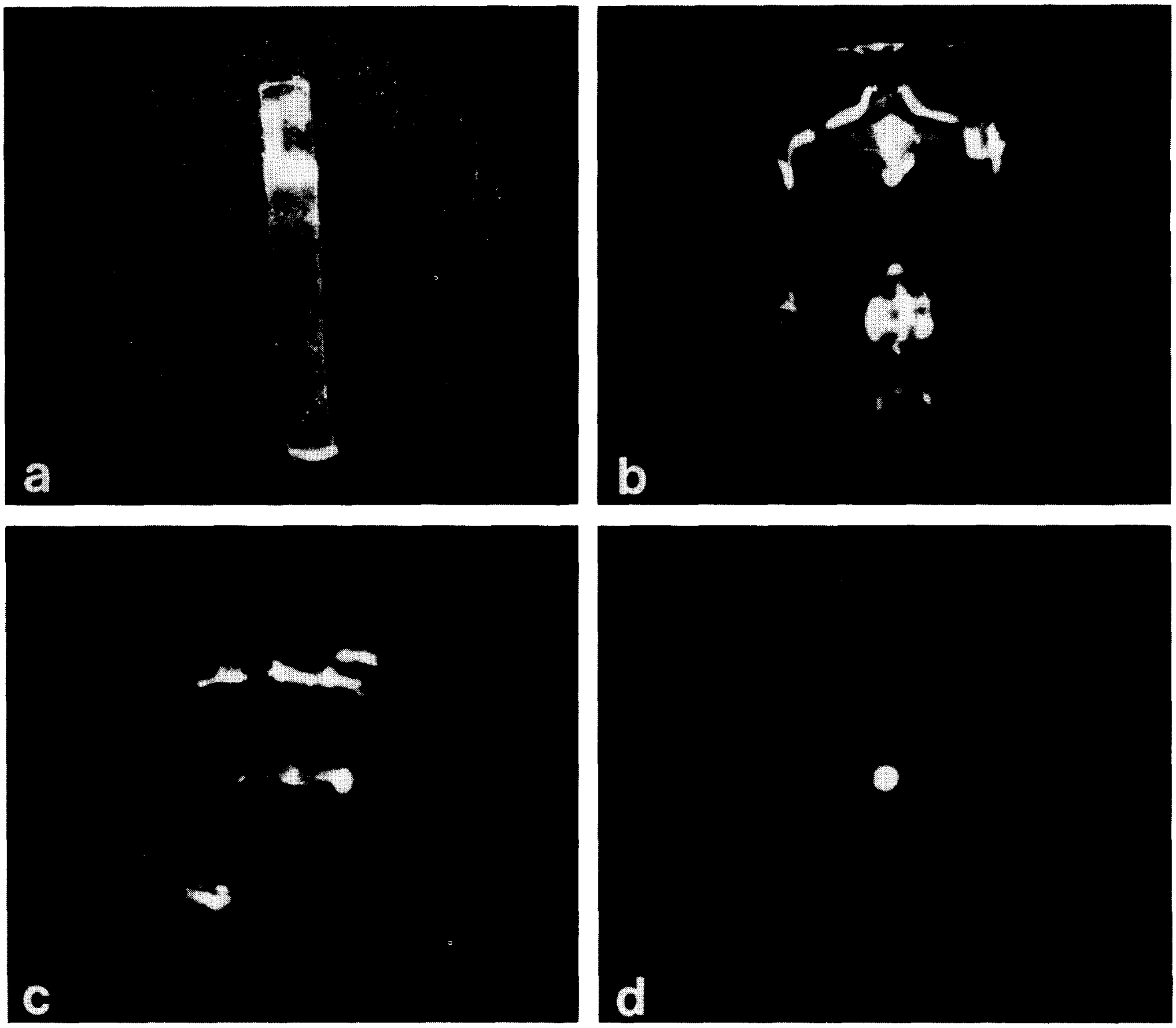

Figure 8 Image-intensified photograph of sonoluminescence in tap water, insonated at 20 kHz by an ultrasonic probe. (a) A Kundt's tube experiment showing the presence of bands of strong and weak sonoluminescence emission. (b) Looking in a horizontal direction into a rectangular tank of water; the ultrasonic probe enters the liquid at top centre. (c) As (b) but viewed from underneath. (d) As (c) but at high power levels. Intensive cavitation at the probe tip, as indicated by localized sonoluminescence, effectively acoustically shields the bulk liquid, where no sonoluminescence is seen to occur. After Walton and Reynolds[52]. Reprinted by permission from *Advances in Physics* (1984) **33** 595–660; Copyright © 1984

if the sound frequency ω is taken to be close to the bubble resonance. This force falls off rapidly with distance r from the bubble wall, where it is the maximum. Coakley and Nyborg calculate that in a 1 MHz sound field of pressure amplitude 0.05 atm, a bubble oscillating such that $R_\varepsilon = R_0/10$, where $R_0 = 3.3\ \mu$m, will collect within 3 ms from the start of insonation all the platelets suspended in a saline solution within the region $R_0 < r < 2R_0$. Close to a bubble, microstreaming circulations may alter the distribution of neighbouring inhomogenities[51§4.4.5,138,139]. In all the formulations of the radiation forces discussed in this section, the bubbles are assumed to be driven into low-amplitude simple harmonic pulsation; however, non-linear oscillations are to be expected in most of the active bubbles in applications involving sonochemistry, erosion, etc.

As discussed above, acoustic radiation forces can produce aggregations of bubbles which may cause regions of high activity in some parts of the sound field, and low in others. This can limit the amount of sample which is exposed to effects of cavitation. The mechanisms by which sample rotation and acoustic pulsing may cause more thorough treatment of the sample are complicated[51§5.3] and can involve the rise-time, saturation, and persistence of the 'activity' of the system[117]; the repeated cycling of a bubble through resonance size as it grows by rectified diffusion and subsequently dissolves when the sound is pulsed[118]; the survival of unstabilized nuclei to seed further cavitation events[12,121]; excitations associated with the start of insonation[140]; and with the local degassing of liquid[114,141]. Shielding the sound field, as discussed earlier, may also be involved[114,141,142]. Shielding of the sample from the sound field may also occur not just at pressure nodes but also very close to an ultrasonic transducer: as the intensity is increased, the cavitational activity in the body of the sample first increases and then decreases, as intense cavitation induced at the surface of the transducer prevents the sound penetrating into the body of the liquid[52] (*Figure 8*). In addition, radiation forces may play a more active role through the high speed of translations of the bubble which they induce. Not only will this tend to mix the sample (and the bubbles, dissolved gases, chemicals, etc.), but also the bubbles may themselves generate a direct effect. The hydrodynamic forces resulting from such rectilinear motion can generate shear stresses capable, for example, of damaging cells[123]. In addition, such transitions can be exploited by coupling the rotation of the sample with the pulsation of the sound field to sweep the bubble repeatedly through the sample; the local cavitation effects (e.g. shocks, chemical species, hot spots) associated with the bubble will therefore be distributed through a greater region of the sample, creating an effect in addition to that resulting from the stresses induced by the motion[122]. Radiation forces might cause transport of the seed bubble nuclei[143-145] for cavitation inception to regions of high acoustic intensity (e.g. foci, pressure antinodes). Such nuclei transport might also be achieved through acoustic streaming[146,147] or stirring[148], although the latter might also introduce tribonucleation[149].

Other factors, of course, in addition to radiation forces, can influence the bubble population distribution. The largest bubble population of all, the oceanic one, is distributed through a variety of mechanisms, including turbulence, buoyancy and Langmuir circulation[51§3.8,150-152]. However, in the intense continuous-wave or tone-burst systems operated in many power ultrasound devices employed for erosion, cell disruption or sonochemistry, it is often the acoustic forces which dominate.

5. Conclusions

The size of the bubble and the nature of the *local* sound field determine whether the bubble oscillations are aspherical or spherical; and, if spherical, whether transient or stable; and, if stable, determine how the bubble grows and migrates.

Rectified diffusion may alter the radii of bubbles present within the population, and therefore their behaviour (stable, transient, migratory, involuting, etc.). Radiation forces may affect the distribution of these bubbles and therefore the local sound field experienced by the bubble. This will in turn affect the bubble activity (mass flux, collapse, etc.). However, the nature of the redistribution of bubbles, and their activity once distributed, depend critically on both the local field and the bubble size. It is the complexity of these interactions, which are dependent both on the current status and the cavitation history of the sample, which gives rise to the rich variety of mechanisms and effects which can be observed.

Acknowlegements

I am very grateful to Professor Ron Roy, who presented this paper on my behalf to the fourth meeting of the European Society of Sonochemistry, and to Sue Hellon, who kindly assisted when I was similarly incapacitated, in preparing the photographic material for presentation.

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