

CHAPTER 11

AN INTRODUCTION TO ACOUSTIC CAVITATION

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11.1. THE ACOUSTIC PROPERTIES OF THE BUBBLE

Perhaps the simplest of the interactions between a gas pocket in a liquid, and an acoustic wave in that liquid, is that of geometrical scattering. This occurs when the wavelength of the acoustic wave is much less than the bubble radius. Sound is scattered because of the strong acoustic impedance mismatch between the gas and the liquid, and the bubble casts an acoustic shadow. In practice the highest frequencies employed in liquids tend to be 30 MHz (giving a wavelength in water of around $50 \mu\text{m}$), and the wavelength even at 1 MHz is 1.5 mm. As such, true geometrical scattering would occur only for bubbles which are so large as to have an important implication: bubbles of a smaller size are likely to be more common in the population. While the scattering of sound from bodies which are not much larger than the wavelength differs from that described above (Morse and Ingard 1986), an even more important factor is this: that smaller bubbles tend to behave less like the inert bodies described above, whose walls act as simple reflective boundaries for the sound, and more as oscillators which can be driven by the sound field to pulsate. Such pulsation enhances the coupling between the sound field and the bubbles. This not only dramatically changes the scattering of sound but also causes a range of other effects which broadly characterise acoustic cavitation.

11.1.1. Stiffness and inertia

An oscillator requires two key elements: stiffness and inertia. In the bubble system, the first of these is provided by the gas. If compressed, it exerts

a force which resists that compression and would tend to make the bubble expand and vice versa. Potential energy is stored in the gas as the bubble volume changes. When the bubble wall moves, the surrounding liquid must also move. If the system has spherical symmetry, then the velocity of an incompressible liquid falls off as an inverse square law away from the bubble wall (Minnaert 1933). Therefore there is a kinetic energy associated with bubble pulsations which is invested in the moving matter. Since the liquid is so much denser than the gas, this is primarily invested in the liquid (though motion of the gas contributes to a much smaller extent; Leighton *et al* 1995a). Comparison of the potential and kinetic energies (which is a comparison of the gas stiffness with the liquid inertia) allows the formulation of the natural frequency f_r of the oscillator. A simple calculation, based on the linear pulsations of a spherical air bubble of radius R_0 in water which is assumed to be incompressible and inviscid, gives

$$f_r R_0 \approx 3 \text{ Hz m} \quad (R_0 \gtrsim 10 \mu\text{m}) \quad (11.1)$$

an approximation which neglects surface tension, making this formulation less valid for smaller bubbles.

11.1.2. Resonance

When a bubble pulsates in a sound field, it loses energy through viscous and thermal mechanisms, and through the radiation of acoustic waves into the liquid (Devin 1959). The existence of a natural frequency and damping such as this means that when driven by a sound field, the bubble can exhibit a resonance at a frequency similar to that given in equation (11.1). This resonance possesses properties in common with others. Bubbles just larger than the size which is resonant with the sound field will pulsate in antiphase to those just smaller than resonance size; the amplitude of pulsation tends to be largest close to the resonance condition (Leighton 1994a, §4.1). However the bubble possesses a complex range of other properties associated with the resonance, which will be detailed in sections 11.2 and 11.3. In brief, as the wall pulsation amplitude increases near resonance, the nonlinear character of the oscillation becomes more pronounced, and the resultant bubble activity can be divided into two classes. In the first category are those effects, such as the emission of harmonics of the driving frequency, which increase continuously with increasing pulsation amplitude. Therefore if the bubble is resonant at a driving (or 'pump') acoustic frequency of f_p , then the emission of signals at $2f_p$, $3f_p$, etc may be taken to indicate that the bubbles are close to resonance size (Miller *et al* 1984, Leighton 1994b). This can be used as a method of sizing bubbles. However it has drawbacks in that phenomena other than resonant bubbles can give rise to harmonic emissions, and for such sizing purposes, a two-frequency technique may be superior (Leighton

et al 1997, Phelps *et al* 1997). A second class of phenomenon occurs as a threshold with the increasing pulsation amplitude, such as the stimulation of surface waves on the bubble wall† (Phelps and Leighton 1996, 1997). While the two examples (harmonic emission and surface wave activity) reflect the dynamics of single bubbles, phenomena which have strong implications for the bubble population as a whole can be found in these two classes (respective examples of radiation forces and rectified diffusion are discussed in section 11.3). The most important of the threshold phenomena, historically studied in populations but with recent important findings through observation of single bubbles, is inertial cavitation, described in the following section.

11.1.3. *Inertial cavitation*

In 1917 Lord Rayleigh provided a theoretical analysis for the phenomenon we now call inertial cavitation. The impetus for this study had come from the problem of the erosion of ship propellers, and consideration was given to the possibility that gas bubbles, excited by the pressure fluctuations generated close to the propeller, were responsible for the 'pitting' seen on the blades. Inertial collapse of a spherical gas bubble in a liquid is characterised by a relatively slow (i.e. timescales of the order of half an acoustic cycle), approximately isothermal growth of an initial bubble nucleus to many times its original size. This is followed by a rapid collapse, the initial stages of which are dominated by the inertia of the spherically converging liquid. During the collapse, the gas temperature rises as it is compressed, and shocks can propagate within the gas. Both high temperatures (Neppiras 1980) and shocks (Bradley 1968) can potentially generate free radicals, the subsequent radiative recombination of which can emit light. Light emission from collapsing bubbles has been observed, and is called 'sonoluminescence'. The production of this emission is sometimes taken as indicative of (but not necessarily requisite for) the occurrence of inertial cavitation, and it is perhaps in this that the main practical exploitation of the phenomenon of sonoluminescence is currently realised. This is because many, but by no means all, of the physical and biological changes which ultrasonic cavitation can induce occur as a result of inertial cavitation (section 11.4).

However, a second, and some would say more important, reason for considering sonoluminescence is its ability to provide experimental evidence against which to test theories of bubble dynamics. In recent years there has been a significant increase in the number of publications supporting one mechanism or another for the generation of sonoluminescence, and the topic remains controversial (Leighton 1994a, §4.2.1). Throughout the 60 years of investigation to date into sonoluminescence, the proposed mechanisms by

† With associated implications for nucleation and microstreaming, which will be discussed in section 11.3.

which the emission is produced have been debated. A range of mechanisms has been proposed, and although there have been several periods where the broad consensus of opinion favours one, these have not lasted. Many people in the 1980s favoured a thermochemical mechanism (Walton and Reynolds 1984), proposed by Griffing (1950, 1952). In this, oxidising agents such as hydrogen peroxide are formed by the high temperatures (~ 5000 K; Suslick *et al* 1986) within the compressed gas, and give rise to chemiluminescent reaction. However, there has been support for over 50 years for mechanisms based on electrical discharge (Frenzel and Schultes 1934, Frenkel 1940, Bresler 1940), though details of the mechanism have changed (Margulis 1985, Lepoint *et al* 1994). Jarman (1960) proposed gas shocks as the source of the emission, and variations of this theory too have survived to present (Vaughan and Leeman 1989, Wu and Roberts 1993, Moss *et al* 1994). However other mechanisms proposed in the early days currently have very few supporters (Chambers 1936, Weyl and Marboe 1949, Gunther *et al* 1959). Replacing these in recent years have been publications introducing into the generation of sonoluminescence the phenomena of Casimir energies (Schwinger 1992); rectified diffusion (Crum and Cordry 1994); molecular collisions (Frommhold and Atchley 1994); quantum radiation (Eberlein 1996); and jetting (Prosperetti 1997).

Perhaps the main reason for the increase in recent years in publications on the proposed mechanisms for the emission was the discovery that it was possible to produce a sonoluminescing bubble which behaved quite differently to those envisaged previously by most people. After reaching minimum size, the bubble rebounds, emitting a pressure pulse into a liquid. Until recently, the bubble was then thought to fragment, and as such the phenomenon we now call 'inertial cavitation' was until recently termed 'transient cavitation'. However, Gaitan and Crum (1990) discovered sonoluminescence over measurement intervals of thousands of acoustic cycles from repeated collapses of a single bubble which apparently† did not break up. The discovery that the sonoluminescent flash in such circumstances is of less than 50 ps duration (Barber *et al* 1992) throws into question the mechanism by which light is generated in this 'single-bubble sonoluminescence' (SBSL). However in practical applications involving the cavitation of many bubbles, the breakdown of spatial symmetry might suggest that each bubble performs such inertial collapses once or only a few times and then fragments on rebound. In such 'cavitation-field sonoluminescence' (CFSL) or 'multibubble sonoluminescence' (MBSL) it may be that the same or another of the various mechanisms for generating light from bubble collapse dominates, depending on the details of the collapse. Currently the most widely accepted theory for CFSL is based on

† Barber *et al* (1992) did not observe fragmentation and re-coalescence occurring during each acoustic cycle, though it is possible that this might occur over timescales too rapid to measure.

the recombination of thermally generated free radicals (Kamath *et al* 1993). There are, however, differences between the predictions of this theory and recent measurements of CFSL emissions (Matula *et al* 1997). The authors suggest an upper limit on the pulse width during CFSL of 1.1 ns.

As mentioned at the close of section 11.1.2, the occurrence of inertial cavitation is a threshold phenomenon. Whether it occurs or not, for a free-floating spherical bubble nucleus of known gas content in a given liquid, depends primarily on the acoustic pressure amplitude, the acoustic frequency, and the size of the nucleus. If the nucleus is too small, then surface tension forces prevent the initial sudden growth, and inertial cavitation does not occur. If it is too large, then the bubble may grow, but be too 'sluggish' to concentrate the energy sufficiently on collapse to generate free radicals etc†. There is therefore a critical size range in which, for a given sound field, the initial size of the bubble must fall if it is to nucleate inertial cavitation (Flynn 1964, Flynn and Church 1984, Holland and Apfel 1989, Leighton 1994a). The lower the frequency, the wider this range.

This is clearly shown in figure 11.1, where the transition threshold between inertial and non-inertial cavitation is plotted, based upon calculations by Apfel and Holland (1991). They assumed that, in response to a single cycle of ultrasound, a bubble which is spherical at all times, would grow; and upon subsequent adiabatic collapse, the gas within the bubble should attain a temperature of at least 5000 K if the collapse is to be 'inertial' (Apfel 1981a,b, Apfel and Holland 1991). Though there are clear approximations and the choice of such a criterion for defining inertial cavitation is not fundamental, this is nevertheless an extremely useful calculation. It illustrates that the acoustic pressure amplitude required to cause a bubble to undergo inertial cavitation is dependent upon the initial radius of that bubble. Since in most applications the frequency is the easiest to control of the three parameters shown in figure 11.1, followed by the acoustic pressure amplitude *at the bubble*, with the radii of the nuclei present being the least accessible, then the graph can be interpreted in another manner. At a fixed frequency, say 10 MHz, an ultrasonic cycle with a peak negative pressure of 1.5 MPa (assumed to be constant throughout the field) will only generate inertial cavitation within a water sample if, according to this model, it contains bubbles having radii between 0.03 and 0.77 μm . As the pressure amplitude decreases, so does the range of bubble sizes which can nucleate inertial

† A simple catapult analogy is informative. Bubbles undergoing inertial cavitation tend to grow, prior to collapse, to a roughly similar size (Leighton 1994a, §4.2.3, §4.3.1b(iii)). This is like trying to achieve the same length of draw (i.e. about one arm's length) on a number of catapults. If the elastic on the catapult is too short, then it is not possible to draw a full arm's length (if the full draw were only a few centimetres, the throw of the catapult would be very short). This is similar to the case of the very small bubble, where surface tension prevents bubble growth. If the elastic in the catapult were, at equilibrium, very long, it would be easy to draw a full arm's length, but the elastic would not be taut, and again the throw would be short. This is akin to the case of the large bubble.

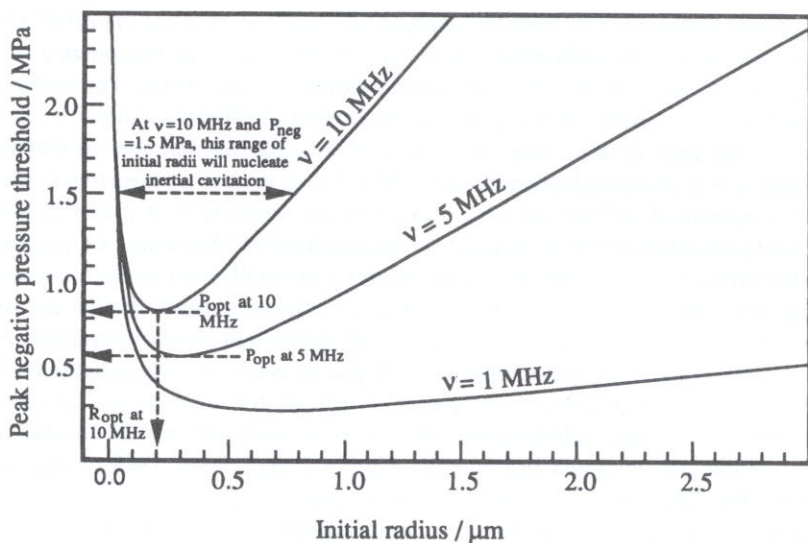


Figure 11.1. *The threshold for inertial cavitation, as predicted by the theory of Apfel and Holland. For each frequency a line can be plotted: if the conditions of peak negative pressure and of the initial bubble radius are such that the point of interest on the graph lies below the line, non-inertial cavitation will occur. If the point of interest is above the line, inertial cavitation will occur. Reprinted by permission of Elsevier Science from 'Gauging the likelihood of cavitation from short-pulse, low-duty cycle diagnostic ultrasound' by R E Apfel and C K Holland, *Ultrasound in Medicine and Biology*, vol 18, pp 267–81, copyright 1991 by World Federation of Ultrasound in Medicine and Biology.*

cavitation. The very lowest peak negative pressure which could give rise to inertial cavitation, according to this model, is p_{opt} (around 0.84 MPa at 10 MHz), and at this pressure only bubbles of a radius R_{opt} ($0.2 \mu\text{m}$ at 10 MHz) could possibly nucleate inertial cavitation.

The theory of Apfel and Holland has given rise to a measure called the 'mechanical index', which may be used to indicate the likelihood of exceeding the threshold required to nucleate cavitation. As described for figure 11.1, the model demonstrates that for a given acoustic frequency there is a minimum in the curve. If the whole range of bubble size classes are present, then there is a specific initial bubble size for which the threshold acoustic pressure required to nucleate inertial cavitation is a minimum. As the insonation frequency increases, the bubble radius which requires minimum pressure to nucleate inertial cavitation decreases. This is because inertial and viscous forces increase with increasing frequency, and there is insufficient time to bring about the required amount of bubble growth. For the same reason the acoustic pressure required to nucleate inertial cavitation in all but

the smallest bubbles increases with increasing frequency. Surface tension dominates the response of the smallest bubbles.

If one is interested in a worst case assessment of the likelihood that inertial cavitation will occur when a liquid is insonated, clearly one must assume that the bubble population contains bubbles at the radius corresponding to the minimum in the threshold curve. At a given frequency, it is bubbles of this radius, R_{opt} , which the analysis predicts will require the smallest peak negative pressure, p_{opt} , to undergo prompt inertial cavitation in response to a single acoustic cycle. Apfel and Holland (1991) generated a plot of p_{opt} against frequency for water and whole blood, using pure fluid bulk property values for the surface tension, density and viscosity relevant to the two fluids. The liquids are assumed to contain the relevant nuclei at size R_{opt} . Apfel and Holland employed a two parameter least-squares fit to these plots in order to obtain a relationship between p_{opt} and insonation frequency $f = \omega/2\pi$. They found a least-squares fit of:

$$\frac{p_{\text{opt}}^a}{f} = b \quad (11.2)$$

where if p_{opt} is measured in MPa and f in MHz, the constant a takes values of 2.10 for water and 1.67 for blood, and b has values 0.06 for water and 0.13 for blood. For a given sound field with a maximum negative pressure of p_r , then by taking a value of $a \approx 2$ to approximate the appropriate physiologically relevant liquid, a mechanical index MI can be defined for the sound in that liquid:

$$\text{MI} = \frac{p_r[\text{MPa}]}{\sqrt{f[\text{MHz}]}} \quad (11.3)$$

The mechanical index MI for prompt cavitation represents an approximate measure of the worst case likelihood of nucleating inertial cavitation. As such it can be used to estimate the potential for nucleating inertial cavitation resulting from insonation by diagnostic ultrasound. Clearly it would be less appropriate to apply this index to tone-burst or continuous-wave insonations, during which a range of complicated processes, including rectified diffusion and enhancement of the cavitation by ultrasonic pulsing, can occur (section 11.3; Leighton 1994a, §4.4.3, §5.3). Holland and Apfel recommend that the pulse length should not exceed 10 cycles, nor the duty cycle 1:100. The peak negative pressure output from the device, as measured in water, must be derated to give the appropriate peak negative pressure that would be attained *in vivo* at the location of the maximum pulse intensity integral (AIUM/NEMA 1998). The centre frequency is used for f which, for accuracy, is expected to be of the order of MHz. Apfel and Holland (1991) suggest that a mechanical index value below $\sqrt{0.5} \approx 0.7$ would indicate that, even in the presence of a broad size distribution of nuclei, the conditions are not sufficient to allow significant bubble expansion. If $\text{MI} \geq \sqrt{0.5}$, Apfel

and Holland suggest that 'the user should be advised of the potential for bubble activity'. The AIUM, NEMA and FDA have adopted the mechanical index as a real-time output display to estimate the potential for cavitation *in vivo* during diagnostic ultrasound scanning (AIUM/NEMA 1998). The use of MI related to practical exposure measurement procedures is discussed in Chapter 7.

Key points should be noted. First, the model for the index is based on the assumption of a free-floating spherical nucleus of optimum size. In certain circumstances it may be that the nucleus is of a different type (section 11.3.3). Second, the mechanical index gauges the likelihood of prompt cavitation, and nothing more: the effect of interest (e.g. a bio-effect) may be related to some other mechanism (section 11.4). Third, the effects of nonlinear propagation (Leighton 1994a, §1.2.3) are not included, and hence the index might underestimate conditions *in situ*. Fourth, when applied to diagnostic ultrasound instruments, the mechanical index describes conditions only at the focus, which is not necessarily the point of interest. Fifth, the mechanical index has arisen from a theory which gives smooth curves of the form shown in figure 11.1; such curves may in fact show peaks when other effects are incorporated (Roy 1996, Allen *et al* 1997).

Lastly, the underlying theory is applied to derive the mechanical index in a way intended to elucidate the conditions which attain the threshold for nucleating inertial cavitation. The amount by which the mechanical index is exceeded is therefore only a guide to the degree of cavitation activity, and by no means an exact predictor. Consider the MHz range illustrated in figure 11.1, where the range of nuclei size which can seed inertial cavitation is relatively narrow. In such a sound field of fixed frequency and increasing acoustic pressure, as one exceeds the threshold in figure 11.1, the range of nuclei which may nucleate inertial cavitation increases. In a field containing a broad range of bubble sizes, with a uniform number of bubbles in each size class, the total number of nucleated events would be expected to increase. However if there is only a narrow distribution of bubble sizes, exceeding the threshold by increasing amounts in this manner would, to first order, have little effect on the *number* of inertial events which are nucleated. What might be expected to increase is the energy associated with each collapse. Similarly, as the acoustic frequency changes, it is no simple matter to predict how the mechanical index might correlate with the amount of cavitation activity observed (Leighton 1997).

11.2. TYPES OF CAVITATION

In section 11.1 an isolated spherical bubble was discussed. Such a bubble can cast a geometric acoustic shadow in an ultrasonic field of sufficiently high frequency. This could, for example, monitor the slow dissolution one might expect of such a bubble (figure 11.2(a)); or the slow growth which

might occur under decompression or heating (figure 11.2(b)). Since the bubble undergoes no oscillation in these two cases, it has no acoustic effect beyond that of an impedance mismatch. However, as a mechanical oscillator possessing stiffness and inertia, the bubble has a natural frequency (figure 11.2(c)), which corresponds to the note emitted when such a bubble is entrained (for example in a waterfall; Leighton and Walton 1987). Further coupling with an oscillatory pressure field occurs when a bubble is driven into pulsation by an incident sound field. The bubble might be driven such that the amplitude of oscillation of the bubble wall is low (for example, the incident sound field has a low acoustic pressure amplitude, or the bubble has a size which differs greatly from that required for resonance with the driving frequency). In such a case, energy is lost from the beam through acoustic re-radiation and, as discussed in section 11.2, through conversion to heat by viscous and thermal damping mechanisms associated with the bubble motion (Devin 1959, Eller 1970). Figure 11.2(c) shows just such an oscillation. Despite damping, the illustrated bubble pulsations are shown not to decrease in amplitude, suggesting a continuous-wave or tone-burst insonation, rather than a short pulse (which is the case illustrated in figure 11.2(d)).

Inertial cavitation, typified by the sudden expansion and then rapid collapse of the bubble, is shown in figure 11.2(e). The bubble may fragment, or repeat the growth/collapse cycle a number of times (Apfel 1981a, b); or, in specialised conditions, can pulsate for thousands of cycles, emitting a sonoluminescent flash at each collapse, as discussed in section 11.1.3. The generic oscillation shown in figure 11.2(f) is a high amplitude pulsation of a spherical bubble. Depending on the amplitude, such oscillation may be inertial, or non-inertial but of high amplitude. If non-inertial, there are a number of interpretations of how this situation, which differs in wall oscillation amplitude from that shown in figure 11.2(c), arises. It might, for example, occur if figures 11.2(c) and (f) show the same bubble, but in figure 11.2(f) the acoustic pressure amplitude of the driving field is greater. Alternatively the bubbles in figures 11.2(c) and (f) might be in the same sound field, but with the bubbles in figure 11.2(f) being closer to resonance. However as the bubble is more closely driven to resonance, other effects begin to occur. Most notable of these are surface waves, which visually cause a 'shimmer' to appear on the surface of the bubble (Neppiras 1980), and acoustically may be detected with a combination-frequency technique (Phelps and Leighton 1997). Such surface waves are illustrated in figure 11.2(g). Surface waves will be stimulated if the amplitude of the bubble wall displacement exceeds a certain value, which it will tend to do as the amplitude of the sound field increases (providing no other effect, such as inertial cavitation or fragmentation, occurs); or, more commonly, the closer the bubble is to the resonance condition. In general, the greater the degree by which the amplitude of the driving field exceeds the threshold condition to excite surface waves on resonant bubbles, the broader the range

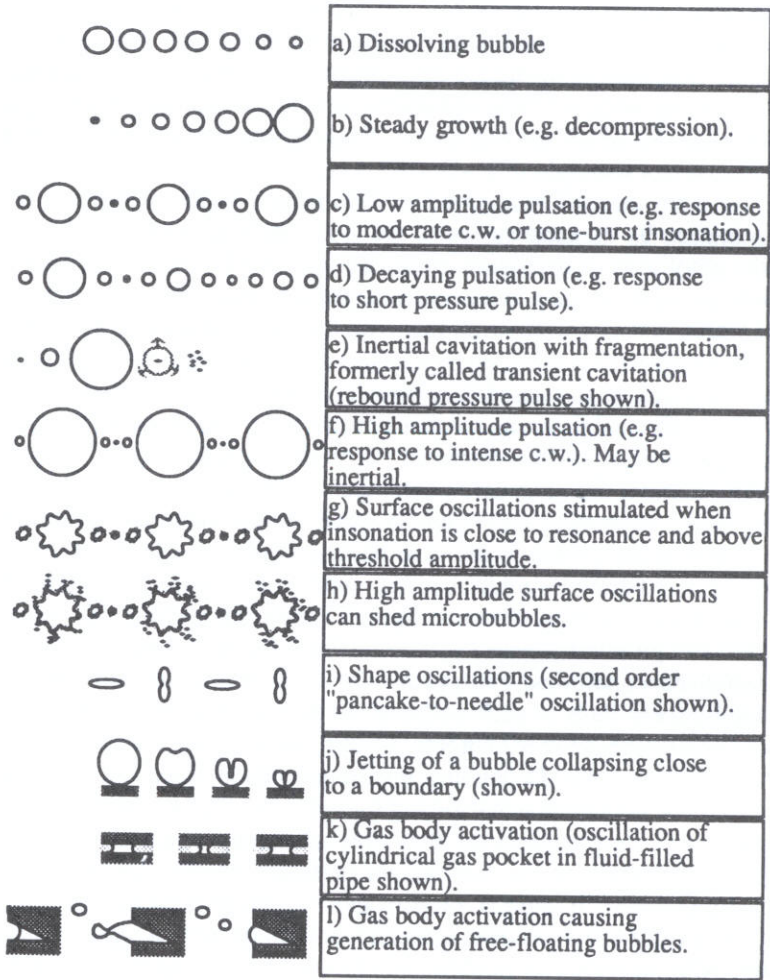


Figure 11.2. A schematic illustration of the range of bubble behaviour. The behaviours are described in the text. The expansion ratios drawn for these bubbles are exaggerated to illustrate the pulsations more clearly.

of bubble sizes, centred about the resonance, on which surface waves will be stimulated (Phelps and Leighton 1996). Surface waves can be associated with an erratic 'dancing' translational motion (Crum and Eller 1969) and, at high amplitudes, microbubbles might break off from the tips of the surface waves (figure 11.2(h); Leighton 1995). Surface wave activity is also associated with microstreaming, local circulation currents around a vibrating body which can influence mass transport in the liquid (section 11.3.1) and generate shear forces (section 11.4) close to a bubble.

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. 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. When the bubble is expanded, each liquid shell contracts. The concentration of dissolved gas in the liquid adjacent to the bubble wall is less than the equilibrium value (Henry's law), but the shell is thinner than when the bubble is at equilibrium radius, so that the gradient across the shell is higher. Therefore 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. Though 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 smaller gradient drives the gas a longer distance. The former effect is dominant.

The result is that both the bubble wall surface area and the dissolved gas diffusion rate are asymmetrical with respect to expansion and contraction: the 'area' and 'shell' effects reinforce one another. The combined effect means that during nonlinear cavitation in acoustic fields which are sufficiently intense, the equilibrium radius about which the bubble pulsates will tend to increase (Eller and Flynn 1965, Eller 1972, Crum 1984, Church 1988a,b).

The threshold acoustic pressure for growth is dependent on the bubble size and the acoustic frequency. Local minima in the acoustic pressure threshold are seen at harmonics and subharmonics of the resonance (Church 1988a). The growth rate once the threshold has been exceeded is strongly influenced by the presence of surface-active agents, and by microstreaming (introduced in 11.2), which affects the transport of dissolved gas beyond the bubble wall (Church 1988b). Microstreaming was not incorporated into the above simple discussion, but its effects are qualitatively simple. 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 further 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 (Church 1988b).

Rectified diffusion and dissolution allow bubbles to steadily change their equilibrium size, and therefore modify the cavitation activity they undertake. Among the wide range of possibilities are the following. A bubble may grow to become closer to the resonance condition (where, for example, surface waves and microstreaming require lower acoustic pressure amplitudes to be stimulated). If a bubble fragments following an inertial collapse, and the resulting small bubbles are smaller than the size required to nucleate inertial cavitation, but larger than that required to grow by rectified diffusion, such growth can make inertial cavitation a self-nucleating process. Church (1988a) outlines some of these possible scenarios, and assesses their likelihood.

11.3.2. *Alteration of the acoustic pressure field at the bubble by radiation forces*

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 degree (Leighton 1994a, §3.3.2(d)). That degree is governed by the magnitude of the difference in density between gas and liquid or, equivalently†, the instantaneous bubble volume. As an acoustic field passes through a liquid, the liquid particle acceleration oscillates, reversing direction periodically such that it is aligned with the direction of propagation for half the acoustic cycle, and contrary to it for the remainder. The bubble, being less dense than the liquid, will follow suit, accelerating in the same direction as the continually alternating liquid particle acceleration‡. However if the bubble is pulsating with the same periodicity as the driving field (which a linear oscillator will do), then the phase relation is such that the bubble will be in the expansion phase (i.e. with its radius greater than equilibrium) as it travels in one direction, and in the compression phase as it travels in the other direction (Leighton *et al* 1990). The bubble accelerates more during the expansion phase than the compression, since then its volume is clearly greater, and its density less. Therefore such a bubble in a sound field will experience forces which reverse direction twice each acoustic cycle. However the net effect will be that it travels in the direction taken by the liquid acceleration when the bubble is in the expansion half-cycle. If the

† Assuming the bubble contains a fixed mass of gas.

‡ Assuming that inertial effects on the response times are small.

bubble is assumed to be an oscillator with a single degree of freedom, bubbles of less than resonance size will pulsate in antiphase to those of greater than resonance size. Therefore if one type of bubble is accelerated in one direction by the sound field, the other type will be forced in the opposite direction.

This behaviour is most readily observed in a standing wave field, where the radiation forces are commonly called 'primary Bjerknes forces'. They cause bubbles of less than resonance size to travel up pressure gradients to collect at acoustic pressure antinodes, while bubbles of larger than resonance size migrate down the gradients to the nodes. Similar comments apply in focused acoustic fields, where bubbles of less than resonance size migrate up pressure gradients towards the focus. Such migrations change the acoustic pressure amplitude at the bubble, and therefore affect the type of cavitation. However there can be important indirect effects, such as the acoustic shielding which such aggregations of bubbles produce, which also affect the local acoustic field (Leighton 1995).

Formulations also exist for the radiation force on a bubble in travelling-wave conditions, where the force is greatest on resonant bubbles; and for the force on a particle suspended in a liquid close to a bubble (Coakley and Nyborg 1978), an example of the latter being the aggregation of platelets in blood (Miller *et al* 1979). The so-called 'secondary' or 'mutual' Bjerknes force is exerted between two pulsating bubbles (Bjerknes 1906, 1909, Prandtl 1954, Batchelor 1967, Leighton 1994a, §4.4.1). There is a general rule that two bubbles which are both less than, or both greater than, resonance size attract; but that if one bubble is greater than, and the other less than, resonance size, they repel. However this is a simplification, and the results may be more complicated if the bubble population density is high or the incident sound field is strong.

Radiation forces therefore can cause mutual repulsion or attraction between bubbles. The latter can cause the net bubble size to increase by causing coalescence; or decrease, because the proximity of one bubble can induce shape oscillations in another (figure 11.2(i)). If sufficiently pronounced, such oscillations can lead to bubble fragmentation (Leighton 1995, Leighton *et al* 1995a, 1998). As discussed with respect to figure 11.2, the equilibrium size of a bubble (along with the acoustic pressure and frequency) strongly influences the type of cavitation it will undergo. Inertial cavitation can be nucleated by bubbles of an appropriate size (section 11.1.3). Details of the nucleation will be discussed in section 11.3.3; and section 11.3.4 will return to the theme of how radiation forces, nucleation and the acoustic field, as well as gas diffusion, determine the cavitation type and effects which will occur when ultrasound is passed through a liquid.

The radiation forces associated with bubbles which have been described in this section can be considered as particular extensions of the more general radiation forces acting at interfaces which have been discussed in Chapter 3.

11.3.3. Nucleation

At the start of section 11.1.3, a free-floating bubble is considered to be an appropriate nucleus for inertial cavitation. The theory outlined there assumes nucleation of inertial cavitation within the first acoustic cycle, so-called 'prompt cavitation', from a free-floating spherical bubble nucleus. Clearly effects relating to longer insonation periods, such as growth by rectified diffusion, are not covered. In practice of course it is not a requirement that such nuclei be present before the start of insonation. Not only may bubbles which are initially too large to nucleate inertial cavitation enter the critical range (through, for example, dissolution, or fragmentation through a shape oscillation or microbubble shedding, as discussed above). Bubbles too small to nucleate cavitation may also enter it through rectified diffusion or coalescence. Radiation forces can affect both size increases and size reductions, by relocating bubbles to regions of greater or lesser acoustic pressure amplitude, or to the presence of neighbours. Radiation forces in focused fields can even convect suitable nuclei into the focus to nucleate inertial cavitation there (Madanshetty *et al* 1991, Madanshetty 1995).

The model employed to produce figure 11.1 is based upon the dynamics of isolated, spherical bubbles, which *de facto* must be free floating. Such bodies are not stable with respect to loss from the liquid as a result of buoyant forces, and indeed will tend to dissolve. Why therefore any suitable nuclei can be found in a sample of liquid which has been left standing for hours is an interesting question (Leighton 1994a, §2.1). Hydrophobic impurities, if present, can collect on a bubble wall over time, and hinder further reduction in size (Akulichev 1966, Sirotyuk 1970, Yount 1979, 1982, Yount *et al* 1984). Other possible nuclei can be found naturally as gas pockets, stabilised against dissolution in crevices and cracks in the container wall or within free-floating particles within the liquid (Harvey *et al* 1944, Trevena 1987, Atchley and Prosperetti 1989, Leighton 1994a). The process by which these bring about nucleation is illustrated in figure 11.2(*l*). High amplitude ultrasonic waves cause the gas pockets to either expand out of their crevice, or conceivably shed microbubbles through surface waves, to generate free-floating nuclei for cavitation. If inertial cavitation is undesirable, then such particles can be removed by filtering. However even this will not completely remove all suitable nuclei for inertial cavitation, which may be generated, for example, by the passage of cosmic rays through the sample (Greenspan and Tschiegg 1967).

11.3.4. Population effects

For a given liquid (including its gas and solid content), it was shown in section 11.1.3 that, to first order, whether a bubble undergoes inertial or non-inertial cavitation depends on three parameters: the acoustic frequency, the acoustic pressure amplitude *at the bubble*, and the equilibrium bubble size. In

section 11.2 this idea was extended to show that, of the wide range of types of cavitation that exist, the ones that occur in a given situation will depend on the above parameters plus others (such as the proximity of the bubble to inhomogeneities such as other bubbles, particles or walls). Section 11.3 has outlined how such key parameters as bubble location and size can be altered during insonation (by radiation forces, rectified diffusion, coalescence and fragmentation etc). It is usually simple to control the acoustic frequency to which a sample is subjected. It is, understandably, often much more difficult to control the bubble size and location.

The type of cavitational behaviour a bubble undertakes depends on the relation between its size and the other critical sizes (figure 11.2). These include: the radius which is resonant with the sound field (governing, for example, radiation force effects, surface wave activity and microbubble shedding etc); the upper and lower limits of the radius range for the nucleation of inertial cavitation; and the threshold for rectified diffusion. How such changes affect the bubble size distribution with respect to the critical sizes mentioned above determines the bubble activity seen. The issues involved may be complex, as the following scenarios suggest. Dissolution and fragmentation provide mechanisms by which bubble size reductions in the population can occur. Coalescence and rectified diffusion provide the ways to produce larger bubbles. To produce a bubble which is larger than resonance, it must either pre-exist (and continue to persist during insonation despite buoyancy and possible fragmentation); form through coalescence; or grow to a size larger than resonance by rectified diffusion. Acting against the latter scenario is the fact that it is on reaching resonance size that the bubble is most likely to lose gas through microbubble emission from surface waves (figure 11.2(h)). Also, if the bubble does pass through resonance intact, then rectified diffusion becomes far less efficient once a bubble exceeds resonance size, the pressure threshold increasing and the growth rate decreasing. There is also the general trend that the larger the bubble, the more easily it is fragmented. Therefore in many fields, it is the aggregation of the smaller bubbles at pressure antinodes or the focus which is more commonly observed (figure 11.3). Bubble aggregations such as the one mentioned above are acoustically active. They may shield, channel, or scatter the acoustic field. This leads to the surprising observation that, of the three parameters mentioned at the start of this section, the acoustic pressure amplitude *at the bubble* may also sometimes be difficult to control.

The types of cavitation (and there may be many) which occur when a liquid is insonated therefore depend on a large number of interacting parameters, introduced above and summarised in figure 11.4 (Leighton 1995). It is possible to control such interactions to produce surprising effects, such as 'pulse enhancement'. This occurs when the magnitude of an effect produced when ultrasound is used in pulsed mode exceeds that observed when the same amount of ultrasonic energy is delivered in continuous wave.

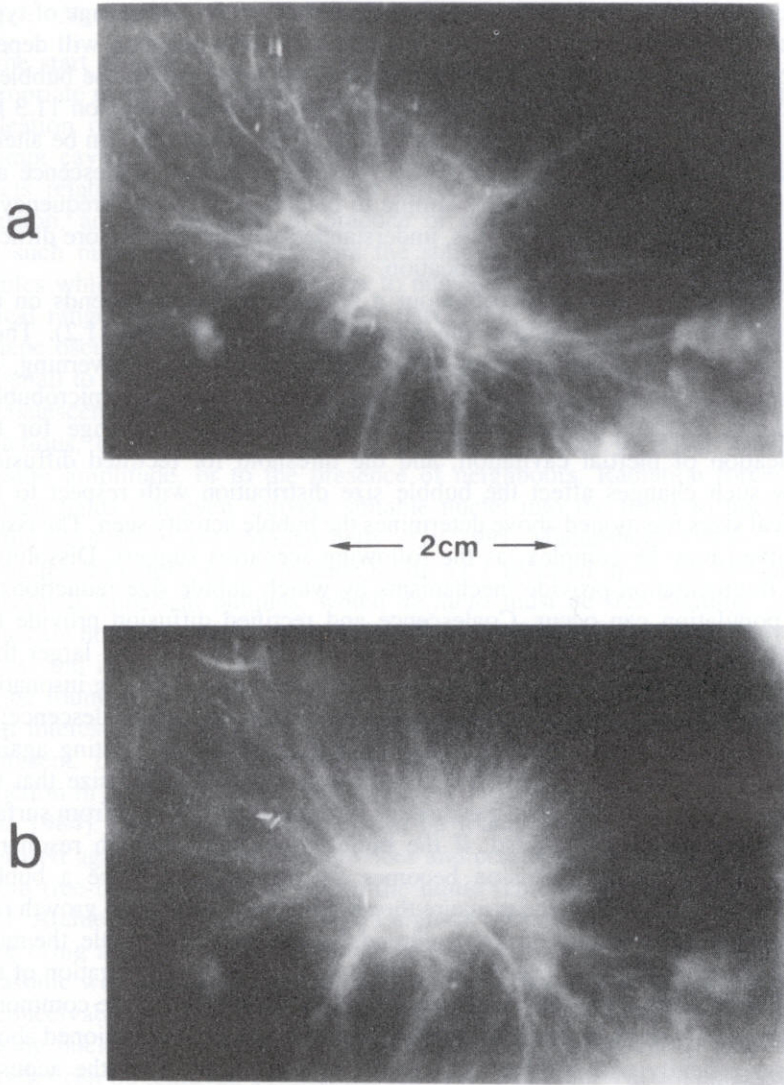


Figure 11.3. *Two photographs of aerated water cavitating in a cylindrically focused 10 kHz continuous-wave sound field, taken approximately 3 s apart. The view is along the line of the axial focus, where the acoustic pressure amplitude is 0.24 MPa. Exposure time, 1/30th second. Streamers are clearly visible, comprising bubbles moving rapidly towards the focus, driven by radiation forces. Comparison of (a) with (b) illustrates that, although the general form is constant, the details change.*

There are a number of mechanisms by which this can be brought about, involving complex interactions of the parameters described above. Detailed

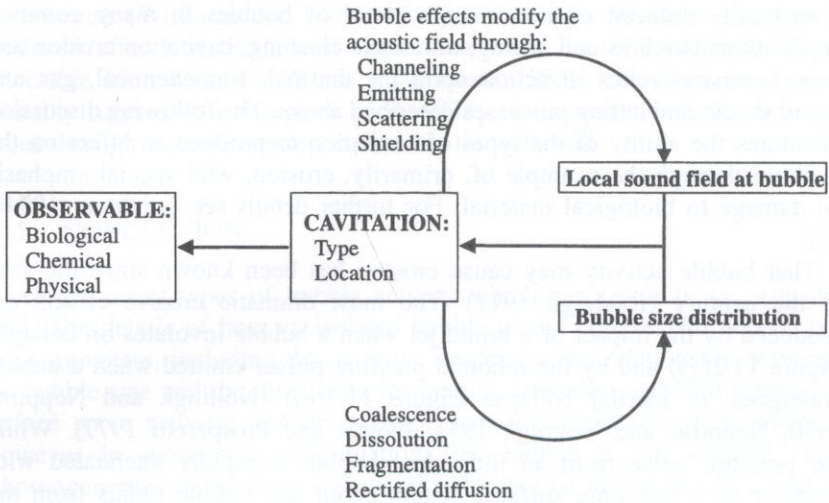


Figure 11.4. A schematic illustration of how the various factors interact to determine the types of cavitation which occur, and hence the effect observed. The latter may be chemical, physical or biological. It depends both on the type of cavitation (e.g. inertial, non-inertial, jetting, fragmentary etc) and its location. Both 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, characterise the inertial 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 is determined by the local sound field and the bubble size distribution. However there is feedback from the cavitation which influences these two key parameters.

discussion of such mechanisms (Leighton 1994a, §5.3) is beyond the scope of this chapter. In the final section, the observables which can be produced by cavitation, the 'end result' in figure 11.4, will be discussed.

11.4. THE IMPLICATIONS OF THE OCCURRENCE OF ONE TYPE OF CAVITATION FOR CAUSING CHANGE TO THE MEDIUM

As shown in figure 11.4, cavitation can produce physical, chemical and biological effects. Given the complexity of interactions shown in that figure and, further, the range of behaviour a single bubble can undergo in an acoustic field (figure 11.2), it is not surprising that the production of a given effect may not be uniquely the result of one form of bubble activity. 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 thermal, sonochemical, gas and liquid shock, and jetting processes described above. The following discussion illustrates the ability of the types of cavitation to produce an effect on the medium through the example of, primarily, erosion, with special emphasis on damage to biological material. For further details see Leighton (1994a, §5).

That bubble activity may cause erosion has been known since the start of the century (Rayleigh 1917). The most dramatic erosive effects are produced by the impact of a liquid jet when a bubble involutes on collapse (figure 11.2(*j*)) and by the rebound pressure pulses emitted when a bubble undergoes an inertial collapse (figure 11.2(*e*)) (Noltingk and Neppiras 1950, Neppiras and Noltingk 1951, Plesset and Prosperetti 1977). While the pressure pulse from an individual 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 pressure emission of a single bubble, clouds of bubbles may collapse co-operatively, enhancing the damage (Vyas and Preece 1976). It is likely that cavitation erosion resulting from jetting and inertial collapses plays a part in lithotripsy (Coleman *et al* 1992, 1993).

If non-inertial pulsations are of sufficient amplitude, bubbles may cause damage by a number of mechanisms. For example, cell disruption may be brought about because the bubbles can travel rapidly through the liquid under the influence of acoustic radiation forces, generating hydrodynamic stresses which have been shown to produce haemolysis (Miller and Williams 1989). If the acoustic field amplitude exceeds a certain threshold, which is lower the closer the bubbles are to resonance (Phelps and Leighton 1996), surface waves are stimulated (figure 11.2(*g*)). Associated with these are the microstreaming currents discussed in sections 11.2 and 11.3.1, which can cause an erosive effect (for example, with dental ultrasonics; Ahmad *et al* 1987). Non-inertial cavitation may indirectly influence erosion through affecting the damage which results from inertial cavitation. This might occur through shielding of the sound field, or through the production of nuclei, through surface wave activity (figure 11.2(*h*)) or rectified diffusion (section 11.3.1).

The ability of non-inertial cavitation to directly bring about physical damage is usually of greatest importance in the absence of inertial cavitation (which would otherwise dominate the erosion, masking the contribution from stable bubbles). Bio-effect from non-inertial cavitation is therefore more readily identifiable when low intensity fields are applied to sensitive chemical or biological systems. Demonstrations include microstreaming-induced cell death associated with gas body activation (Miller 1985, Vivino *et al* 1985) and the use of photometry to detect ATP release from *in vitro* human

erythrocytes (Williams and Miller 1980), a technique which has probably produced effects at the lowest continuous-wave intensity recorded for a detectable bio-effect, 4 mW cm^{-2} (Williams 1983). The mechanism was probably rupture or a change in permeability of the cell membrane brought about through microstreaming stresses at the cell wall.

11.5. CONCLUSION

There are many types of bubble activity which can occur in an acoustic field. The details of how an isolated bubble would behave will depend on key parameters, including the acoustic frequency; the relationship between the bubble size and the thresholds for inertial cavitation, rectified diffusion, surface wave activity; and the acoustic pressure amplitude *at the bubble*. However in most practical situations the bubble is not isolated, and inhomogeneities in the fluid and sound field may be important. That said it is often the effects of cavitation, rather than the type of bubble activity *per se*, which are important, and these will depend on the interactions between the bubble population, the sound field, and the medium.

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