SONOLUMINESCENCE IN SPACE: THE CRITICAL ROLE OF BUOYANCY IN STABILITY AND EMISSION MECHANISMS

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INTRODUCTION

Sonoluminescence ("light from sound") is the result of extremely nonlinear pulsations of gas/vapor bubbles in liquids when subject to sufficiently high amplitude acoustic pressures. In a single collapse, a bubble's volume can be compressed more than a thousand-fold in the span of less than a microsecond. Even the simplest consideration of the thermodynamics yields pressures on the order of 10,000 ATM. and temperatures of at least 10,000K. On the face of things, it is not surprising that light should be emitted from such an extreme process. For a general review of some of the remarkable experimental results issuing from the study of sonoluminescence in acoustically levitated bubbles we refer to a number of articles [Roy 1994; Crum 1994; Crum & Roy 1994; Putterman 1995] The phenomenon has achieved prominence in the public eye [Browne, 1996; Glanz, 1996; Knight, 1996]. The increase in scientific interest can be gauged by noting that, while as recently as 1990 (the year after Gaitan discovered light from a single bubble) there were 4 SL-related articles in refereed journals, this number increased to 13 in 1995, 24 in 1996, 31 in 1997, and 14 refereed journal articles have already appeared in 1998!

Experiments [Gaitan et al. 1992a,b, 1997; Barber et al. 1991, 1992, 1994; Hiller et al. 1992, 1994; Holt et al. 1994, 1996; Löfstedt et al. 1995; Matula et al. 1995, 1996, 1997a,b,c,d; Lepoint et al. 1997, among others] have uncovered the existence of many unexplained features surrounding the observation of sonoluminescence. Of these, we have identified four which are fundamental, and in which gravity plays (or is predicted by theory to play) a critical role. These are:

- the light emission mechanism itself,
- the mechanism for anomalous mass flux stability,
- the disappearance of the bubble at some critical acoustic pressure, and
- the appearance of quasiperiodic and chaotic oscillations in the flash timing.

Gravity, in the context of buoyancy, is implicated in all four of these unexplained phenomena.

PROBLEM STATEMENT

The scientific objectives of the planned research are:

(1) To test the predictions of the fractoluminescence light emission theory of single bubble sonoluminescence [Prosperetti 1997] which relies on the asymmetry induced by buoyancy-induced translatory oscillations to postulate that a jet forms during the final stages of bubble collapse. This hypothesized jet shoots through the bubble interior and makes contact with opposite bubble wall at near Mach speeds and 'fractures' the water, with resultant light emission. Vortices in the liquid carry off excess linear momentum [Longuet-Higgins 1996], so that the process can repeat the next acoustic cycle. This theory has many specific predictions, which can be summarized here by noting that, if gravity is reduced to the ambient orbital level of 10^{-5} g, then this theory would predict a completely different picture of the parameter space and its regions of stability and light emission than the one which has recently been uncovered in 1g experimentation [Holt and Gaitan, 1996, 1996a; Gaitan and Holt, 1997]. Simply by making careful measurements of the details of light emission and mechanical oscillations of bubbles in μg, where buoyancy-induced coupling of translation and oscillation motions is absent, we can definitively establish the viability of this theory.

(2) To measure the mass flux and mechanical stability boundaries which constrain the appearance of light emission in the parameter space of acoustic pressure, dissolved gas concentration and equilibrium radius. These measurements will test both the fractoluminescence and the chemical reaction/diffusion theories that model the non-diffusive anomalous mass flux observed in 1g experiments [Löfstedt et al. 1995; Holt & Gaitan 1996a]. The latter theory [Lohse et al. 1996a,b] relies on the assumption that gravitational effects are not important in the convective-diffusion treatment of mass transfer.

(3) To measure the precise values of acoustic pressure and equilibrium radius where a light-emitting bubble disappears, an unexplained phenomenon which is a ubiquitous feature of 1g experiments. The disappearance represents an upper bound on the acoustic pressure (and hence energy input to the system) which can be employed to drive the bubble oscillations. These measurements will directly test a theory [Cordry 1996; Matula et al. 1997c] which postulates that the variation in buoyant force with each acoustic cycle's expansion and contraction of the bubble leads to a loss of levitation above some critical acoustic pressure.

(4) To test whether chaotic and quasiperiodic timing of the flashes observed in 1g [Holt et al. 1994] is

due to buoyancy-induced effects or germane to the phenomenon of light emission itself. The nonlinear coupling of translational and radial motion via the buoyant force is postulated to result in quasiperiodic and chaotic modulation of the volume oscillations, but this would not occur in μg. Simultaneous monitoring of the timing of individual flashes during detuning of the system will allow us to determine the presence or absence of the nonlinear effects, and thus to prove or disprove the conjecture.

BACKGROUND

Rather than giving a comprehensive review of light emission and nonlinear bubble dynamics in general, we will use the four above features to both motivate and organize our background section. Reviews of nonlinear bubble dynamics are numerous; a compilation is available in Leighton [1994]. Since the research is so topical, no comprehensive review of competing theories for the light emission mechanism can be written. We briefly list some of the top contenders, which include electron conduction-quenched plasma [Moss et al., 1997], shock-wave mediated Bremmstrahlung [Wu & Roberts 1994], plasma discharge [Lepoint et al. 1997], superradiance [Trentalange & Pandey 1995], acoustic superresonance [Brenner et al. 1996], quantum vacuum fluctuations [Eberlein 1995], confinement of post-ionization electrons [Bernstein and Zakin 1995] and collision-induced dipole emission [Frommhold & Atchley 1994]. Other than the fractoluminescence theory discussed above, only one of these various theories for the light emission mechanism would be directly influenced by gravity, and that is the plasma discharge theory, which relies on a shape instability (Rayleigh-Taylor mechanism) to concentrate electric charge on the surface of a critical microjet that penetrates the collapsing bubble interior. The net result is "lightning" inside the bubble due to plasma discharge associated with spray electrification of the jet as it breaks up into droplets. While this theory has not gained widespread acceptance, it nevertheless has been able to explain some features of the light emission. Except to say that it remains a contender and falls under the heading of "affected by gravity", we will not discuss this theory further. In the next section, we concentrate on one theory in particular which has the potential to explain in a unified fashion many of the experimental observations.

Time-varying buoyancy: the direct effect of gravity

Consider the situation of a single air bubble in water, subject to a harmonic acoustic field which is also a standing wave. The acoustic wavelength is much larger than the bubble radius R_0 (kR_0 is small, where k is the acoustic wavenumber). On the acoustic time scale (typical acoustic periods are 50 microseconds), the bubble will contract during the compression phase of the field. The pressure and temperature will increase. During the expansion phase of the external field, the bubble expands. Thus the bubble volume, $V(t)$, can change significantly during an acoustic cycle.

An oscillating bubble in a standing pressure wave experiences a nonlinear body force dominated by the large relative compressibility between the bubble and the host fluid. The force depends on the time-average of the product of the bubble volume and the pressure gradient, known as the primary Bjerknes force [Rayleigh 1894; Bjerknes 1906; Yosioka & Kawasima 1955; Doinikov 1994]. Its functional form is $F_a \propto \langle V(t) \bullet \nabla P_a(r,t) \rangle$, where $\langle ... \rangle$ denotes a time-average over one acoustic cycle, and $P_a(r,t)$ is the acoustic pressure field. F_a acts in the direction of the pressure gradient of the standing wave, and thus it can balance the buoyant force and cause stable trapping of a bubble.

Finally, the bubble experiences a buoyant force $F_b = \rho\, gV(t)$ directed opposite the acceleration due to gravity. In practice, the fact that this force varies in time with the bubble volume is usually ignored, due to the small change in volume coupled with the inertia and drag resistance to translational motion in the liquid. Thus, one normally considers an acoustically levitated bubble to have a fixed vertical position at constant acoustic pressure which will depend on the ratio of the magnitudes of the forces F_b/F_a. However, in the parameter regimes where sonoluminescence occurs, the ratio of V_{max}/V_0 can reach 10^3 and greater, as shown in Fig. 1.

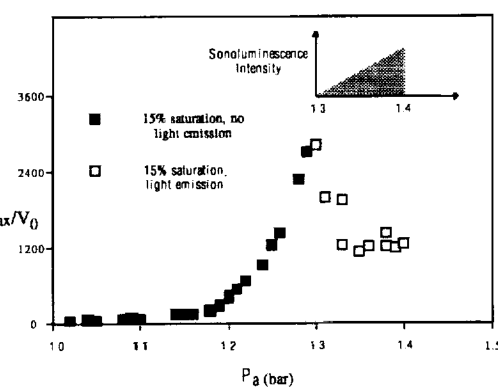


Figure 1. The measured ratio of V_{max} / V_0 for a bubble as a function of pressure. Light emission begins very near the peak value of the normalized response. The dissolved air concentration in the water was 15% of the saturation concentration at 1 atmosphere ambient pressure. The equilibrium or ambient radius varies with pressure in a complicated fashion, and is implicit in these measurements. Derived from Gaitan and Holt (1998).

The resulting ratio of F_b/F_a can vary by a factor $O(10^3)$ during a single acoustic cycle -- thus the bubble must undergo translatory oscillations driven by the time-varying buoyant force F_b! This is a fundamental result, independent of any proposed theory for sonoluminescence. It is the basis for the fact that gravity plays a tremendously important role in these nonlinear bubble oscillations (despite that fact that a straightforward calculation of a static Bond number for earth-based levitation yields $\rho \; g(R_0)^2 / 2\sigma \;\; \sim O(10^{-7})$ for a 10 micron bubble!). The effect of gravity lies in the magnitude of the changes in F_b. The ratio F_b/F_a can be considered a coupling strength between the radius of the bubble and the position of the bubble's center of mass. Because of this nonlinear coupling, volume oscillations can drive translatory oscillations. If gravity is effectively removed, this coupling constant will become much smaller than unity, and thus its effects will virtually disappear.

INVESTIGATIVE APPROACH

Our short-term plan is to perform 1g laboratory, drop tower, and parabolic flight experiments. Certain basic questions can be answered scientifically by drop tower and parabolic flight tests. But going beyond a "light or no light" level of sophistication will require the experimentation time to change key parameters. Significant experimentation, as we have documented, has been performed in 1g leading to a rationale for an experimental design in which the critical measurables are already identified and the relevant parameters are well characterized. Since 0g experimentation time is limited, the scope of our plan is finely focused: this investigation is mainly concerned with probing mechanisms for the light emission and related mechanical stability, not with a parameter study. In the section that follows we detail the experimental plan; here, we present the plan's essential features for rapid consideration. The crucial host fluid is water. The important dissolved gases are air and argon. Controlled dissolved gas concentration must be achieved in preparation and/or storage. The acoustic system must be capable of generating controlled acoustic pressures ranging from 1/10 ATM to 3 ATM. The dissolved gas concentration must be varied, either by pre-flight preparation, in-flight preparation, or variation of the test cell's ambient pressure. Temperature must be controlled. The critical measurables and their associated method are:

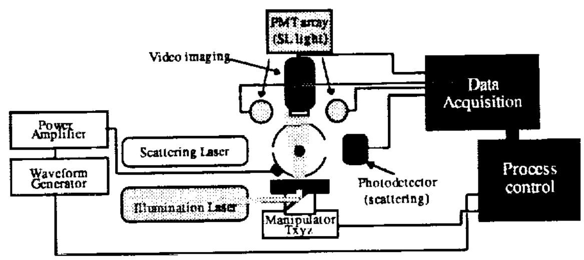
- acoustic pressure versus time, $P_a(t)$
  (in situ hydrophone)
- instantaneous radius versus time, $R(t)$
  (laser light scattering)
- maximum and equilibrium radius, R_{max} & R_0
  (imaging)
- flash timing, intensity and duration
  (time-resolved photometry)
- flash spatial distribution
  (multiple coincidence detectors)

We emphasize that these are all techniques with which we have extensive experience from ground-based laboratory experimental work. This is not to say that the experiments have all been done, but that the measurements we propose are eminently feasible.

Method and Diagnostics

The experimental techniques will build on an optical scattering/acoustic levitation/imaging system used in Holt and Gaitan [1996, 1996a, 1997] and a photometric system used by Holt et al. [1994], and by Matula & Roy et al. [1997a,b]. This section will simply highlight features of the apparatus and method, as well as discuss calibration and procedures.

Figure 2. Schematic of the experimental setup.



To monitor the oscillations of the bubble wall, it is illuminated by a linearly-polarized laser beam, and the scattered intensity as a function of time at a specific angle from the forward is recorded. The voltage output of a photodetector (typically a fast photodiode, but PMT's are useful for low intensity work) positioned at the aforementioned angle is the time signal of interest. For purely spherical volume oscillations, a monotonic transfer function can be theoretically derived and experimentally calibrated [Holt & Crum 1990] relating the voltage and the bubble radius R(t).

To determine the presence and nature of shape oscillations, a variety of techniques will be utilized in the manner of Holt & Gaitan [1996]. First, to detect the onset and frequency of shape oscillations, the scattered light intensity $V_{exp}(t)$ can be monitored for either a period-doubling or a rapid amplitude change [Gaitan & Holt, 1996]. Time-gating techniques can be used to provide a trigger criterion for shape oscillations coupled to the ringing oscillation after the main bubble maximum expansion. The values of acoustic pressure P_a and equilibrium radius R_0 at the onset of shape oscillations will be measured. Secondly, to determine the mode of oscillation, high-resolution, short-illumination-pulse video will be used.

High-resolution video imaging with short (<1μsec) pulse illumination will be employed to obtain mechanical response information, R_0, R_{max}, shape mode, and translational motion can all be obtained via this technique by improving on the system used by Holt and Gaitan [1996]. Digital image processing techniques will be utilized to derive statistics from individual images.

Calibration

1. Cell pressure:

The method used to calibrate the cell pressure is outlined in Crum [1983]. A small hydro-phone will be permanently mounted inside the cell. The pressure gradient in the gravitational direction will be measured. While levitating a bubble in the cell in 1g we measure its equilibrium position with respect to the measured pressure gradient in the cell, its equilibrium radius, and the hydrophone voltage as the pressure is varied. Using the equation expressing the balance of the acoustic force and the buoyant force for small oscillations to solve for the pressure, a calibration constant can be determined from fitting techniques. It is a null method, and is very sensitive to small changes in pressure. As a check, numerical fitting of the equations of motion for small-amplitude oscillations will be performed with the calibrated photodetector output as the data to be fit by varying the acoustic pressure amplitude in the model. Gaitan & Holt [1998] have verified that, while the Rayleigh-Plesset equation cannot predict where in the P_a, R space the asymptotic equilibria exist, it can accurately predict one of the triple (P_a, R_0, R_{max}) given the other two. Thus we can calibrate in situ in 0g as well.

2. Bubble radius

The quantity $I_{exp}(R,t)$, the incident scattered light intensity falling on the photodetector, is equal to $I_0 \int\int I_{rel}(R(t))\, d\theta \;\; d\phi$, where the limits on the spherical angle variables θ and ϕ in the integral are determined by the particular photodetection scheme used. I_0 is the intensity incident on the bubble. I_{rel} is the normalized component of the intensity (with linear polarization parallel to the scattering plane) scattered into an angle θ from the forward. I_{rel} is calculable from Mie theory. Thus, the photodetector output $V_{exp}(R,t) = F \int\int I_{rel}(R(t))\, d\theta \;\; d\phi$, where F is an apparatus-dependent constant to be determined empirically.

First, a stably oscillating bubble is obtained in the cell and moved into the beam. The output voltage is monitored on an oscilloscope, and the time average is recorded. During calibration, only linearly oscillating bubbles are used, since $R_0 = \langle R(t) \rangle$ only for linear oscillations, and hence $\langle I_{exp}(R,t) \rangle = I_{exp}(R_0)$ only for linear oscillations. As soon as $V_{exp}(R_0)$ has been recorded, an independent measurement of R_0 is made by instantaneously turning the sound field off while acquiring video images [Holt & Gaitan, 1996a]. The video image can be calibrated by a known scale. The calibration constant F is then determined by taking the ratio of the experimental voltage to the relative intensity Dividing the photodiode output by F gives the experimental relative intensity, which can then be used to find the radius. In this fashion, bubble radii can be obtained to within about 4% [Holt 1989].

An alternate technique has recently been introduced [Lentz et al. 1995]. This technique relies on the angular variation in the Mie scattering, where the number and location of intensity maxima are unique for a given bubble radius (at fixed laser wavelength). Thus, an absolute determination of R is possible. While the technique is very accurate, it is much slower than the previous technique. Nevertheless, it will be a valuable independent check on the fixed-angle method, and is able to provide in situ calibration without accessing the interior of the acoustic test cell.

Procedure

We present here the generic procedures for obtaining data by varying the accessible para-meters of the system. These can be divided into two classes:

1. Mechanical parameters

P_a will be varied between 0.5 and 1.5 ATM, depending on the dissolved gas concentration, and increasing to higher pressures if the pressure for disappearance is not the same as in 1g. The practical problem this introduces in 1g, i.e. changing the position of levitation, can be addressed with feedback control on the vertical translational stage via a PC-controlled stepper mike motor. Pressure control itself will be accomplished via a PC-controlled step sequence, increasing and decreasing the pressure in a controlled fashion with operator intervention possible at every step.

R_0 will be implicitly varied via the constraints of mass flux equilibrium and shape instability. From our experience in 1g, the approximate range should be 1 - 25 microns in equilibrium radius, and 20 to 70 microns in maximum radius. This may, of course, change dramatically with the removal of buoyancy-induced translatory oscillations.

The acoustic frequency f_d will be fixed at the resonance of the acoustic cell; with temperature control this should remain constant. When investigating variations in flash timing, f_d will be varied no more than a few hundred Hz from the resonance frequency.

2. Material parameters

The host liquid for all experiments will be water, distilled, micro-filtered and de-ionized to 17Mohm resistivity at 20°C. Two gases will be used, air and argon. They will be dissolved in water at varying percentages of saturation with respect to 100% saturation at 760 mm-Hg ambient pressure at 20°C for air over water. Air as a mixture of gases in the proportions found at sea level will be dissolved in equilibration at ambient pressures from 400 mm Hg down to the vapor pressure. Argon will be prepared in the same way, only the absolute pressure of the argon will reflect its partial pressure in air at sea level. Thus, argon will be dissolved in equilibration at ambient pressures from 8mm down to 3mm Hg.

Data Acquisition

The optical Mie scattering data will be the primary source for both information and triggering purposes for the other data acquisition methods. A photodiode or photomultiplier tube with a scattering laser-line bandpass filter will detect the scattered light. A calibrated system will yield both equilibrium and time-varying radius values. This signal will be digitized at 2 GS/s, and minimum 10 bit resolution, with record lengths up to 1Mb.

The Mie signal will be used to trigger a short light pulse (diode laser, superbright LED or short halogen strobe light) backlighting the bubble for obtaining high resolution video/CCD images Appropriate optical filters will be used to block out the Mie signal.

Matched photomultiplier tubes (or a mini-array) will be used to detect the sonoluminescence flashes. Flash timing circuitry will include a Constant Fraction Discriminator and Time-Amplitude Converter. Relative intensity measurements will be made using pulse height analysis with appropriate coincidence criteria, both to yield intensity as a function of parameter variation, and to detect spatial anisotropies in the emitted light.

MICROGRAVITY RATIONALE

The aspect of gravity which affects the proposed measurements is the buoyant force on the gas bubble. This is unavoidable in a material sense, since the density and compressibility contrast are required to obtain volume oscillations in an isotropic pressure field. In addition to the transla-tional oscillations discussed in the previous sections, the presence of the buoyant force (and the need to overcome it) results in: 1) a perturbation from the ideal spherical symmetry in which one would like to perform experiments; 2) a requirement of a minimum pressure below which experi-ments cannot be performed simply because the bubbles will rise to the container surface; 3) non-linear acoustic streaming, which will affect measurements of mass flux and mechanical stability.

We can quantify our discussion in the background section by considering the difference in the nonlinear coupling constant F_b/F_a in 1g and μg. To obtain simple order-of-magnitude results in this section we ignore the dependence on the relative phase of the bubble oscillation with respect to the acoustic field. The time-average ratio F_b/F_a is exactly unity on earth when the bubble is displaced above the central antinode of the standing wave acoustic field to a point where the gradient of the field is strong enough so that the forces balance -- this is the definition of acoustic levitation. As long as the volume oscillations of the bubble remain on the order of 25% or so, then the volume in the force equations $V(t)$ can be replaced by V_0, the equilibrium volume of the bubble. As is borne out in laboratory results, for these mild oscillations the bubble remains at the location given by $F_b/F_a = 1 \approx \rho\ g\ /\ K(P_ak)^2z$, given suitable low-amplitude approximations, where K is a material constant, P is the acoustic pressure amplitude, k is the acoustic wavenumber and z is the vertical height above the antinode. Thus $z \approx \rho\ g\ /\ K(P_ak)^2$ has no time dependence and depends only on P_a for low amplitudes.

However, as Fig. 1 shows, the ratio V_{max}/V_0 for the high-amplitude oscillations during sonoluminescence can exceed 10^3. Thus in 1g the ratio of F_b/F_a can vary roughly by a factor $V_{max} / \langle V(t)\rangle \approx 10^3$ during a single acoustic cycle. This means that the gravity-induced nonlinear coupling to the translational mode will be a maximum precisely when sonoluminescence occurs!

If, however, gravity is reduced to orbital ambient levels of $< 10^{-4}$ g, this means that F_b/F_a is also reduced to $O(10^{-4})$ or less, since F_b is linear in g. Thus even in the sonoluminescence regime, the coupling strength is only of order 10^{-2}. A bubble in microgravity will thus be located almost precisely at the acoustic pressure antinode, where the field gradient is zero. The nonlinear coupling between the volume mode and translation mode is removed.

The time scales in the problem dictate the need for extended periods of low acceleration. There are, of course, very short time scales involved in the problem, such as the picosecond scale for individual sonoluminescence flash widths. However, beginning with the diffusive time scale for the growth, dissolution or equilibration of bubbles (on the order of seconds), the need for long time experiments become clear. For a given dissolved gas concentration, both mechanical and photometric data must be gathered as phenomena develop on the diffusive time scale at a constant acoustic pressure. Then the pressure must be incremented and measurements repeated while still at the same dissolved gas concentration, until the critical pressure and radius where the bubble disappears. Such a sequence, even if fully automated, could require tens of minutes.

ACKNOWLEDGEMENTS

Work supported by NASA.

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