Chaotic Sonoluminescence

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Sonoluminescence (SL) is generated from single, stably oscillating bubbles in a stationary, time-periodic acoustic field. By measuring the time delay between flashes, the dynamics of the phenomenon has been investigated. While other researchers have concentrated on the remarkable periodic stability of the system, present results indicate that, for small variations in the governing parameters, period doubling, chaos, and quasiperiodicity can occur. The implications of the complex temporal behavior for the problem of determining the mechanism(s) for production of SL are discussed.

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The phenomenon of *sonoluminescence* (SL) [1] has recently enjoyed a rediscovery of sorts [2–5] in connection with the stable nonlinear oscillations of single bubbles in a periodic acoustic field. The energy-focusing effect of bubbles [$O(10^{11})$ times the energy density of the external acoustic field [6]] gives rise to extremely high temperatures and pressures within the bubble during its collapse. That these conditions yield light emission would seem an obvious conclusion, and yet SL still defies even heuristic explanation, let alone rigorous theoretical description.

Since SL spectra [3–5] have been equally well (or poorly) explained by such diverse processes as blackbody radiation, bremsstrahlung, plasma formation, and pressure-broadened OH^{-*} transitions, investigation of a less complex effect than a spectrum may be useful in gaining fundamental insight into at least one of the nonlinear processes involved. Towards this end, we have made novel measurements of the *timing* of the individual flashes from a stably oscillating bubble. We report here the results of some preliminary measurements.

Description of the measurements and techniques. —Figure 1 shows a schematic of the experimental setup. A spherically symmetric acoustic resonator is driven near one of its normal modes, setting up a stationary sound field in the water contained in the interior. Typically, the acoustic field pressure amplitude P and frequency f_d are about 1.3 bars and 27 kHz, respectively. A single air bubble is thus levitated near the center, the pressure gradient of the resonator mode providing a nonlinear radiation force which counteracts the buoyant force. The product kR_0 of the acoustic wave number and the bubble's equilibrium [7] radius is always much less than unity; hence only the volume (breathing) mode is directly forced by the time-varying pressure, because of the large compressibility contrast between air and water.

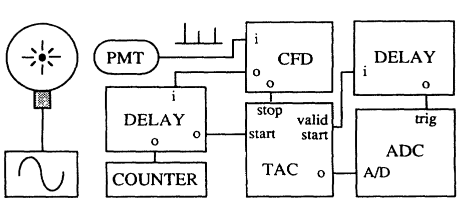

FIG. 1. Schematic of the experimental apparatus.

When the conditions for SL are met, the bubble emits a flash of light every acoustic period. A photomultiplier tube (PMT) is used to capture the light pulses. The time between each successive flash was measured to within 0.25 ns with a time-to-amplitude converter (TAC) and an appropriate delay circuit as described in the next paragraph.

An incoming voltage pulse from the PMT triggers a NIM pulse from the constant fraction discriminator (CFD). The NIM pulse (a) triggers the delay generator, and (b) stops the time-to-amplitude converter. At the end of the preset delay (~ 36 μsec, just shorter than an acoustic period) the TAC is started by a pulse from the delay generator, continuing the measurement cycle. The TAC output is a long pulse whose height is proportional to the start/stop time interval; when calibrated, the product of the TAC voltage and the time/voltage constant yields Δt, the variation in time between each successive flash. The valid-start signal from the TAC, which occurs simultaneously with the output pulse, is used to trigger the analog to digital conversion after a 6 μsec delay.

The time series thus generated consists of the variation Δt of the time between flashes, measured once per acoustic period for up to 16000 periods (see Fig. 1). Δt is small (~ 1 μsec) compared with the acoustic period $T \sim 37$ μsec, but it is not insignificant, as will be seen. The experimental error in Δt is Gaussian distributed, having a typical half-width of 0.25 nsec.

One feature of single-bubble SL that has direct bearing on these measurements is that it has been found to occur only in a very small region of the pressure-frequency-

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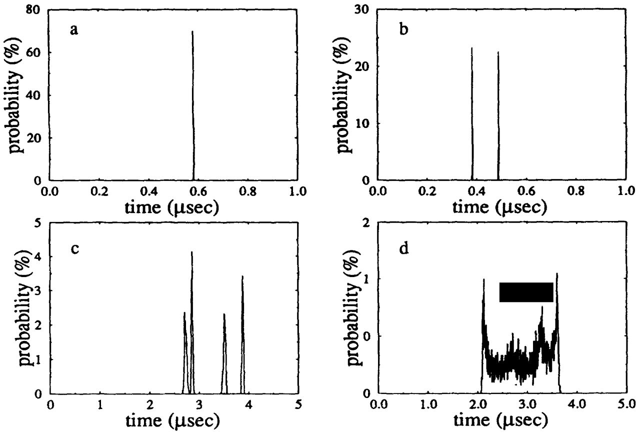

FIG. 2. Sequence of variation in time Δt between SL flashes from a single bubble. The data are histograms of the number of occurrences of a given Δt, measured sequentially during part of a bubble lifetime. The bin width is 0.25 nsec. Time zero is always defined as the time of the previous flash plus the ~ 36 μsec delay. The histograms show (a) a single maximum, (b) 2 maxima, (c) 4 maxima, and (d) a broad distribution. The frequency was slowly detuned about 0.01 kHz to initiate the bifurcation. $R_0 \sim 5$ μm, $P \sim 1.3$ atm, and $f_d = 27.0$ kHz.

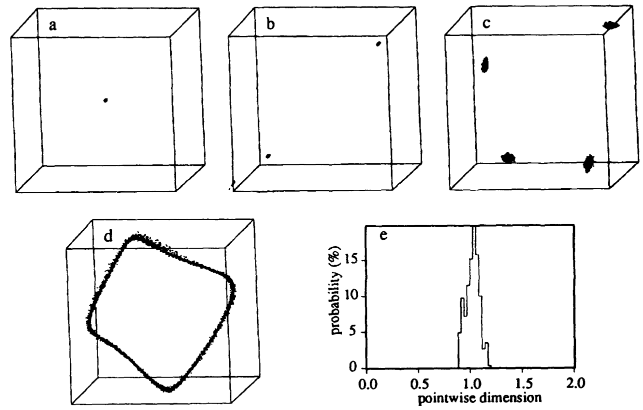

FIG. 3. Phase-space reconstruction of a typical bifurcation sequence of time series Δt. Each individual data point is a tuple of the form $(\Delta t_n, \Delta t_{n-1}, \Delta t_{n-2})$ generated using a single time series of flash data, where $3 \leq n \leq N$, and N is the number of data points (acoustic cycles) in each time series. Bifurcation of the variation Δt from period 1 (a) to 2 (b) to 4 (c) to a quasiperiodic state (d) is clearly shown. (a)-(d) are the attractors reconstructed from each of the Δt time series in Figs. 2(a)-2(d), respectively. (e) is the distribution of pointwise dimensions for (d).

radius space (P, f_d, R_0). Its discovery was fortuitous. The practical consequence is that very slight parameter variation can eliminate the phenomenon altogether. Thus, very sensitive experimental control is needed for the external driving parameters P, f_d. Controlling R_0 is even more difficult due to mass diffusion [7]. Further, departures from strictly singly periodic dynamical behavior occur via small variations in (P, f_d, R_0) within the already minuscule window for SL. The resonator itself must have a very high Q in order to reach the pressures necessary for SL. Thus, as the driving frequency is varied, the pressure also varies. In the experiments, fine control was achieved only via frequency control. The results presented here occur most often as functions of small frequency detuning from the resonator resonance, on the order of 0.01 kHz at 27 kHz. Additionally, no real-time measurements of R_0 were made; we can only specify a range, most likely between 4 and 10 μm.

Finally, sometimes the dynamics we report here occurred spontaneously, i.e., while the external controllable parameters were held constant. The most likely explanation is that R_0 changed due to mass diffusion—this is dynamically the same as varying the driving frequency with fixed R_0.

Results and analysis.—Figure 2 shows a histogram of the measured variation Δt of time between flashes as a function of slow detuning from resonance. The histograms show (a) a single maximum, (b) 2 maxima, (c) 4 maxima, and (d) a broad distribution, which is clearly not related to the measurement noise distribution. Some variation of this behavior was often observed in the experiments. Other methods of analysis must be used to say more about the dynamics at work here.

Because of the nature of the data acquisition, spectral methods such as fast-Fourier transforms are not as useful, and are even misleading, since aliasing is a problem. Methods from nonlinear dynamics are, at least qualitatively, much better at identifying the nature of the dynamics. In particular, they allow us to distinguish periodic, quasiperiodic, and chaotic behaviors.

The data are analyzed by transformation into a reconstructed, equivalent state space where the coordinates of each state are represented by time-delayed data values [8]. Specifically, each data point in Figs. 3(a)-3(d) and Fig. 4(a) is a tuple $(\Delta t_n, \Delta t_{n-1}, \Delta t_{n-2})$ generated using a single time series of flash data, where $3 \leq n \leq N$; N is the number of data points (acoustic cycles) in each time series. We note that our data acquisition scheme naturally (via the subtracted delay) generates a Poincaré section of the original multidimensional state space. The attractors (assuming a steady state) thus created have a dimension decreased by 1 when compared to the original state space.

Using the method of the previous paragraph, Fig. 3(a) shows the attractor reconstructed using the data from Fig. 2(a). The resulting fixed point expresses the dynamical fact that each flash occurs *at the same time* each successive acoustic period—the SL is phase locked to the acoustic field. Using the data from Fig. 2(b), Fig. 3(b) shows an attractor consisting of two points—a *period-doubling* (PD) bifurcation of some sort has occurred. Two acoustic periods must now pass before the flash interval repeats itself. Figure 3(c), using the data of Fig. 2(c), shows that another PD bifurcation has taken place;

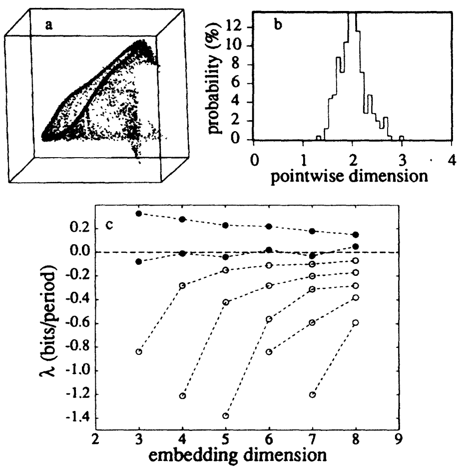

FIG. 4. Chaotic behavior in flash variation Δt. (a) is the attractor reconstructed from the flash time series. (b) is the distribution of pointwise dimensions for (a). (c) shows the results of calculation of the Lyapunov exponents for the attractor in (a).

also, that each "fixed point" of the attractor is the locus of many thousands of individual data tuples, indicating the decreased stability of the period-four state. Finally, Fig. 3(d) [data from 2(d)] clearly shows the existence of a limit cycle, a closed curve instead of discrete fixed points. This indicates that a Hopf bifurcation has taken place; the system dynamics occurs on a two-dimensional torus (a closed curve in the Poincaré section). The flashes are still occurring once every acoustic cycle, but the phase with respect to the acoustic field is now varying at a frequency *incommensurate* with the driving frequency—SL is no longer phase locked.

Figure 3(e) plots the results of calculating the pointwise dimension [9] for the attractor in (d), with the average being almost exactly 1.0. This, along with the zero Lyapunov exponent [10] calculated for (d), confirms that the behavior is *quasiperiodic* (QP), the sum of two pure frequencies (one of which is the driving frequency or an integral submultiple thereof, which is eliminated by our acquisition technique of subtracting the acoustic period T from the measurement).

The surprising appearance of an incommensurate *phase frequency* warrants some further observations. Its dependence on the external parameters is unknown. The phase frequency was not always the same in all time series obtained, but generally fell into two categories: "slow" and "fast" time scales. The slow behavior was an incommensurate modulation of the f_d,$f_d/2$ or $f_d/4$ periodic component. The reconstructed attractor was a single limit cycle in the state space, similar to Fig. 3(d). The fast behavior resulted in each periodic point being expanded into a limit cycle (or something more complicated) in the state space.

Figure 4 illustrates chaotic behavior observed in the SL time series. The attractor reconstructed in 4(a) is no longer a simple closed curve. The pointwise dimension calculated for 4(a) is approximately 2.0. Although the dimension is not fractional, a calculation of the associated spectrum of Lyapunov exponents for trajectories on the attractor 4(a), explicitly shown in 4(c), yields one positive exponent, indicating chaos. The Fourier transforms of the original flash interval data used in both Figs. 3(d) and 4(a) (not shown) reveal a strong two-frequency mixing, with multiple sum and difference peaks. The frequencies, however, are not the same for 3(d) and 4(a).

Discussion: mechanisms for light production.—Viable explanations for the light production in SL include [11] (a) *ionization* of the gas inside the bubble (Wu and Roberts in [4]); (b) *excitation* of vapor species present inside the bubble (Flint and Suslick in [4]); and (c) a "dynamic" *Casimir effect* due to the rapid acceleration of the interface (the bubble wall) between two dielectrics (air and water) (Schwinger in [4]). In this section we will briefly introduce three conjectures which could explain our results and remain in harmony with the body of experimental results of other groups. While the above results will neither disprove nor confirm (a)-(c), they can provide bounds for their sufficiency.

The effects we have seen in the flash interval data which are important in this context are *period-doubling* (PD), *chaos*, and *quasiperiodicity* (QP). Although PD and chaos could in theory be explained by purely spherical oscillations of the bubble [see 12], such behavior has *never* been observed in single-bubble experiments [13-15].

QP *definitely* requires more than spherical bubble dynamics, since a bubble oscillator (without internal spatial dependences) is a strictly dissipative system [16]. We can organize our remaining discussion by considering how our proposed conjectures yield quasiperiodic flash intervals.

Conjecture 1: The bubbles lose spherical symmetry via the onset of the *Faraday instability* [17]. *Stable* shape oscillations of well-defined spatial mode occur even in the isotropic acoustic field via a nonlinear resonant coupling to the volume oscillations. Some recent theoretical work [18] has shown (albeit under rather limited assumptions) that a quasiperiodic modulation of the volume mode can result for such resonant interactions. Experimentally [15], period-doubled, chaotic, and quasiperiodic temporal behaviors have been observed for shape oscillations of different modes in bubbles which were *not* luminescing. Note that this argument is compatible with any of the three mechanisms (a), (b), or (c) for light production—the SL merely "illuminates" the bubble wall mechanics.

Conjecture 2: For this conjecture, we assume that the excitation mechanism theory (b) for SL is the only mechanism operative. In one version of this theory [19], photon emission occurs when an excited state of a vapor species inside the bubble, notably OH^{-*}, decays. The nonlinear chemical reactions governing the concentrations of such species present inside the bubble are periodically forced by the violent temperatures and pressures during the collapse phase. Period-doubling, chaos, and quasiperiodicity have all been observed in concentration oscillations for similar forced systems of chemical oscillations [20]. Thus, (b) alone could account for the SL dynamics reported here.

Conjecture 3: A shock wave propagates inside the bubble (see Greenspan and Nadim or Wu and Roberts in [4]), converging, reflecting at the center, and then diverging. Our idea follows from the fact that the shock's shedding from and subsequent collision with the bubble wall gives rise to a momentum exchange, hence a change in the velocity of the bubble wall. Thus the mechanical oscillation will be slightly phase shifted each acoustic period from the last period. The point in time at which the implosion (and flash) occurs will continually vary, since the relevant time constant, $(R_{\text{shedding}}-R_{\text{return}})/c_{\text{shock}}$ will not bear a constant relationship to the acoustic period. This conjecture would admit ionization and excitation as mechanisms, but would probably rule out Casimir light, since the shock front bounds a discontinuity in the dielectric constant (due to the density discontinuity) which is much less than unity. Schwinger's [4] proposed light energy goes as the square of the difference in dielectric constants.

Conjecture 4: Roy [21] has suggested a form of mode locking between the bubble and the resonant acoustic driving chamber as the source of the remarkable periodic stability that Barber and Putterman observe. The frequency shift (indirectly indicating the necessary nonlinear coupling) of a resonant acoustic standing wave field due to an inclusion (bubble in water, drop in air) is a well-known and, in the drop-levitated-in-air case, well-studied phenomenon [22]. We imagine that detuning of the driving frequency moves the system off of a mode-locked step in the incomplete devil's staircase [23], and into a quasiperiodic state. This final conjecture is compatible with any of the light emission mechanisms presented above.

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[1] N. Marinesco and J. J. Trillat, Proc. R. Acad. Sci. **196**, 858 (1933).
[2] D. F. Gaitan, L. A. Crum, C. C. Church, and R. Roy, J. Acoust. Soc. Am. **91**, 3166 (1992); B. P. Barber and S. J. Putterman, Nature (London) **352**, 318 (1991).
[3] K. J. Taylor and P. D. Jarman, Aust. J. Phys. **23**, 319 (1970); E. B. Flint and K. S. Suslick, Science **253**, 1397 (1991); R. Hiller, S. J. Putterman, and B. P. Barber, Phys. Rev. Lett. **69**, 1182 (1992).
[4] M. Degrois and P. Baldo, Ultasonics **12**, 25 (1974); E. B. Flint and K. S. Suslick, J. Am. Chem. Soc. **111**, 6987 (1989); H. P. Greenspan and A. Nadim, Phys. Fluids A **5**, 1065 (1993); C. C. Wu and Paul H. Roberts, Phys. Rev. Lett. **70**, 3424 (1993); J. Schwinger, Proc. Natl. Acad. Sci. U.S.A. **89**, 11118 (1992); **90**, 958 (1993).
[5] L. A. Crum, J. Acoust. Soc. Am. **95**, 559 (1994).
[6] R. Löfstedt, B. P. Barber, and S. J. Putterman, Phys. Fluids A **5**, 2911 (1993).
[7] There is no simple conception of a static "equilibrium radius," due to passive and convection-aided mass diffusion across the bubble/water interface. The long-lived existence of bubbles in high-intensity acoustic fields implies some dynamic equilibrium state exists. This would depend in a complicated way on nonlinear mechanical resonances of the bubble, as well as on parameters such as the external pressure and frequency of the acoustic field, and material parameters (solubility, dissolved gas concentration, temperature). See C. C. Church, J. Acoust. Soc. Am. **83**, 2210 (1988).
[8] N. H. Packard *et al.*, Phys. Rev. Lett. **45**, 712 (1980).
[9] J. Holzfuss and G. Mayer-Kress, in *Dimensions and Entropies in Chaotic Systems*, edited by G. Mayer-Kress (Springer, Berlin, 1986), p. 114.
[10] J. Holzfuss and W. Lauterborn, Phys. Rev. A **39**, 2146 (1989).
[11] The coronal discharge theory has recently been rehabilitated [T. LePoint and F. Mullie, in Proceedings of the 3rd Meeting of European Society of Sonochemists, 1993 (to be published)].
[12] A. Prosperetti, L. A. Crum, and K. W. Commander, J. Acoust. Soc. Am. **83**, 502 (1988); U. Parlitz *et al.*, J. Acoust. Soc. Am. **88**, 1061 (1990).
[13] R. G. Holt and L. A. Crum, J. Acoust. Soc. Am. **91**, 1924 (1992).
[14] J. Holzfuss and W. Lauterborn, Phys. Lett. A **115**, 369 (1986).
[15] R. G. Holt *et al.*, in *Proceedings of the 12th International Symposium on Nonlinear Acoustics, Austin, Texas*, edited by M. F. Hamilton and D. T. Blackstock (Elsevier, New York, 1990), p. 497.
[16] J. Guckenheimer and P. Holmes, *Nonlinear Oscillations, Dynamical Systems and Bifurcations of Vector Fields* (Springer, Berlin, 1983).
[17] John Miles, J. Fluid Mech. **248**, 671 (1993).
[18] C. C. Mei and X. Zhou, J. Fluid Mech. **229**, 29 (1992); S. M. Yang, Z. C. Feng, and L. G. Leal, J. Fluid Mech. **247**, 417 (1993).
[19] V. Kamath, A. Prosperetti, and F. N. Egolfopoulos, J. Acoust. Soc. Am. **94**, 248 (1993).
[20] J. Maselko and H. L. Swinney, J. Chem. Phys. **85**, 6430 (1986); D. Barkley, J. Ringland, and J. S. Turner, J. Chem. Phys. **87**, 3812 (1987).
[21] R. A. Roy (private communication).
[22] E. Leung, C. P. Lee, N. Jacobi, and T. G. Wang, J. Acoust. Soc. Am. **72**, 615 (1982).
[23] The generic model for such behavior is the *circle map*, introduced by V. I. Arnold, Am. Math. Soc. Trans. **46**, 213 (1965), and numerically studied by M. H. Jensen, P. Bak, and T. Bohr, Phys. Rev. A **30**, 1960 (1984). Note that the rational step [heuristically defined as (mode of chamber)/(mode of bubble)] upon which stable, period-1 SL occurs may not be the 1/1 resonance: Independent confirmation of R_0 would help determine this ratio. If it is defined as (driving frequency)/(response frequency), it would appear to be $\sim 1/10$ in the experiments of Barber and Putterman [2].

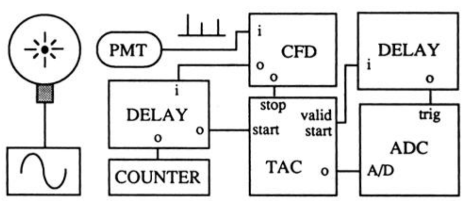
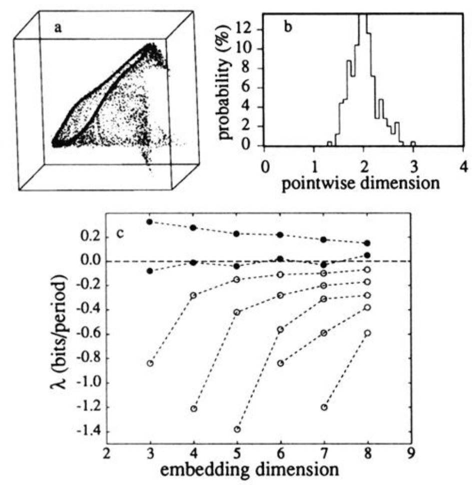


FIG. 1. Schematic of the experimental apparatus.



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