Period-Doubling Bifurcations from Breaking the Spherical Symmetry in Sonoluminescence: Experimental Verification

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Using a fiber-based four-channel correlation scheme to investigate spatial and temporal correlations, we show that observations of period-doubling phenomena in single bubble sonoluminescence are primarily a result of spontaneously breaking the spherical symmetry in the bubble collapse and, at most, may show up as secondary effects in the flash-to-flash spatially integrated light output.

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Period doubling (PD) and chaos are common phenomena in nonlinear systems. Single bubble sonoluminescence (SBSL) [1] represents an extreme example of such systems. Here we present evidence that these phenomena in SBSL are linked to spontaneous breaking of the spherical symmetry, giving additional evidence [2] that stable SBSL does not necessarily imply perfect spherical bubble collapse.

Sonoluminescence from a single gas bubble levitated in a liquid by ultrasound has been a fascinating subject of recent studies. At high values of the driving sound field, the oscillations of the bubble can get so violent that in each period the gas in the bubble is compressed close to its van der Waals hard-core radius. In the process, the gas heats up to an extent which leads to light emission while the bubble is smallest (for recent reviews, see [3,4]).

The first experiments on single bubbles reported a synchronicity with the drive (typically ≈ 27 kHz) of the order of 50 ps, showing a remarkably stable dynamics [5]. Later Holt *et al.* reported PD, quasiperiodicity, and chaos in the timing of successive flashes [6]. Jensen observed a single PD directly in the pulse heights (i.e., the height alternates between two values) and a quadruple peak in the pulse height statistics [7]. A double peak in the statistics was reported by Ketterling and Apfel [8], who also cite Gaitan and Holt for this observation. However, these observations were all done using a single photomultiplier, and the underlying mechanism responsible for the observed behavior has yet to be identified.

Lauterborn and Suchla [9] investigated a dynamical bubble model in the context of acoustic cavitation [10,11] and showed that the radial oscillations of a gas bubble can undergo PD and become chaotic. It was proposed [6] that the same mechanism could explain the observations in the SBSL measurements; however, the numerically studied parameter space [9,10] is, alas, irrelevant for SBSL. A detailed study on SBSL based on the Rayleigh-Plesset equation [12] was performed by Simon *et al.* [13]. This study showed that the parameter space for PD is far from that where light emission takes place. Also found was that the afterbounces were involved in these models in catalyzing the instabilities. In the regime of SBSL, the afterbounces die out long before the next collapse and thus can hardly provide the necessary memory. A suggestion in the latter paper pointed to the returning echo of the bubble collapse as a possible memory mechanism (see also [14]). However, these model calculations all assume that the bubble collapse preserves spherical symmetry; i.e., the bifurcations are solely due to a size effect. In the correlation measurements presented here, contrary to this assumption, we find that instabilities leading to pulse height PD are always associated with a break in the spherical symmetry of the collapse and will, at most, show up as secondary effects in the spatially integrated light output per flash if at all.

The vessel used is a 6.5 cm diameter glass sphere hanging acoustically isolated in a 7 mm inner diameter neck tube. A pair of piezoelectric transducers is glued on to opposite sides with epoxy. The drive signal of approximately 25 100 Hz is delivered by a computer controlled HP 33120A function generator through a power amplifier and tuning circuit. The flask is filled with outgassed distilled water and placed in a modified refrigerator. After cooling to a temperature of approximately 5 °C and allowing for temperature equilibrium to be reached, a bubble is generated by a computer controlled blast of air on the free surface. The computer is programmed to automatically redo this, if bubble extinction occurs (or none is generated), and also automatically search for stable light emission above a preset intensity limit.

The correlation setup consists of four quartz fibers of 1 mm diameter leading the emitted light to four photomultiplier tubes (PMTs) (Hamamatsu H5783P-03, rise time 0.65 ns). The fibers are placed at well defined longitudes in the equatorial plane of the sphere pointing at the bubble from a distance of $\sim$4.5 cm. Using four fibers allows for simultaneous measurements of cross correlations involving six angles. This creates a comprehensive spatial knowledge simultaneously for all these angles. The signals from the PMTs are amplified by preamplifiers and shaping amplifiers (shaping time 3 μs) and fed to a 20 MHz four-channel simultaneously digitizing analog-to-digital (A/D) data acquisition card (ADLink 9810). A fifth photomultiplier tube that looks directly at the bubble provides the digitizer timing signal, which is delayed so the A/D card measures the peak value of the pulse. The reliability of the trigger system has been carefully checked to ensure exactly one sampling per flash. The peak value of the pulse observed in each channel for every flash is recorded in time series of length 2 min ($\approx 3 \times 10^6$ flashes). To avoid cross talk between channels, all PMTs and amplifiers have separate power supplies and the performance of the system has been carefully checked in trial runs.

Frequently, time series obtained in this fashion showed that the light emission was period doubled. In the raw time series, a sliding point average (of the order of 100 points or more) for odd and even times separately was necessary to observe this, and, although the PD could be present for minutes, the phase would slip. Bifurcations are seen in all channels simultaneously. However, individual time tracks are rather confused, mostly due to the small number of photons (notice that pulse height depends on the wavelength of the detected photon) detected from a single pulse as the solid angle seen by the bubble is very small, but also due to the statistical nature of the PMT detection process. Apart from long time oscillations of frequencies of a few Hz, the output is stable and the bubble can survive for many hours without extinction.

The picture becomes much clearer when we look for time dependent and spatial correlations. These are calculated as

$$C_{k,l}(t) = \frac{\sum_{m=1}^{N}[CHk(m) - \overline{CHk}][CHl(m+t) - \overline{CHl}]}{N\overline{CHk}\,\overline{CHl}}, \tag{1}$$

where N is the total number of samples in the two channels CHk and CHl ($k, l \in \{0, 1, 2, 3\}$) over which the correlation is being calculated. Here m denotes a specific sample and t the offset time normalized by the sampling interval. $\overline{CHk}$ is the average of $CHk(m)$ over all samples in the time series. (Notice that higher than average signals in both channels or lower than average signals in both channels both give rise to positive contributions, while higher than average in one channel together with lower than average in the other channel give a negative contribution to the correlation.) The results presented in the following are calculations on a single time series ($\approx 3 \times 10^6$ flashes). Different symbols are used for correlations belonging to odd and even time steps for clarity.

In Fig. 1 is shown the autocorrelation [$k = l$ in Eq. (1)] for four channels placed in the horizontal plane of the bubble at respective longitudes CH0: 30°, CH1: 75°, CH2: 90°, and CH3: 0°. Several features are conspicuous. Most obvious, the curves are doubled, meaning that the temporal correlations for odd offset times are different from those with even offset times; i.e., we have period doubling. Differences up to 40 parts per thousand have been observed, though values around 10 are more common. Moreover, the size of the effect is independent of the channel. So either we see a radial PD in a spherical collapse or an averaging over space due to no preferred direction of a possible PD shape distortion.

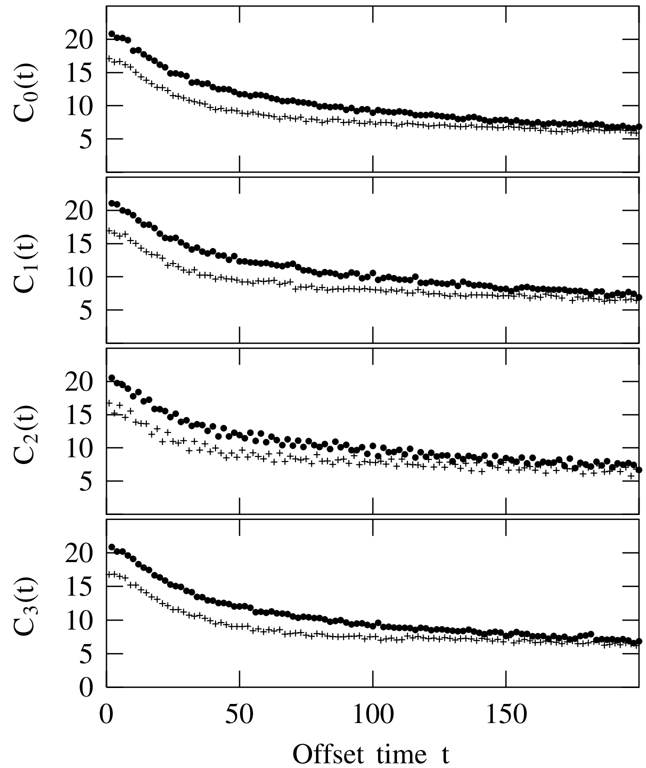

FIG. 1. Autocorrelations (parts per 1000) versus time. Period doubling is seen as a splitting in correlations for odd and even values of t. Odd time steps (+) and even time steps (•) are given different symbols for clarity.

To distinguish between these possibilities, we have to look at cross correlations [$k \neq l$ in Eq. (1)] between channels (Fig. 2) that also provide spatial information. With four channels, six different angles are involved. Different symbols are again used for clarity to designate correlations belonging to odd and even offset times. For t below ≈ 200, the even offset time correlations are seen to lie above the odd offset time correlations for angles below $\sim 50°$, but below for angles above. This is easily seen if the difference between correlations belonging to consecutive even and odd offset times (see Fig. 3) is plotted instead. Measurements on different configurations of fiber positions show that the situation is symmetric around 90°. Figure 3 also shows that the lifetime for the bifurcated state in this time series is about 300 flashes. Consistency checks, such as, e.g., interchanging positions of fibers, all validate these results.

The angular dependence of the cross correlations means that if an observer A sees a strong flash, then an observer B placed at 180° measured from A with respect to the bubble simultaneously also most likely will see a strong flash, while an observer C placed at an angle of 90° is more likely to see a weaker signal. In the case of PD, one time step ahead the situation is reversed, and thus it keeps alternating for progressive time steps. Moreover, there is an angle of $\sim 50°$ where the flash most likely will be of an

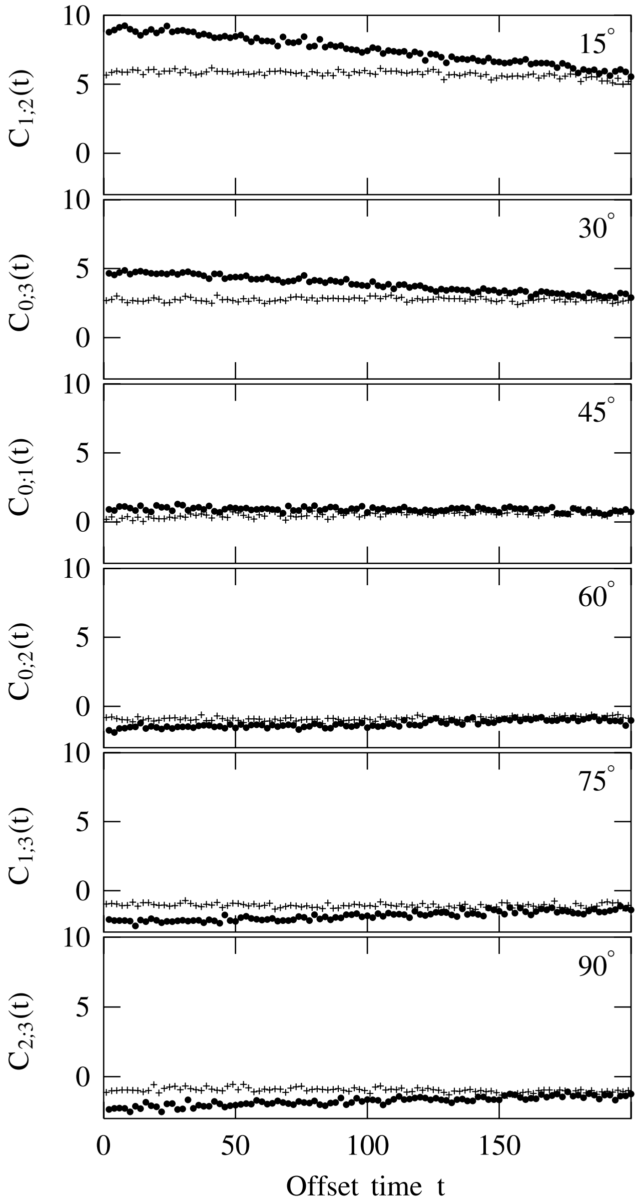

FIG. 2. Cross correlation (parts per 1000) versus $|t|$ for different angles, $\Theta_{k,l}$. The period doubling is evidently predominantly geometric as even time correlations are larger for small angles (below 45°) and smaller for large angles (above) than uneven time correlations. Odd time-steps (+) and even time-steps (•) are given different symbols for clarity.

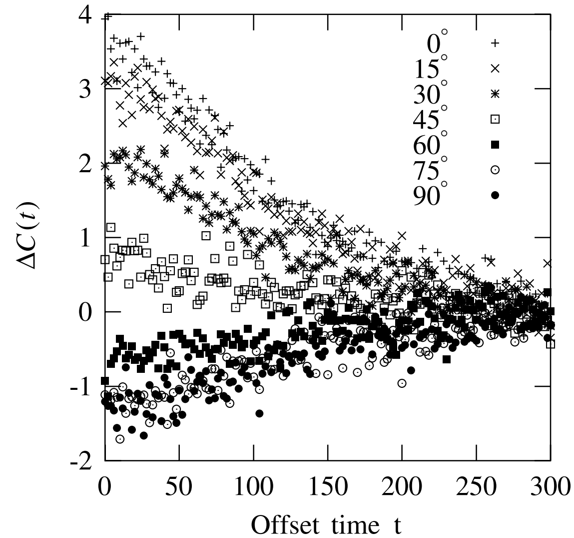

FIG. 3. $\Delta C(t) = C(t) - C(t-1)$ (parts per 1000) (t even) plotted as a function of t for different angles. The geometric nature of the bifurcation shows up here, as small angles have a positive ΔC and larger angles have a negative ΔC. A PD of the total intensity from the bubble would result in a positive ΔC for all angles.

intermediate magnitude. This observation is linked to the relative positions in space. If the bubble collapse was spherical symmetric, all observers would most likely see strong (or weak) flashes simultaneously. A translational movement in space also affects correlations. However, if A sees a strong flash because the bubble moves closer to A, then B most likely would see a weak flash as the bubble moves away [15]. From symmetry arguments, these possibilities must therefore be excluded as explanations of the observed PD.

The only way to interpret these results is that the PD is geometrical in nature, i.e., is breaking the spherical symmetry in the bubble collapse. This means that the bubble shape is distorted, and that the distortion alternates in size from flash to flash. Clearly a memory effect must be present to give rise to the existence of PD states. Most likely, the distortion in shape is reflected in the velocity field of the surrounding fluid [16], but also the returning echo from the previous bubble collapse may play a role as shown by a simulation on a simple model [17].

Although the main conclusion that the PD bifurcation is geometrical in nature seems solid, there are still unsolved puzzles presented by the data shown. The answers to these must rest on the specific nature of the geometric forms of the two states involved in the PD. Presumably, the distortions involved are spherical harmonics [18,19]. Measurements by Weninger *et al.* [15] using a two point angular correlation setup suggested that a dipolar state with random orientation ($n = 2$ spherical harmonic) could be excited (see their Fig. 1). As can be seen from the sign change of $C(t)$ for both odd and even offset times as the relative angle is increased (see Fig. 2), the basic state is predominantly an $n = 2$ state. On top of this, as seen from Fig. 4 where we have plotted the difference in cross correlation between even and odd time steps (average over $t = 2$ to 20 time steps) as a function of angle, we have the period doubling as an $n = 2$ state with the sign alternating with period 2. The data points fit reasonably well to the equation $C_{k,l}(t) \sim [1 + 3\cos(2\Theta_{k,l})]$ calculated by Weninger *et al.* for the emission from an $n = 2$ state. The fit assumes a random orientation of the excited state. Strictly speaking, this cannot be true since the phase of the PD is sustained for hundreds of cycles. Only over longer times is the motion uncorrelated, resulting in the size of the effect being the same for all channels as seen in the autocorrelations.

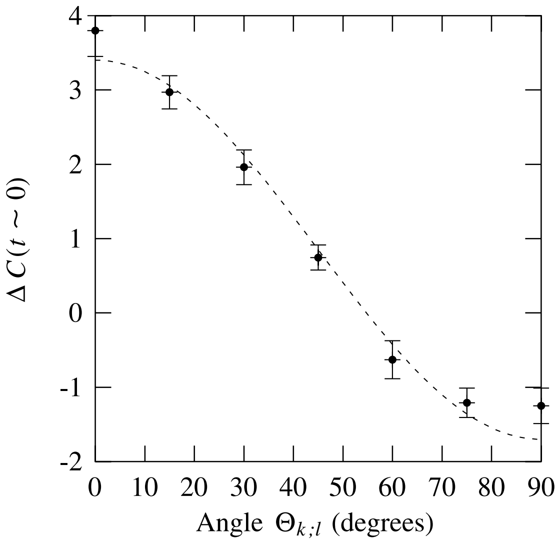

FIG. 4. The mean value of $\Delta C(t)$ (parts per 1000) for $t = 2$ to 20 plotted as a function of angle. For the angle 0° the mean value of the autocorrelation from the four channels is used. The dashed line is $0.000\,85[1 + 3\cos(2\Theta_{k,l})]$.

An open question is whether the correlation measurement can tell anything about the mechanism giving light, the extent of the emitting region, and the opacity of the bubble. However, the angular dependence of the cross correlation in the small wavelength limit resulting from either the diffraction of the light pulse at the bubble surface, as suggested by Weninger *et al.* as a possible explanation, or from surface radiation is nearly identical for not too large ellipticity and a relative refractive index (water/air) of 1.33 (unless one assumes complete incoherence and the surface an ideal Lambertian diffuser, in which case there is no angular dependence). No conclusion can thus be reached on the question of the degree of opacity inside the bubble before the index of refraction inside the bubble at the time of emission is known, although certainly the strong compression of the gas in the bubble will increase the refractive index of the gas, thereby reducing the effect of light diffraction.

An extended account relating to these questions and of measurements showing a preferred direction in space for very stable PD states is in preparation. This will also include results on non-PD states.

To conclude, we have performed spatial and temporal correlation measurements on the light emission from a sonoluminescing bubble using a four-channel fiber-based detection system. These measurements show unequivocally that PD is observed as a result of spontaneous breaking of spherical symmetry in the bubble collapse and is not seen in the light output per flash integrated over space. That the distorted states can survive for a very long time (hours) provides additional evidence that stable SBSL does not imply perfect spherical bubble collapse. The picture evolving is that the bubble first experiences excitation of a state of broken spherical symmetry (Refs. [8,15]) that is then subjected to a second (pitchfork) bifurcation into a PD state. Thus the explanation for these and previous observations [6–8] on PD and chaos should rather be sought in the three-dimensional Rayleigh-Plesset equation (see, e.g., [18,19] for a theoretical treatment of symmetry broken states) than in the symmetric radial Rayleigh-Plesset equation [9,10,13] that is essentially one-dimensional. We have looked for evidence for higher period states and have some evidence for a second PD (i.e., period 4), but no odd period states have been seen. A memory effect has to be involved, and we believe that either the flow field around the bubble [16] or the echo of the shock wave emitted into the water [13,14] are involved.

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