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Direct observation of period-doubled nonspherical states in single-bubble sonoluminescence

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We present direct observations of period doubling in the flash to flash pulse heights in single-bubble sonoluminescence. States involved are stable, spherically symmetry broken. Observations are made using seven detectors distributed in the equatorial plane of the bubble. Contrary to earlier experiments by Holt *et al.* [Phys. Rev. Lett. **72**, 1376 (1994)], where period doubling was observed in the time intervals between flashes but *not* in the pulse heights, we observe period doubling in pulse heights, but *no* corresponding period doubling is seen in the time intervals. In parameter space the period doubling is observed below the $n=2$ shape instability boundary line where extinction is shown to take place.

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Single-bubble sonoluminescence (SBSL) [1], where a bubble caught in the pressure anti-node of an intense resonant sound field emits flashes of light with the periodicity of the sound field, is one of the most extreme nonlinear systems ever known. For reviews see Refs. [2–4].

Not surprisingly, effects such as shape distortion (shape instabilities) and period doubling are encountered in SBSL (see, e.g., Refs. [5–10]). From a practical view the efficiency of the bubble as a chemical reactor is expected to decrease since the energy will be less focused, thus effectively lowering the resulting temperature inside the bubble. This should then give rise to a smaller light output (see Ref. [8] though). Recently, we have shown that the period doubling implicates a periodic change between two spatially anisotropic distributions of the light intensity. However, in these experiments statistical methods were necessary to discern the period doubling [9,10].

Here we report measurements of stable geometric period doubling seen directly in the flash to flash intensity without any resort to statistical methods. In the present context, by period doubling we mean a periodic change between 2^m different intensity levels such that $I(\vec{r},t_{n+2^m})=I(\vec{r},t_n)$, where $I(\vec{r},t_n)$ is the intensity at flash time t_n in the spatial direction $\vec{r}$ and $m\geqslant 1$ a positive integer. The obvious implication is that the bubble collapses nonspherically. Surface diffraction of light [6] emitted from a (distorted?, blackbody?) central hot spot or partial correlation by stimulated processes could then result in an anisotropic intensity distribution. While the previous experiment by Weninger *et al.* [6] on anisotropic intensity distribution has largely been ignored, perhaps because of a heavy reliance on statistical interpretation (and measurements at different angles being done sequentially), stable anisotropic emission that may even be period doubled does raise questions that any plausible theory must be able to answer.

In our previous experiments the 2 cycle ($m=1$) was often preceded by a shape distortion. While the uncertainty on a single flash in these experiments was too large for the period doubling to be seen directly due to the very small solid angle (0.4 msrad) under which the detectors were viewing the bubble, the very long time series obtainable made the observation of the shape distortion possible even for a spatially stable distortion. The case here is the reverse, so whether the period-doubled state is actually preceded by a shape distorted state is unknown. On the other hand, the stability of the system allows us at the same time to measure the timing of the flashes. In the first experiment [5] reporting period doubling in sonoluminescence, the effect was seen *only* in the timing of the flashes and *not* in the pulse heights. Contrary to these authors we observe the effect *only* in the pulse heights but *not* in the timing. This is the case for both the cylindrical cell used here and for the spherical cell used in our previous experiments [9,10]; thus we believe the effect to be cell independent.

The vessel used is a quartz cylinder of 6.5 cm outer diameter and wall thickness 2 mm. The height is 6.0 cm. Top and bottom are made of aluminum with a pair of piezoelectric transducers glued on with epoxy. The drive signal is delivered by a computer controlled HP 33120A function generator through a power amplifier and tuning circuit. Bubbles are generated through electrolysis by applying 30 V to a pair of electrodes for a few seconds. A distilled water sample was prepared with a partial air pressure overhead of 214 mbar (after correcting for water vapor pressure) at a temperature of 29 °C. Since air contains 0.93% of argon, this corresponds to a partial argon pressure overhead of 1.99 mbars and an argon concentration of $C_i=0.00196C_0^{29}$, where C_0^{29} is the equilibrium argon concentration in water at temperature of 29 °C and ambient pressure of 1 atm (see, e.g., Ref. [11] for the temperature dependence of C_0). The water was transferred to the resonator using gravity flow. During water preparation and filling the gas pressure in the resonator was kept at the degassing pressure to ensure a gas concentration in the resonator equal to the desired preset value. After filling the resonator was subsequently cooled to the operating temperature of 9 °C allowing for thermodynamic equilibrium to be reached. At the operating temperature the resonance frequency is ≈22.165 kHz and the relative argon concentration is $C_i/C_0^9=0.00135$.

The detection system consisted of seven photomultiplier

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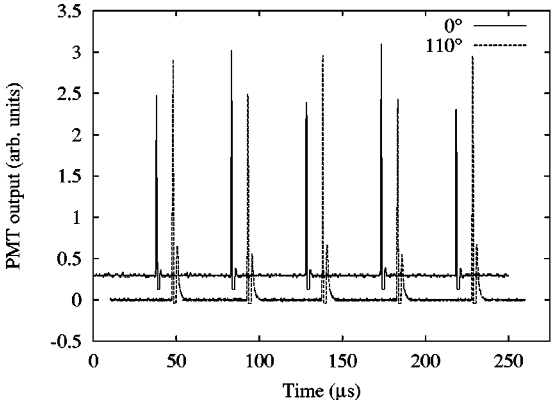

FIG. 1. PMT signals from two channels 110° apart. The upper trace has been displaced up and to the left for easier viewing. The period doubling is seen to be out of phase showing it to be a geometric effect.

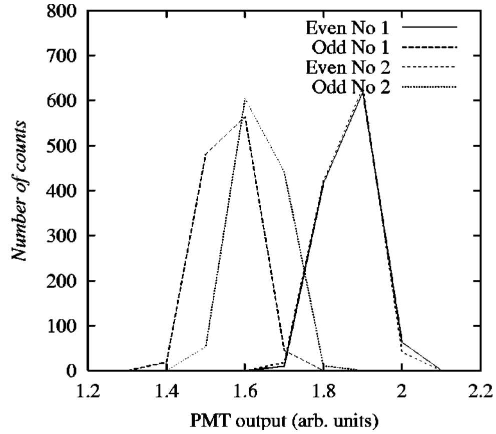

FIG. 2. Distribution of the odd and even numbered peak heights from one channel for tracks taken 2 min apart.

tubes (PMT Hamamatsu R3478, rise time 1.3 ns) situated in the horizontal plane of the bubble viewing it at a solid angle of 50 msrad and placed at relative longitudinal angles of 0°, 60°, 100°, 140°, 180°, 220°, and 280°. Using seven detectors allows for a simultaneous measurement of intensities creating a comprehensive spatial knowledge. The signals from the PMTs are amplified by time-shaping amplifiers (shaping time 0.1–3 μs) and fed to two 100-MHz 4-channel oscilloscopes (HP 54624A). Time series containing about 2000 flashes can be obtained with a reasonable resolution and the peak value $I(t_n)$ of the pulse observed in each channel for every flash calculated. The direct signal from one channel is furthermore led to a 500-MHz oscilloscope (HP 54616C).

In Fig. 1 we display a few periods of the raw PMT signal from two of the channels placed 110° apart in a situation of stable period doubling. For clarity the upper trace is displaced up and to the left. The period doubling is easily discernible directly in the signals and also the fact that the two signals are in antiphase with respect to the period doubling. Thus this is a geometric effect. The direction in space of the period doubling can be stable for minutes. A histogram of peak heights shown separately for even and odd numbered flashes over $\approx$2000 periods for two tracks (No 1 and No 2) taken 2 min apart is displayed in Fig. 2 to give an idea of the stability. Even after more than 2.5×10^6 collapses the period doubling is very clear. The higher intensity peak is nearly unchanged while the lower intensity peak has shifted a bit up.

The difference $\Delta I=\langle I_{even}\rangle-\langle I_{odd}\rangle$ between the separate averages of the even I_{even} and odd time I_{odd} flash intensities (taken over $\approx$2000 periods) is displayed in Fig. 3 as a function of channel position. Within the uncertainty the data can be fitted to $a\cos(2\Theta-2\Theta_0)+b$, with Θ being the longitude of the different PMT's, $\Theta_0=108$ the angle of maximum ΔI, and $a=0.097$ the amplitude of ΔI normalized with the average intensity. The offset $b=-0.028$ is a result of the total amount of light emitted in the plane of measurement, not necessarily being the same for odd and even flashes. The frequency range of the drive where, without changing the amplitude of this, we can observe the stable period-doubled state is $\approx$6 Hz. By further detuning, tumbling, i.e., a slow change in the directions of the symmetry axes of the geometric period-doubled state, can be induced.

These experimental results show directly that stable bubble states exist that are shape distorted at the instant of light emission, and that these can undergo period doubling. The direction of the symmetry axis does not seem to be bound to any fixed direction in space but seems to be randomly chosen whenever the bubble is brought into the regime of the period-doubled state anew. Also at some choices of parameters, the period-doubled state exists in a bounded regime of amplitude variations.

In the following we shall investigate the connection of our experiment to the other independent report documenting period doubling in SBSL. The first experiment found period doubling in the interflash timing $\Delta t_n=t_{n+1}-t_n$ alone but not in the intensity of the flashes [2]. If averaging was necessary this is understandable. However, several other explanations can be found for the lack of observation of an intensity variation. First, a single PMT was used. Thus this could accidentally be placed looking at a node in the intensity field from a spatially stable period doubling; or if the symmetry axis is tumbling, the resulting variation in the intensity could be interpreted as just noise fluctuations. Also, the statistical noise from the photon detection process itself could have

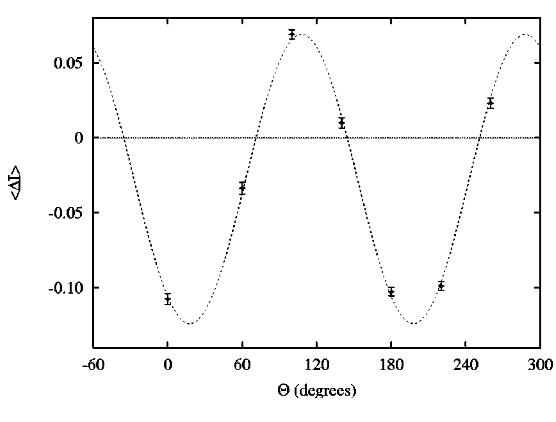

FIG. 3. Angular variation of the intensity difference between the even and odd timing averages, fitted to $a\cos(2\Theta-2\Theta_0)+b$.

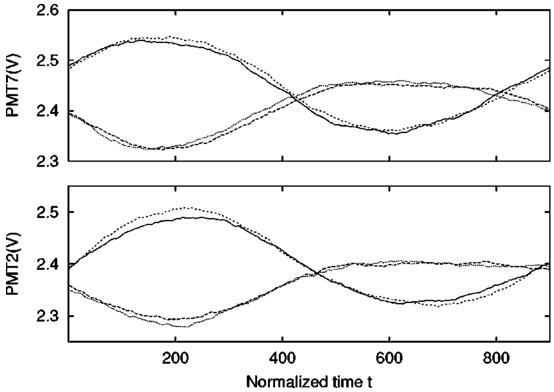

FIG. 4. Observation of 4 cycle for two channels spaced 160°.

masked the effect especially if too large a solid angle was covered by the PMT, or the measurement is made at room temperature, as in Ref. [5]. It is therefore natural to look at the interflash timing in the present experiment.

Needless to say, there is no difference in the timing of the flashes seen by the different PMT's, but rather surprising is that there is no period doubling seen in the interflash timing either. A careful inspection with the 500-MHz oscilloscope shows this to be true within 2.5 ns. In fact, we have never seen the slightest evidence of a splitting in Δt_n. This should be compared to the more than 100 ns difference in Δt_n for a 2 cycle ($\Delta t_{n+2}=\Delta t_n$), and even a microsecond long time difference for a 4 cycle, i.e., $\Delta t_{n+4}=\Delta t_n$ measured by Holt *et al.*, Ref. [5]. Thus, it is an open question whether the effects seen in the two experiments are actually related.

To emphasize that we do indeed see the beginning of a Feigenbaum tree of period doubling, in Fig. 4 we display traces of PMT output peak values from two channels (PMT2 and PMT7) displaying a 4 cycle ($m=2$). A 50 point running average is taken for each cycle point independently since the statistics do not allow for a clear direct observation. A slow change in the axes of orientation is apparent in the crossings of the tracks, differing for the two channels as they are spaced 160° apart.

Earlier attempts to explain period doubling in SBSL have centered on comparisons with the one-dimensional radial Rayleigh-Plesset equation. However, the parameter space where this is susceptible to period-doubling instabilities is far from overlapping with the parameter space where SBSL is encountered [12]. Nevertheless, some of the additional feedback mechanisms suggested for period doubling to be found for spherically collapsing bubbles may still be relevant for the nonspherical case. One such mechanism is the coupling to the motion of the fluid around the bubble which in turn is induced by the growth and collapse of the bubble [13]. Another mechanism is the back coupling in the following cycle of the shock-wave emitted into the fluid by the bubble collapse [12,14]. The latter mechanism has been shown by simulations on a simple model to be able to induce period doubling in the regime of sonoluminescence [15]. A third mechanism is the detuning of the drive frequency from the resonator frequency [5]. The last mechanism (and probably the others, too), though, would most certainly involve the timing of the flashes.

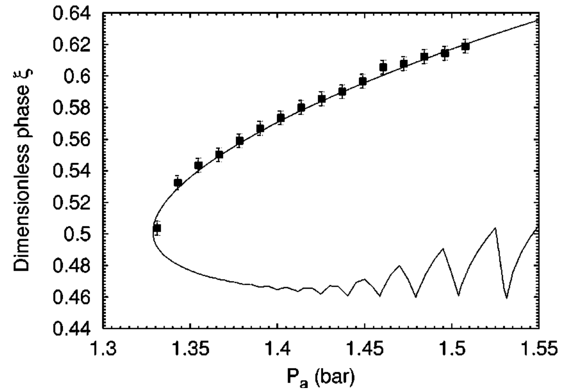

FIG. 5. Fit relating the measured phase shift and drive amplitude to the acoustic pressure. The dimensionless phase is defined as $\xi=\Delta t/T$, where Δt is the elapsed time from the beginning of the acoustic period T until the instant of the flash.

We have therefore measured the timing as a function of frequency detuning. While this changes the relative phase of the collapse with respect to the drive, the time between flashes is still constant equal to the drive period within the digitizing error of 2.5 ns.

In order to understand the mechanism behind the period doubling, it is essential to determine where in the R_0, P_a parameter space the operating point of the bubble is situated. Here P_a is the acoustic pressure and R_0 is the ambient bubble radius. These parameters can be determined by the method described in Sec. IV of Ref. [16]. More details on the method can be found in Ref. [17].

After locating the operating point and stability range for a stable period doubled-bubble, the amplitude of the drive was lowered while measuring corresponding values of the drive amplitude, the pulse height, and the phase of the flash with respect to the drive. Care was taken that the flash height had stabilized before the measurement was performed. The result for the phase of the flash is displayed in Fig. 5. We would

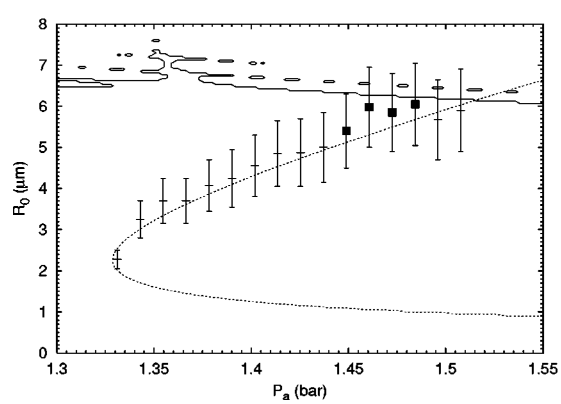

FIG. 6. The results for the acoustic pressure P_a and ambient radius R_0 parameters found from the fit of Fig. 5. The numerical diffusive equilibrium curve (dotted line) is within the error bars for all the data points, implying the self-consistency of the fit. Also shown is the boundary for the parametric shape instability (continuous line). The period doubling (■) is observed below the regime of parametric instability.

like to point out that the upper boundary of the range of period doubling can at least sometimes be reached before the bubble actually breaks up. Also, we would like to point to the rather unexpected continuing increase of the space/time averaged light intensity with the drive amplitude even after the bubble becomes shape distorted.

Following the procedure of Refs. [16,17], we have fitted the data to the Rayleigh-Plesset (RP) equation in order to determine the phase diagram. We used the particular form of the RP equation which incorporates heat transfer through a variable polytropic exponent [18]. The result is displayed in Fig. 6.

In this plot we have also inserted the stability curve relating to the parametric shape instability. At ambient radius values above this line initial small shape distortions grow exponentially to the size of the bubble in a few acoustic cycles. The curve was calculated with the model of Ref. [19] using the same external parameters including the preset argon pressure ($C_i/C_0^9=0.00135$) as in the experiment and assuming a zero boundary-layer thickness, a choice motivated by the work of the authors of Ref. [20], who find this to give the best agreement with experiments. From Fig. 6 it is apparent that this model indeed can accurately predict the extinction threshold of SBSL, as also found in this experiment.

To summarize, we observe spatial period doubling that is stable in both phase and direction in space for minutes. The parameter range for period-doubling is located below the parametric shape instability threshold and is in some cases separated from this by a narrow non-period doubled regime. This seems to indicate that the two shape instability phenomena are not linked. Furthermore, whenever the parameters for period doubling are preset, any newly seeded bubble will go directly into this state within a few seconds, evidence that the period-doubled state is intrinsic to SBSL. Finally we find no connection to the previous observations of period doubling in the timing of the light pulses.

In conclusion, we want to emphasize that any plausible explanation for sonoluminescence has to be able to explain the observation of anisotropy in the light. In fact, the most interesting aspect of our findings may not be the period doubling itself but the constraints that the very observability sets on the theory for SBSL.

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