Second Mode of Recycling together with Period Doubling Links Single-Bubble and Multibubble Sonoluminescence

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We report the existence of a second type of recycling mode that occurs for air-seeded bubbles. Observation of period doubling in both the stable, the first type, and the second type of recycling mode, together with simultaneous measurement of the relative phase of light emission compared to the drive, shows that the instability boundaries of period doubling and bubble extinction are mainly determined by the bubble size irregardless of the gas composition. The second type mode seems to represent a link between single-bubble and multibubble sonoluminescence.

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The collapse of the bubble in single-bubble sonoluminescence (SBSL) [1] is an extremely nonlinear phenomenon. Driven close to the boundary for extinction, stable anisotropic emission can be observed [2], and a further increase in ultrasound power will lead to destruction of the bubble due to uncontrollable growth of shape instabilities. While the dynamics and chemical processes in SBSL are fairly well understood, the connection to multibubble sonoluminescence (MBSL) is still unclear. Here we present observations on a new mode of recycling SBSL (type 2) that seems to represent a link between air-seeded SBSL and MBSL.

Both argon-seeded bubbles [3] and air-seeded bubbles [4–6] may survive in a recycling mode (type 1 Refs. [4,5]) at sound pressures close to the extinction level when partial gas pressure in the liquid is high. The second type of recycling is observed for partial gas levels between approximately 25% and atmospheric pressure. At 25% the mode exists in a very narrow band of sound levels at even higher levels than the type 1, but at higher gas levels the band widens up with a hysteretic overlap to the type 1 mode. The size of the equilibrium radius at 25% is found to be $\sim 6\ \mu$m and the sound pressure at the threshold to be 1.5 atm at the instability level in a previous experiment. As we have shown for air-seeded stable and type 1 SBSL [2,7,8], period doubled light emission is observed for the type 2 mode. Since the emission in all cases is spatially anisotropic, this phenomenon is presumably coupled to shape distortion.

The type 2 mode is a true single-bubble mode in the sense that with a resolution of 2.5 ns we never see more than one flash per sound cycle. If the light emitting bubble disappears, a new bubble has to be introduced by external means. This then enters in the previous mode of operation if outside the region of hysteresis. Thus the type 2 mode is completely reproducible. In the following we present studies of the anisotropic period doubling, spectra, spatial stability, and relative phase of the type 2 mode.

The type 1 collapse is believed to be connected to pinch-off (and possible recombination) of microbubbles, which in the case of air-seeded bubbles can lead to a renewed intake of air. According to the dissociation theory [9,10], the surplus of nitrogen and oxygen is burned off while the level of argon builds up to its former level through rectified diffusion. In turn this leads to a slow increase in the light emission until the cycle repeats. The gas in the surviving part of the bubble thus mainly consists of argon. In what follows we shall present arguments that the new recycling mode is related to mostly air filled bubbles that reach the instability threshold before the burn off becomes efficient.

The vessel and the filling procedure is described in detail in a previous publication [8] with experiments performed at drive frequency 22.114 kHz and temperature 9 °C. However, the detection and data collection systems have undergone several changes. The detection system consists of eight photomultiplier tubes (PMT Hamamatsu R3478 & R2076, rise time 1.3 ns) to provide spatial information. These are nearly evenly distributed in the horizontal plane of the bubble viewing it at a solid angle of approximately 0.050 sr. The positioning of narrow band optical filters in front of the PMTs allows for the recording of an eight point frequency spectrum. The signals are fed to preamplifiers (Hamamatsu C7319) and then to two simultaneously digitizing analog-to-digital (A/D) data acquisition cards (ADLink 9810, 12 bits, 20 MHz, 4 channel) run in parallel. Continuous time series (for convenience partitioned into 300 0000 peak values $I(t_n)$ of the flashes for each channel) can thus be obtained. A second HP 33 120A function generator provides a triangular signal phase locked to the ultrasound drive. Suitably phase shifted, this unambiguously produces a voltage proportional to the timing of the pulse. This voltage can be recorded instead of one of the PMT signals by the A/D card, with the external timing signal for this produced by an ninth PMT coupled through a fast discriminator. Finally stroboscopic recordings are obtained using a digital camera together with a homemade long range microscope.

The timing of the flash relative to the phase of the sound field (denoted relative phase) has been shown in the stable case to be related monotonically to the size of the bubble

[11,12]. Since the time scale of the recycling is much longer than the cycle of the sound, it is fair to assume the bubble to be in a semistable equilibrium state, where a similar relation between size and relative phase holds. Therefore we have investigated the relationship between period doubling and relative phase (indicative of size) in the recycling modes and also the relationship to the surprising existence of period doubled states in the weak emission regime right after the sudden drop in light emission. 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 $m \geq 1$ a positive integer. For some of the observations we will for clarity in the displays make a running average. This has to be done separately for n even or odd.

When a recycling state is reached either by slowly increasing the forcing or by jolting the bubble with a sudden peak in sound amplitude, the recycling may take place for a very long time before the bubble either breaks or settles down again in a quiet state. Part of a recording of such a state is displayed in Fig. 1 where the intensity from one PMT (upper trace) shows the development in light emission in the type 1 recycling state for two near-extinction events. Degassing is done to 25% atmospheric pressure. However, the bubble suddenly settles into a second type of recycling state (type 2), the nature of which we shall discuss shortly. Just before the sudden drops in intensity a period doubling of the intensity is observed. The lower trace shows the corresponding development of the relative phase of the light pulses with time.

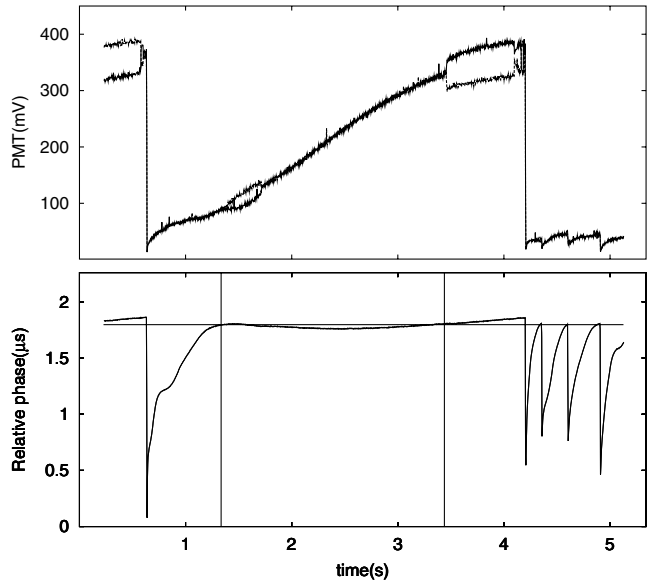

FIG. 1. Upper trace displays part of a recording of the light intensity with the bubble in the recycling mode. Running average over 30 peak values. Note the various period-doubling regimes. Simultaneous measurements from the other PMTs show the period doubling always to be geometric of nature. Lower trace shows the corresponding development of the relative phase. Vertical lines denote the timing of the onset of period doublings, while the horizontal line shows the associated relative phase. Degassing to 25%.

For the steady state sonoluminescing bubble we also find a completely stable period doubling. The intensity level here is just about the same as that of the recycling mode. However, the sound level needed is always somewhat lower than that needed to instigate the type 2 mode. Since the relative phase depends not only on the size of the bubble but also on sound amplitude, a direct comparison between the relative phases of the light pulses is not possible. However, information about the relationship is gained in the following manner. The ultrasound amplitude is set at the threshold value for stable period doubling. The level is then abruptly changed to that of the recycling mode. The time constant of the change of the sound level is given by the quality factor Q of the system which is of the order of ~ 100 corresponding to ~ 5 ms. Since this time is much shorter than the diffusion times involved, the phase corresponding to the collapse of a bubble of the right size for period doubling under stable conditions, can be measured at the pressure of the recycling mode by extrapolation of this exponential approach in phase. The conclusion from this kind of measurement is that the period doublings in the two cases take place at nearly exactly the same relative phases and that the states therefore are analogous in nature.

Surprisingly a small region of period doubling is seen shortly after the sudden drops in intensity at a much smaller light intensity. Both period doublings are observed to be anisotropic with the same 180° symmetry as observed for the steady state period doubling. The vertical lines in the lower part of Fig. 1 denotes the timing of the onset of the period doublings, while the horizontal line shows the relative phases associated with the two period doubled states.

Remarkably we again find a very close agreement between the relative phase of the timing of the period doubling in the two cases even though the pulse height is much reduced in the second case (from 350 to ~ 90 mV). This we interpret as showing that the phenomenon takes place at a certain bubble size and as a strong indication of the validity of the above assumption of semiequilibrium.

Conversely we can interpret the above observations as indicating that the relative phase gives a true measure of the size of the bubble also throughout a recycling mode (note though that the relative phase also depends on acoustic pressure). This aspect is of great interest in cases like the present one, where a direct measurement of the size, e.g., by Mie scattering is prevented by the preference of the measurement of other parameters.

Let us now turn to the new type 2 recycling case. In Fig. 2 we display an enlarged picture of the type 2 mode seen in the last part of Fig. 1. Altogether the recycling took place for 85 cycles before the bubble finally succumbed, although much longer tracks can be obtained. With only slight degassing, the type 2 mode can stay on for hours.

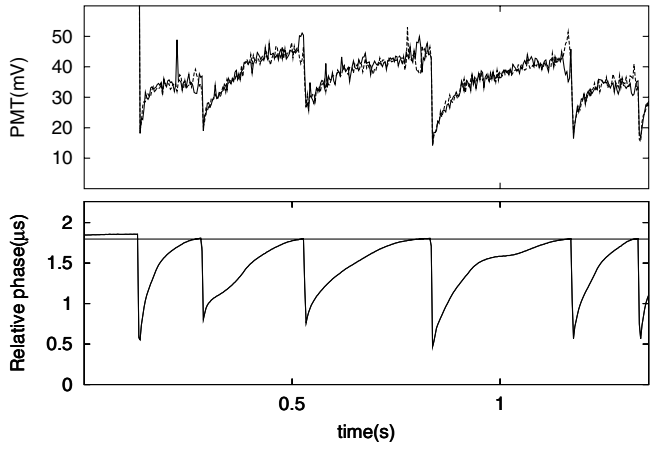

FIG. 2. Upper trace displays an expanded part of type 2 recycling (later half of the recording shown in Fig. 1). The trace starts with the drop from the end of a cycle of the type 1 recycling mode. Notice the period doubling. Lower trace shows the corresponding development of the relative phase of the light pulses with time with the horizontal line indicating the relative phase at the level of period doubling in the type 1 recycling mode.

The upper trace shows the variations in the light intensity seen by one PMT. The maximum light intensity varies quite a bit, but in all the cycles period doubling is observable. The variations are partly statistical in nature, but also a sign of the radiation from the bubble being anisotropic as can be ascertained from viewing the other channels. Now, however, the light intensity never reaches the level from before the drop, but stays at a much reduced level of around 45 mV.

The lower trace shows the simultaneous recording of the relative phase. Here we see an extraordinary consensus as regards the bubble size at the period doubling and the catastrophic events, the horizontal level drawn being that of the period doublings of the type 1 recycling mode and the stable mode corrected for the pressure dependence. As this indicates the same size for all the three cases the difference in light emission is most likely caused by a change in gas composition leading to a change in maximum temperature reached in the collapse.

To check this, we have attempted to measure the spectrum of the recycling modes. Since the lower light level of the type 2 mode compared with SBSL and the intermittent nature makes obtaining a spectrum with a spectrometer extremely difficult, we have resorted to making a crude eight point spectrum using narrow band filters (FWHM 10 nm) in front of the PMTs. As a reference we use the spectrum of a stable SBSL bubble. This has, in a previous measurement, been found to be well approximated by blackbody radiation at 14 000 K for ~25% degassing. In Fig. 3 we show the result of such a relative measurement for the spectrum of a type 2 event. Compared to the stably emitting bubble spectrum, the spectrum falls off drastically toward the ultraviolet. From the position of the peak a fit to blackbody radiation would give $T \sim 8000$ K but the spectrum falls off too fast toward both the ultraviolet and the infrared. Whether that feature is related to the 310 nm OH^* emission line is not known at present. A similar spectrum is obtained in the beginning of the cycle for a type 1 event although it looks slightly hotter, while at the end of the cycle the spectrum is close to that of the stable bubble. These spectra are also shown in Fig. 3. Note that the spectrum for type 2 looks rather similar to that of air-seeded MBSL. The spectrum represents an average over many different events but this is obviously also the case for MBSL. We have employed a homemade long distance microscope together with stroboscopic lighting to image the bubble with a CCD camera. Within the 10 μm resolution obtained there only seems to be a single bubble moving vertically a few tens of micrometers in the beginning of the type 2 cycle.

At a slight degassing level (~ 90%), the range of the sound pressure is sufficient to see the dependence on this as displayed in Fig. 4. Here the stable state spectrum is not known but the figure clearly shows the same qualitative behavior, with the bubble being much colder in the type 2 mode. Furthermore, we see the content of infrared growing as the sound pressure is raised. The sound pressure is not known at this degassing level. However, the drive amplitude is nearly constant throughout the entire range from ~25% to atmospheric pressure, where we observe the type 2 mode. This indicates that the sound pressure too is constant.

As noted above, the type 1 recycling is believed to be caused by the combined effect of pinch-off and diffusion. The bubble is mainly composed by argon before and right after the pinch-off. As indicated by the spectra the bubble is much colder after the collapse. The experiments described above therefore lead us to the following interpre-

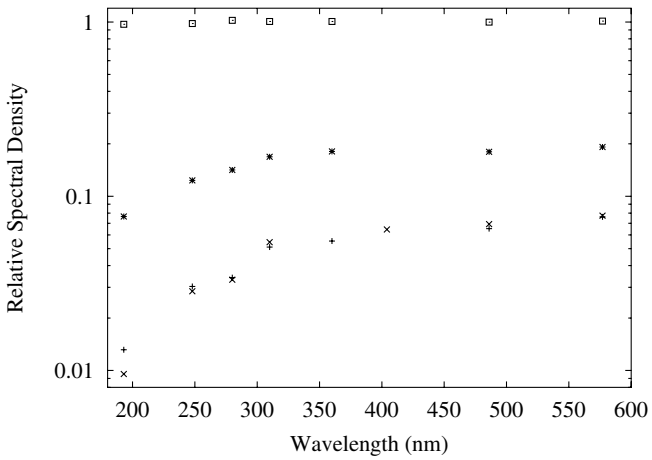

FIG. 3. Lower curve (× and +): Relative spectra of a bubble in type 2 mode (degassing to ~25%) normalized by the radiation from a stably emitting bubble. A switch of the 404 and 360 nm filters has been performed. Upper curves: Same for type 1, at, respectively, the low (*) and the high emission end (⊡).

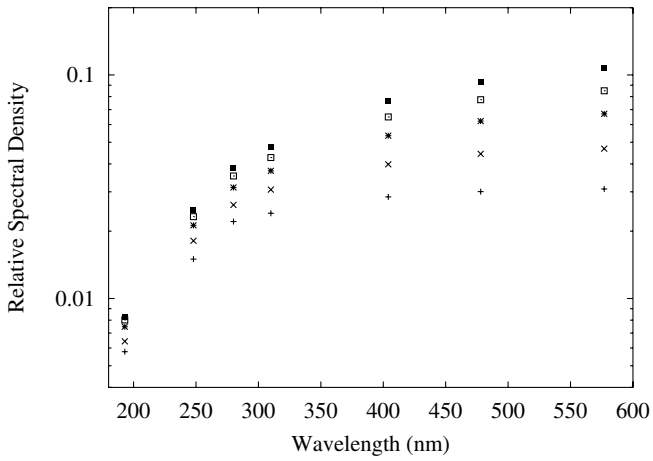

FIG. 4. Relative spectrum of a bubble in type 2 mode normalized by the radiation from a stably emitting bubble with drive amplitude as parameter. From below 137.6, 139.5, 140.8, 142.0, and 143.2 mV.

tation. After the collapse the bubble grows by diffusion toward the size called for by the applied sound pressure but is too cold for efficient burn off. The emission in this regime might in part be due to chemiluminescence since some of the possible reactions in fact produce light [13,14]. At some stage the increasing amount of argon in the bubble makes the bubble sufficiently hot that the nitrogen and oxygen starts to get burned off faster than these gasses diffuse into the bubble and the bubble starts to shrink as seen from the behavior of the relative phase (lowest trace, Fig. 1). As even more argon gets into the bubble this shrinking eventually stops and the bubble starts to grow again until large enough for the shape instability to set in and the cycle repeats. This interpretation agrees with the simulations of Holzfuss and Rüggeberg [15].

The essential difference between the two modes is in this picture that the type 2 starts out as an air bubble. From the spectrum we know that it is even colder than the type 1 so the burn off is even slower, the bubble grows faster by diffusion and enters the instability region before the temperature gets high enough to make it shrink by burning off the excess diatomic gas. Thus the breakdown again. The bubble now never has time to collect sufficient argon before the next breakup and therefore never leaves this state again as long as the sound field strength is not lowered. This explains why the bubble seemingly grows colder with increased sound pressure.

This view is strongly supported by looking at representative times for the recycling in the two modes. The time scale for a type 1 is of the order of 10 s while that of the type 2 is of the order of 0.1 s. Since the diffusion rate into the bubble is the determinant factor according to our picture, this difference in time scales agrees well with the amount of argon being only about 1% of the total amount of dissolved gasses.

To conclude, we have found a new recycling mode in air-seeded SBSL that, as indicated by the spectrum, represents a cold state of inefficient rectified diffusion thus having a high content of diatomic gasses. This picture is supported by considering the existence of period doubling and the difference in diffusion times for argon and the diatomic gasses caused by the strong difference in concentration. Measurement of the relative phase of the flash indicate that the onset of period doubling and shape instability depends mostly on size. This second mode of recycling may give valuable insight in the connection between multibubble and single-bubble sonoluminescence with this mode being a kind of SBSL equivalent of the MBSL state. It is worth noting here the 1 order of magnitude change in MBSL radiation going from argon to air-seeded bubbles found by Sehgal, Sutherland, and Verrall [14].

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