Multi-bubble sonoluminescence resolved in space, time, and colour

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ABSTRACT

A setup for space-, time- and colour-resolved sonoluminescence measurements is presented. By this technique, the acoustic cycle is split up into small time intervals to image differences in the regions and colours at different phases of the acoustic cycle. Time intervals of down to 100 ns can be exposed by a gated multichannel plate intensifier, whose images are then averaged by an EMCCD camera. Colour resolution is achieved by three filters of disjunct wavelength ranges in the visible, which is a compromise for having a full spectrum at every time at every position. As an example, multibubble sonoluminescence has been investigated at 420 kHz for pure water, water with SDS, and with luminol.

INTRODUCTION

Strong ultrasonic fields in liquids can cause cavitation and sonoluminescence, commonly known as multi-bubble sonoluminescence (MBSL) [Leighton (1994)]. In addition to the all-up intensity of the light emission from MBSL, there is also information hidden in its temporal and spatial distribution [Ohl et al. (1998), Otto et al. (2008)]. Ideally the whole optical spectrum could be resolved in time and space. To achieve this aim at least partially we use a set-up with a gated multi channel plate (MCP) which is triggered with a fixed phase relative to the acoustic driving. This phase coupling allows us to repeatedly expose only short time windows (down to 100 ns) per cycle. To collect sufficient MBSL light, these 100 ns windows are accumulated over many cycles. A delay line allows to shift the window position over the full acoustic driving period. Furthermore, three colour filters are used to achieve a rough spectral resolution. This arrangement can be used for various cavitation structures and different ultrasonic field parameters. The results show the regions and time periods of MBSL in the spectral filter ranges. One goal is to draw conclusions on the modalities and temperatures of bubble collapse in different spatial regions of the cavitating liquid.

MEASURMENT PRINCIPLE

For space- and time-resolved measurements a light sensitive EMCCD camera (IXON, Andor) and an intensified relay optics (IRO) are coupled. The working principle of the IRO is a multi channel plate where the photons are multiplied and maintain their spatial resolution. In addition, the IRO can be gated electrically, so that small exposure times of 100 ns and repeatabilities of up to 1 MHz can be achieved. The experimental setup is shown in fig. 1. In a light proof box the camera and the IRO are installed with an additional filter wheel, where several filters can be mounted. In front of the filter wheel, the cuvette is positioned which should be inspected. An additional halogen lamp is in the box for scattered light photographs for calibration. All of the equipment can be controlled from the outside of the box.

For phase resolution, a trigger to a the periodic (driving) oscillation is needed. For that we use the driving sine. This makes sense if we assume that the bubbles oscillate coupled to the
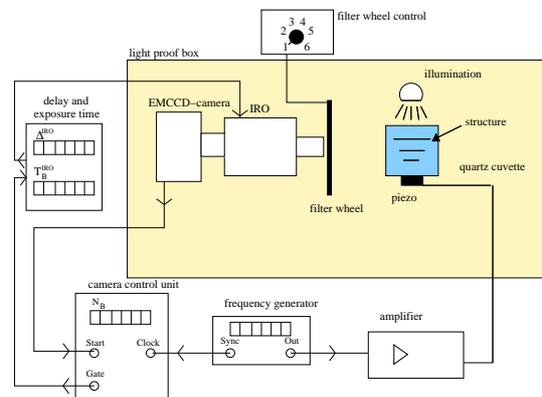
Figure 1: Scheme of the setup (see text).

driving and perform the same oscillation in every cycle. Although this is not true for every single bubble (as the bubbles appear, move, split, merge or disappear), this assumption holds for the time averaged cavitating bubble field, if it is sufficiently stable over the observation time. The camera is opened for several seconds, and thus integrates over a large amount of cycles. As the IRO is triggered by the driving sine, from every cycle only a small time slot of 100 ns is recorded, always at the same driving phase. If the delay time is increased in steps of 100 ns, a whole acoustic cycle can be covered.

DATA ACQUISITION AND PROCESSING

For three different liquids the sonoluminescence was recorded. The liquids (always 50 ml) were filled in a cuboid quartz glass cuvette (feed size of 4 cm) with a circular plate piezo ceramic (thickness 2 mm, diameter 45 mm) glued to the bottom. All liquids were saturated with argon by means of bubbling 30 min argon through it. The driving frequency was for all liquids 420 kHz, that is to say for a time slot of 100 ns 24 frames are needed to cover one acoustic cycle (acoustic cycle time: 2.38 μs). All measurements were performed in the same way. For every colour filter a sequence of 24 frames was recorded with increasing delay times for the increasing frame number. That means frame one corresponds to delay time 0 ns till 100 ns, frame 2 to 100 ns till 200 ns, and so on. The three raw se-
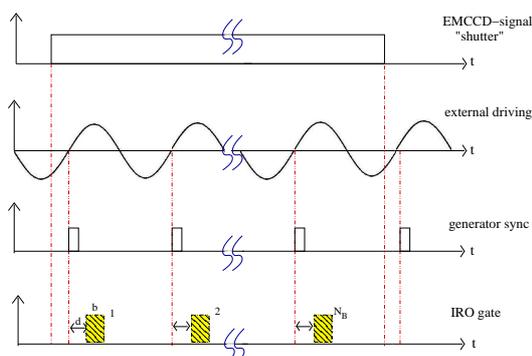
Figure 2: Principle for the time resolved measurements. As soon as the camera shutter opens, all positive zero crossings of the driving sine are converted to a rectangular pulse which triggers the IRO. In addition, the IRO has an internal delay time and an exposure time which can be controlled. If this delay time is increased in several steps, a whole acoustic cycle can be scanned.

quences are edited by contrast enhancement, colour inversion and then colorizing with the filter colour. The resulting three movies are overlaid frame-wise to one movie. White colour means no detected light, and red, blue or green parts mean light detection with the corresponding filter. The used filter transmission ranges are red ($\lambda > 600$ nm), green(500 nm $< \lambda < 575$ nm), and blue ($\lambda < 500$ nm) in the visible.

RESULTS

The first liquid is filtered and deionized (DI) water. The result is shown in fig. 3. It can be seen that there are time slots where no light can be detected (frames 7, 8, 9 and 19, 20) and there are parts where a maximum of light can be seen (frame 2 and 14). So the light intensity increases and decreases twice per cycle, which is due to the standing wave in the cuvette: In the first half of the acoustic cycle the bubbles in every second antinode are around collapse phase, which is where light is emitted. All the bubbles in the other antinodes are at growing phase and dark. In the second part of the cycle all the bubbles in the antinodes which where emitting light in the first part are now in the growing phase, and the ones which were in the growing phase in the first part are now in the collapse phase and therefore emit light. Thus in every half period only half of the antinodes are emitting light. An average in time over many cycles would lead to light planes of half a wavelength, i.e. the doubled amount of the planes of one frame of the movie.

DI-water

The first and the third as well as the second and the fourth row in the figure resemble each other. In the first row a decay of intensity can be seen, as well in the third. In the second row only at the fourth frame the light intensity starts to increase, just as well in the last row. So there is the same light emitting process which is repeated for the antinodes.

On top of the distinct standing wave field in the vertical direction there is a weak propagating wave in the horizontal direction from the right to the left overlaid. In the first frame the light pillar is tilted over to the right, in the second it is straight and in the following frames it tilts over more and more to the left (the same counts for frame 13 and following).

For the different colours arises nearly no space dependency. Everywhere where light can be detected all three colours are present. There is only a slight dependence on time. From this movie is seems as if the light emission would start in the green
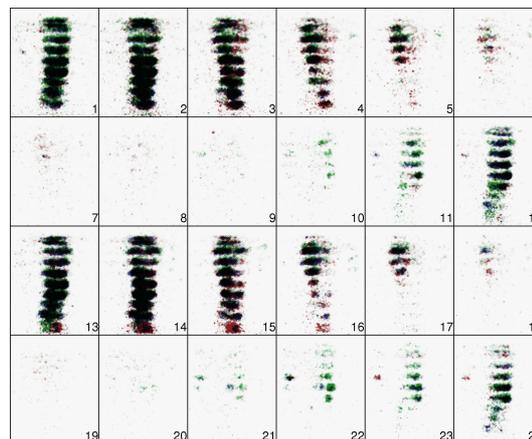
Figure 3: Phase- space- and colour-resolved measurement of argon saturated DI water at 420 kHz. For each frame the exposure time was 7 seconds. The width of the image is 3.1 cm.

(frames 10, 11 and 20, 21, 22) and end in the red wavelengths (frames 4, 5, 6, 7 and 16, 17, 18). That would indicate that small bubbles, which collapse earlier than bigger bubbles, cause a hotter emission. One explanation might be that the larger bubbles develop asphericities (due to their size, relative motion, or neighbouring bubbles) which results in a lower compression rate of their interior. Alternatively or additionally, larger bubbles might have larger vapour content, which reduces their collapse temperature.

In conclusion, from standard (DI) water no spatial dependence of sonoluminescence can be measured. However, Sunartio et al [Sunartio et al. (2007)] observed in SDS and NaCl solutions spatial differences of colours with a conventional digital mirror reflex camera. Therefore, we decided to study other liquids than pure water.

SDS solution

SDS is a surfactant which arranges itself to the surface of the bubbles so that the hydrophobic parts sticks to the bubbles [Ashokkumar et al. (2007)]. In addition the bubbles get loaded due to the end chain which points into the water and so it gets harder for the bubbles to coalesce and the bubble structures decluster. Over and above, the surface tension is lowered. In high speed movies it could be seen that big cigar shaped bubbles evolve.
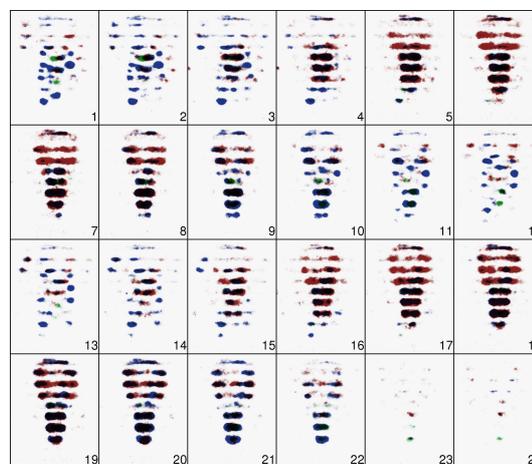
Figure 4: Phase- space- and colour-resolved measurement of argon saturated 1 mM SDS solution at 420 kHz. The image width is 2.7 cm.

Compared to water, a different light emission can be seen in an argon saturated 1 mM SDS solution (see fig.4). It is possible to detect light in every phase. But there is again a similarity between the first and the third as well as in the second and the last row. In row one and three the first three frames are dominated by the blue light and the last three frames are dominated by the red light. All the time when the red light gets dominant the light structure narrows conical. When the blue light is dominant the light structure looks like a lined block where in every distance to the transducer the horizontal lines have the same length. But when the red light gets dominant the light structure looks like a triangle with the pit close to the transducer. Overall there is again a wave motion of the light intensity visible. But this time it is not a change from no light to much light collectively, but a change from nearly no red light to much red light. It is again not possible to see parts where only one colour is present. Here the green light is very dim, so maybe the light is more divided into red and blue light, what would indicate, that there are no big bubbles which collapse very heavily.

If we compare the SDS emission with the water emission, the strong light emission in the water for all colours corresponds to the blue dominance in the SDS solution, and the missing of light emission in water corresponds to the red dominance in SDS. So maybe the red dominant parts are the Na* emissions from the SDS, and the blue dominant parts are from the normal bubble collapse. A final interpretation, however, still has to be found.

SDS-luminol solution

Again a different light emission pattern can be seen from a 1 mM SDS- 0.2 mM luminol solution. Luminol causes sonochemiluminescence [McMurray and Wilson (1999)]. That is to say, the light which can be detected from luminol solutions is not completely due to the collapsing bubbles and the light emission from them, but an additional chemically induced light emission occurs. The ground state luminol molecule needs to react first with an OH radical and then an $O_2^{(-)}$. The next step is the release of an $H^{(+)}$. The last step is the release of an N_2 which yields to an excited state of the molecule where a photon is emitted when it relaxes back to the ground state. This photon has a wavelength of 430 nm.
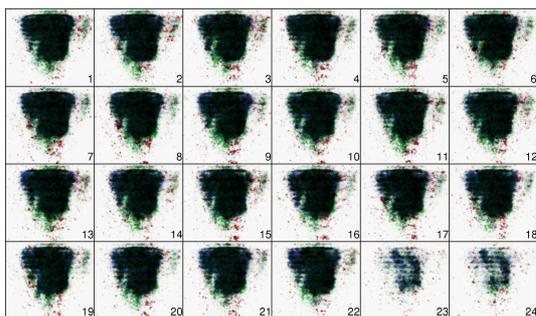
Figure 5: Phase- space- and colour-resolved measurement of argon saturated 1 mM SDS and 0.2 mM luminol solution at 420 kHz. The width of the image is 3.1 cm.

Our results are shown in fig.5. A dominant light emission in the blue range can be seen in every frame. This time the red light is very dim. No strong intensity changes appear. That means all the time, even when no bubble should collapse, there is still light emission. This is apparently due to the time the luminol molecule needs to hit the right reaction partner and to be excited and then finally to fall back to the ground state including light emission. The time for all the steps together is in the ms range. That means that the time for the afterglow is long enough to cover one or even 30 cycles. Nevertheless, the

diffusion/convection of the excited luminol molecules is too slow to get into the regions where the nodes are. This can be best seen in the last two frames. In the blue colour it is still possible to see the horizontal lines. If the resolution would be better and the exposure time would be shorter the individual lines could be seen much better. At lower frequencies it could be very nicely seen that the nodes do not emit light when luminol is added, even if the bubble streaming from node to node was 0.5 m/s.

CONCLUSIONS AND OUTLOOK

Phase- space- and colour-resolved sonoluminescnece measurements were done for three different liquids. Until now no distinctive spatial colour distribution could be seen. If light could be detected, all three colour ranges were present. To improve this, new filters will be used with a better differentiation between the different colours. The time dependency is only very small. For water, a beginning in the green and an end in the red colour could be seen which suggests that the smaller bubbles, which emit light first, have a stronger collapse (in the sense of temperature), and the emission of the larger bubbles is less hot (quenched by aspherical collapse and/or higher vapour content). For SDS solutions, a double intensity cycle can be seen: one for the red part and one for the blue, which are in maximum intensity against each other. A final explanation for these findings still has to be found. For the luminol, no time dependency is measurable. In all phases there is a strong blue light emission, apparently due to the long reaction time of the luminol molecule.

Next steps will be to improve filter numbers and also to include point-wise measurements of the whole spectrum to get a better overview of the involved wavelengths and the corresponding intensities. Further acoustic geometries and other liquids will be investigated for spatio-temporal patterns of sonoluminescence.

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