

## ACTION OF ULTRASOUND ON PARTICLES AND CAVITATION BUBBLES

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Action of ultrasound on suspending particles and on cavitation bubbles is discussed from a viewpoint of sonochemistry. As examples of acoustic radiation pressure on solid particles, aggregation and manipulation of a small object are shown. Acoustic radiation pressure on bubbles, or Bjerknes force, acts in a different manner because of large compressibility of a bubble. Observation of sonoluminescence from oscillating bubbles reveals that the action of the primary Bjerknes force and the secondary Bjerknes force is crucially important for the efficacy of sonochemistry.

**Introduction**

Particles placed in a sound field receive action of acoustic radiation pressure. This phenomenon is used for aggregation, separation, and non-contact manipulation of particles as well as visualization of the sound field. High-intensity ultrasound in water induces cavitation and generates many tiny bubbles. Acoustic radiation pressure also acts on bubbles and it is called Bjerknes force. Since compressibility of a bubble is large, the bubble oscillates in accordance with ultrasound and the action is different from that on solid particles. Ultimately, the oscillation leads to collapse of the bubble attaining extremely high temperature and pressure in a very short time. These extreme conditions make the basis of sonochemistry. The present paper discusses the action of ultrasound on acoustically induced bubbles in contrast with that on solid particles.

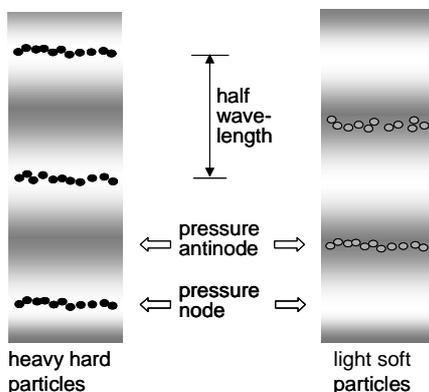


Figure 1: Action of acoustic radiation pressure on solid particles in a standing-wave field.

**Action of radiation pressure on solid particles**

It is well known that particles placed in a sound field receive action of acoustic radiation pressure. In a standing-wave field, the direction of radiation force, which is obtained by integrating the radiation pressure around the particle surface, is different depending on the density and compressibility of the particle [1]. Heavy and hard particles, whose size is sufficiently smaller than the acoustic wavelength, receive force toward nodes of sound pressure distribution, whereas light and soft particles go to antinodes of sound pressure (Fig.1).

This phenomenon is used for aggregation, separation, and non-contact manipulation of particles as well as visualization of the sound field. Figure 2 shows aggregation of polystyrene beads in a standing-wave field generated in saline water of the same density as that of beads [2]. Slight change in ultrasonic frequency alters the aggregation pattern, which corresponds to the distribution of sound pressure.

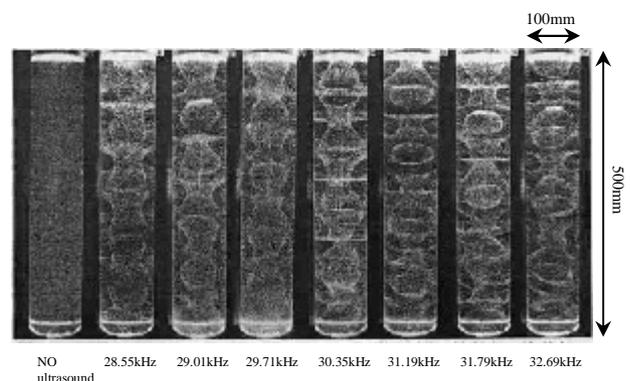


Figure 2: Aggregation of polystyrene beads in a standing-wave field of saline water at various ultrasonic frequencies.

Figure 3 shows an example of non-contact manipulation of a polystyrene bead in a standing-wave field generated by three crossing sound beams of 1.75 MHz [3]. The sound field was calculated by superposing three sound beams emitted from each transducer of 11 mm in diameter and a honeycomb-like standing-wave field is seen in the center of the figure. This pattern was also confirmed through visualization of the sound field by the schlieren method. Since the standing-wave field can be shifted by changing the mutual phases of driving signals, a particle trapped at the pressure node of the standing wave field moves two-dimensionally along with the

shift of the sound field. The inserted photograph shows an experimental result of acoustic manipulation of a particle in this sound field.

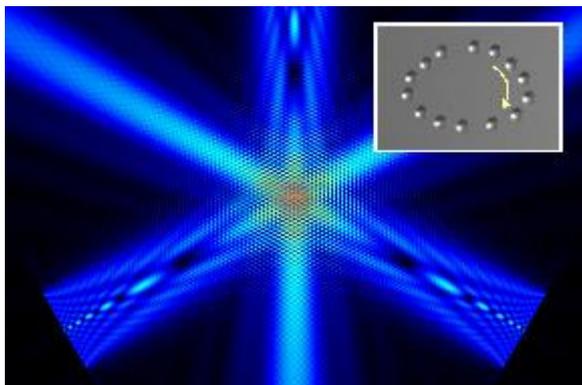


Figure 3: Non-contact ultrasonic manipulation of a particle in a standing-wave field generated by three crossing sound beams of 1.75 MHz in water.

**Action of radiation pressure on bubbles**

Acoustic radiation pressure also acts on bubbles and is called Bjerknes force. This force dominates behavior of bubbles. Although Fig.1 suggests that “light and soft” bubbles go to antinodes of sound pressure in a standing-wave field, there exists bubble-size dependence because of large compressibility (Fig.4) [1]. If a bubble oscillates sinusoidally and its radius is smaller than the resonant radius corresponding to the driving ultrasonic frequency, it goes to the antinodes of sound pressure distribution as in the case of Fig.1. On the contrary, if the bubble is larger than the resonant radius, the bubble cannot oscillate in phase with the pressure change of the ultrasound. Then, it goes to nodes of sound pressure distribution where the bubble cannot oscillate because of the least amplitude in sound pressure.

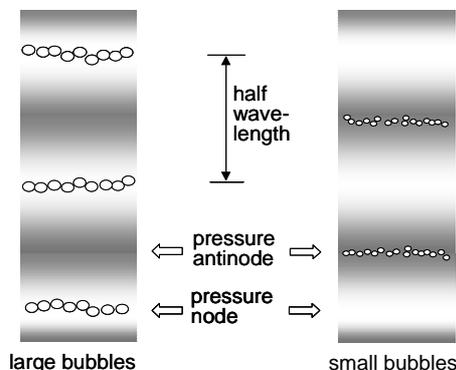


Figure 4: Action of acoustic radiation pressure on bubbles in a standing-wave field.

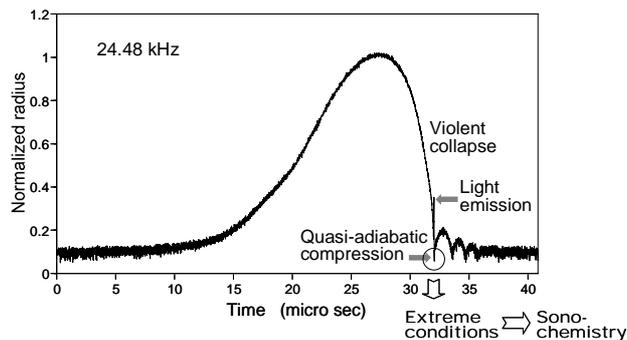


Figure 5: Nonlinear oscillation of a bubble measured by the light-scattering method.

Since compressibility of a bubble is large, the bubble oscillates in accordance with ultrasound. But the oscillation is not sinusoidal except under very low sound pressure levels. Generally, bubbles make nonlinear oscillation as shown in Fig.5 [4]. This figure shows a radius versus time curve of an oscillating bubble measured by the light-scattering method. At the compression phase, the bubble collapse with large compression ratio in the order of  $1:10^{-6}$  and quasi-adiabatic compression generates extreme conditions in the bubble. The temperature in the bubble attains several thousands kelvins and the bubble emits light called sonoluminescence. Analyses of sonoluminescence revealed that small bubbles in the order of several micrometers are responsible for sonochemistry and they go to pressure antinodes. Hence, effective sonochemical reaction fields correspond to the antinodes of sound pressure distribution.

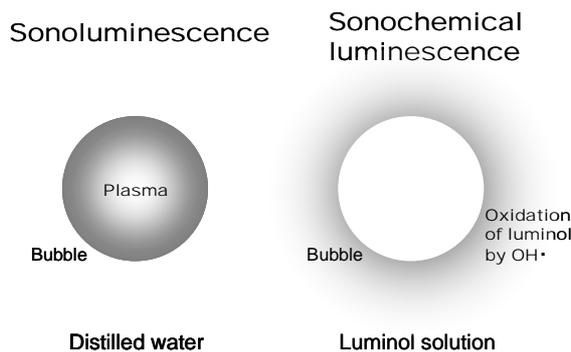


Figure 6: Comparison of luminescing regions between sonoluminescence in distilled water and sonochemical luminescence in a luminol solution.

**Sonoluminescence at high sound pressure levels**

The mechanism of light emission is different depending on the medium. If the medium is distilled water, the light is generated from accelerated motion

of electrons of plasma in the bubble. This light emission is sonoluminescence from inside of the bubble. On the other hand, if the medium is a luminol solution, the light is due to the oxidation of luminol by OH radicals generated in the bubble by decomposition of water vapor and immersed into liquid surrounding the bubble. This case is sonochemical luminescence and the light comes from the shell of the bubble (Fig.6). Anyway, acoustically induced luminescence comes from the neighborhood of bubbles.

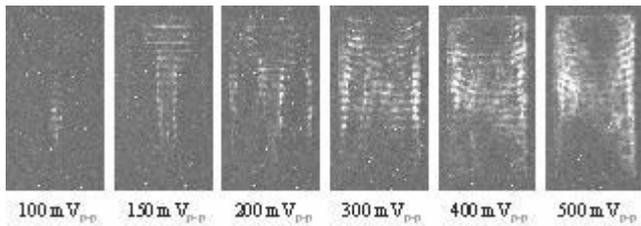


Figure 7: Changes of luminescing regions in a standing-wave field as the sound pressure level increases. The label of each figure indicates output voltage of the function generator (FG) or input voltage to the power amplifier. Rectangular glass cell, luminol solution, 140 kHz.

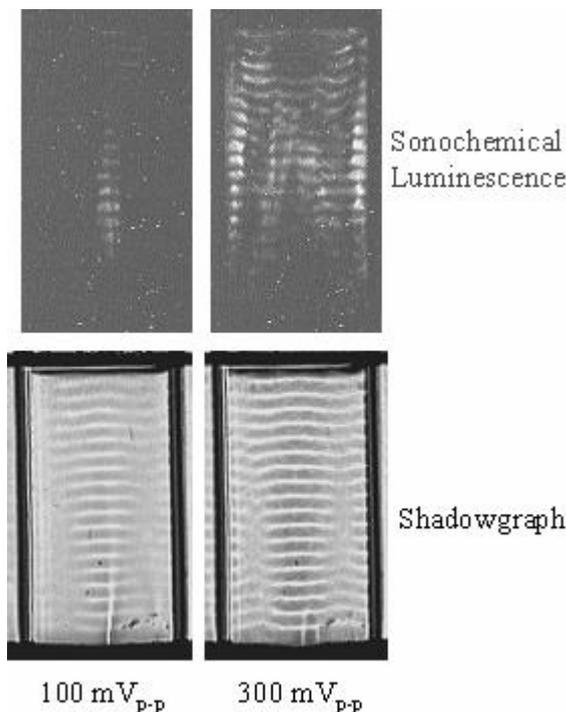


Figure 8: Comparison of luminescing region (upper) and visualized sound field (lower) at different sound pressure levels. 140 kHz.

Figure 7 shows changes of sonochemical luminescence of a luminol solution in a rectangular glass cell driven at 140-kHz ultrasound [5]. The indicated

voltage means input voltage to a power amplifier. As the input power increases, the luminescing region becomes larger, but it is interesting to note that the region changes depending on the input power. Figure 8 compares the sound field visualized by the shadowgraph method with the luminescing field at two input levels [5]. The distributions of the sound field do not change except their amplitude, but the luminescing region at the lower input level disappears at higher input level; namely, quenching of sonoluminescence occurs at high input levels.

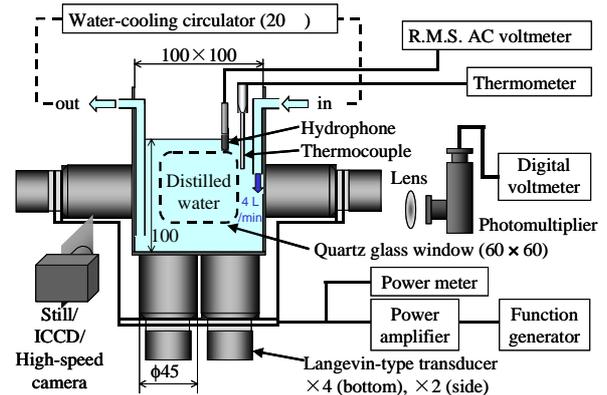


Figure 9: Experimental setup to observe bubble behavior at various sound pressure levels.

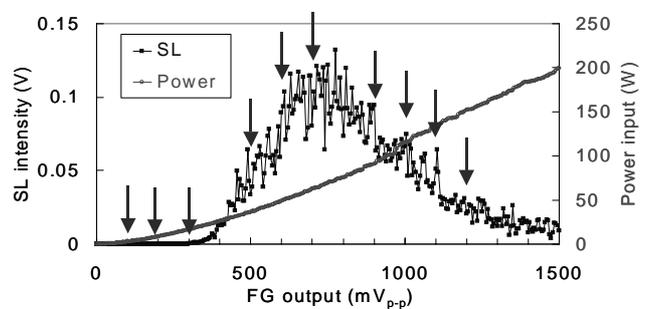


Figure 10: Changes of input power to the ultrasonic transducers and the corresponding sonoluminescence (SL) intensity at 23 kHz.

Using experimental apparatus shown in Fig.9, changes of sonoluminescence intensity and input power were compared at lower frequency of 23 kHz so that observation of bubble behavior became easier [6]. The results are shown in Fig.10. As the output level of the function generator (FG) increases, input power to the ultrasonic transducers increases. But sonoluminescence (SL) intensity becomes lower beyond a certain input level. Figure 11 shows the bubble behavior taken with a still camera. The center of each photograph is the antinode of sound pressure

distribution in the cell. The arrows in Fig 10 correspond to the photographs in Fig.11. It is seen that tiny cavitation bubbles gather to the antinode of sound pressure at lower sound pressure levels. But, at higher levels where SL intensity decreases, the bubbles are expelled from the pressure antinode.

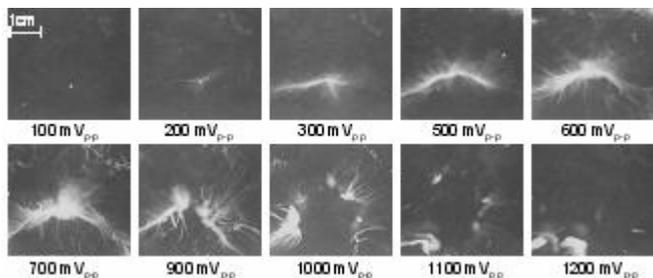


Figure 11: Changes of cavitation behavior at various FG output voltages indicated by arrows in Fig.10. Each photograph was taken with a still camera and the center part corresponds to the antinode of sound pressure distribution in the cell.

**Cluster formation by the secondary Bjerknes force**

These phenomena can be explained by cluster formation of bubbles (Fig.12) [7]. Bjerknes force mentioned in the previous sections is the primary Bjerknes force and determines macroscopic motion of bubbles whether they go to antinodes or nodes. Since the bubbles undergo oscillation in a sound field, re-radiation of ultrasound from oscillating bubbles induces the secondary Bjerknes force. If the bubbles oscillate in phase, attractive force is generated between the bubbles, which forms a cluster of bubbles. Once these clusters are formed, tiny bubbles become large and they are expelled from the pressure antinodes because they grow too large to stay there under the action of the primary Bjerknes force. The bubble cluster also shields the immersion of ultrasound into the inner part of the cluster because of difference in acoustic impedance.

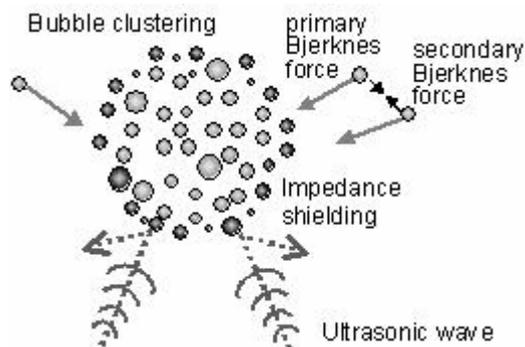


Figure 12: Schematic diagram of cluster formation of cavitation bubbles by the secondary Bjerknes force.

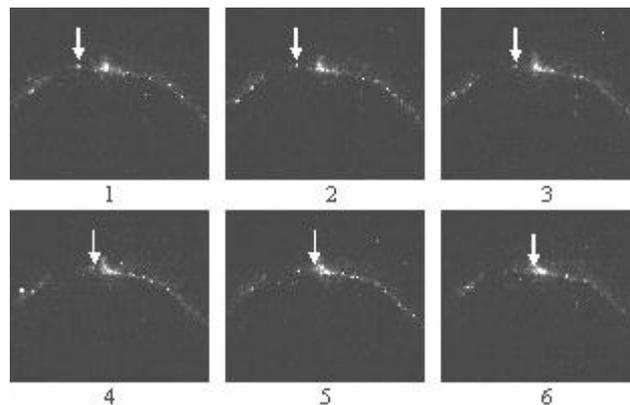


Figure 13: High-speed video images of cavitation bubbles taken at 1000 frames/s. 600 mV and 23 kHz corresponding to Figs.10 and 11.

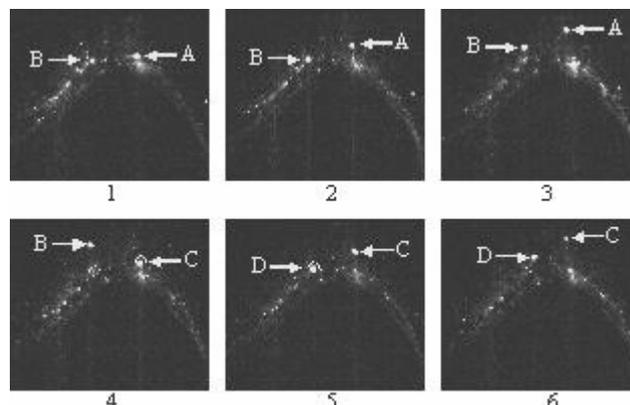


Figure 14: High-speed video images of cavitation bubbles taken at 1000 frames/s. 900 mV and 23 kHz corresponding to Figs.10 and 11.

Figures 13 and 14 show images of bubble behavior recorded with a high-speed video camera taken at 1000 frames/s in the same experiment as Figs. 9-11 [6]. Figure 13 is the case at lower input level and tiny bubbles gathers around the antinode of sound pressure. On the other hand, in the case of higher input level as shown in Fig.14, tiny bubbles form clusters and they are expelled from the antinode at the center of each photograph.

Since, roughly speaking, the secondary Bjerknes force is inversely proportional to the second power of distance between bubbles and the forth power of ultrasonic frequency, it is weak generally. So clustering of bubbles may be affected by fluid motion in a sound field (Fig.15). This speculation was confirmed by working the pump of water-cooling circulator in Fig.9 [7,8].

**Conclusions**

The action of radiation pressure on bubbles is very different from that on solid particles, because

compressibility of a bubble is very large. Although irradiation of intense ultrasound generates cavitation bubbles in a liquid, only tiny bubbles are responsible for sonochemical reactions, because they stay at antinodes of sound pressure distribution in a standing-wave field and their violent collapse generates extreme conditions. The primary Bjerknes force determines macroscopic motion of bubbles whether they go to antinodes or nodes. The secondary Bjerknes force dominates clustering of bubbles, which is influential at high input power levels and crucially important in the efficacy of sonochemistry.

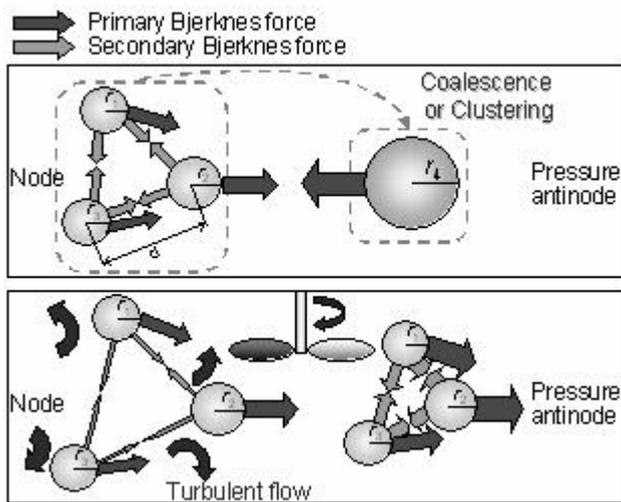


Figure 15: Influence of fluid motion on cluster formation of cavitation bubbles.

### Acknowledgments

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