Comment on "Mie scattering from a sonoluminescing bubble with high spatial and temporal resolution" [Physical Review E 61, 5253 (2000)]

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A key parameter underlying the existence of sonoluminescence (or SL) is the time dependence of the radius $R(t)$ of the collapsing bubble from which SL originates. With regard to the use of light scattering to measure this quantity, we wish to note that we disagree with the statement of Gompf and Pecha—highly compressed water causes the minimum in scattered light to occur 700 ps before SL—and that this effect leads to an overestimate of the bubble wall velocity. We discuss potential artifacts in their experimental arrangement and reply to their criticisms of our experiments on Mie scattering.

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Sonoluminescence (SL) occurs when the energy density of the contents of a bubble are rapidly concentrated by its implosion [1]. The implosion dynamics are an important aspect of SL and so various techniques [2–10] have been applied to the experimental determination of the bubble radius as a function of time $R(t)$, and the response of the water. Various realizations of Mie scattering [3–10] in particular have proved useful in obtaining bubble parameters. Mie scattering occurs when variations in the index of refraction cause light to be scattered out of the direction of the incident beam.

In a recent paper [11] Gompf and Pecha (GP) have used a streak camera to image Mie scattering. We wish to reply to statements of GP that are critical of our work, and disagree with our published results.

In the Abstract GP claim that "In the last nanoseconds around minimum bubble radius most of the light is scattered at the highly compressed water surrounding the bubble and not at the bubble wall. This leads to a minimum in the scattered light intensity about 700 ps before the SL pulse is emitted." They go on to say that "neglecting this change leads to a strong overestimation of the bubble wall velocity."

We disagree with a number of aspects of these statements. The 700-ps interval that GP quote is specific to their particular experimental arrangement and is unrelated to the physics of a bubble collapsing in highly compressed water. In Fig. 1 of Ref. [8] the flash will be seen to occur 100–200 ps *before* (not 700 ps *after*) the minimum apparent radius (i.e., y axis), which for this experiment is the minimum in total light scattering. We agree [8] that light scattering is due to index of refraction changes at the wall of the bubble as well as the highly compressed water. But in this case, attribution of the Mie scattering exclusively to the bubble wall would lead to an *underestimate* of its velocity, not an *overestimate*, as quoted above from GP. Perhaps the observation that the flash *precedes* the minimum in light scattering could be due to this effect.

On page 5254 GP discuss our previous experiments on Mie scattering [3,4,5] and state that "In...former investigations...the scattered light intensity was assumed to be proportional to the square of the bubble radius that totally neglects the complicated angular distribution of the Mie scattering." This statement comprises an inaccurate description of past experiments. The complicated angular distribution can be seen in Fig. 6 of [4], which was taken from our first paper on Mie scattering from SL [3]. One of the steps that enabled us to obtain quantitative information about bubble radii from Mie scattering was to simplify the scattered intensity as a function of R by collecting light from a large solid angle: such as 30°–80° [8] or 46°–94° [3]. In this case the intensity of light scattering is within 20% of R^2 for bubbles bigger than 0.6 μm [8], or 1 μm [3]. These corrections and their connection to the "complicated angular distribution of Mie scattering" were discussed in these papers. A plot typical of calculations that formed the basis for these corrections was published in Ref. [8]. The strong deviations from R^2 Mie scattering displayed in Fig. 5 of GP results from their collecting light in a small solid angle [14°–36°] near the forward direction. On page 5255 GP state that our papers neglected the effect of changes in the refractive indices due to the implosion. This is true; the index of refraction inside the bubble was reckoned to unity for the purpose of deconvolving the scattered intensity. Light scattering techniques have not yet reached the point where changes in the index of refraction, due to say the formation of a plasma, can be extracted. Analysis of our data also neglected the effect of bubble asphericity (see discussion relating to Fig. 6 of Ref. [5]).

The time scale of 700 ps enters GP in two entirely different contexts: (1) it is a "pronounced minimum in the scattered light intensity 0.7 ns before the SL pulse due to Mie lobe clusters" and (2) "from this time on most of the light is scattered at the highly compressed water around the bubble leading to a strong increase in the scattered light intensity before minimum bubble radius" which is the moment of SL. For the choice of angles over which GP collect scattered light we agree with (1) but emphasize that a different choice of angles eliminated this artifactual minimum. Regarding (2) we reiterate our disagreement with GP's Abstract.

At more than one location in GP it is claimed that "the bubble wall velocity 1 ns before the SL pulse is about 950 m/s. This value is much lower than the values found by Weninger, Barber, and Putterman'' (Ref. [5] this comment). First, the bubble wall velocity 1 ns before collapse, where R is about 1.7 μm, is for our data 900 m/s [5], which is in good agreement with the results of GP. Second, the 500-ps timing resolution of GP means that a 500-ps smoothing function has been applied to their rapidly changing data. There can be no question that their value of 950 m/s is an underestimate of the bubble wall velocity in their experiment. Furthermore, a statement claiming a significant discrepancy between experiments would have more weight if it were accompanied by a discussion of ''error bars.'' The paper of GP contains no such discussion. An example of experimental ''error'' is shown in Fig. 6 of [5]. It has sources in the various processes discussed above, gas concentration, and also run-to-run variations.

The data of GP for $R(t)$ cut off at about 1.7 μm up to which point they are largely in agreement with our previously published results [4–6]. There remains the issue of whether the bubble wall velocity for some systems [4–6] approaches higher velocities (e.g., in excess of 1200 m/s) for smaller radii. Systems are characterized by the gas mixture used, acoustic frequency, and ambient temperature. (For 1% Xe 99% O_2 dissolved at 150 Torr driven at 40 KHz at 20 C, the maximum velocity was actually found to be less than 950 m/s [8].) In the range 1.7 μm$>R>R_c\approx 0.5\,\mu$m, where R_c is the collapse or minimum radius, GP provide no data for R. They claim that this is due to the difficulty in subtracting out a large signal due to scattering from highly compressed water in a 5 ns gap around R_c. Except for a much smaller window ($\sim$200 ps) around R_c we disagree. We suggest some possible complications that may affect their experiment in this range.

(1) The GP choice of angles leads to Mie lobes that are sufficiently complicated that the intensity of scattered light is not monotonic with radius, so that deconvolution is difficult.

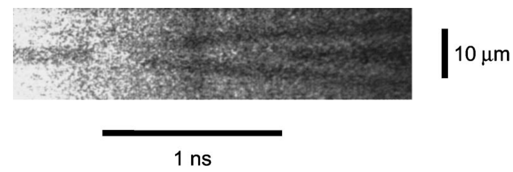

FIG. 1. Single shot streak camera shadowgraph of a collapsing bubble launching a pulse of sound into the surrounding water. The image of the bubble is the center line, and the radiated pulse of sound moves at a supersonic velocity relative to the speed of sound in water. As this particular bubble is not centered on the entrance slit its image is lost during the indicated 1-ns time span. The experimental details and a photo lacking this artifact can be found in [8].

(2) As the GP images are magnified and averaged, small translational motion and concentric pulsation of the bubble can throw its image off the slit [12].

(3) The level of scattered laser light is less than the intensity of SL, obscuring dynamics near R_c. In Fig. 1 one sees a very common example of the image of an imperfectly centered bubble falling off the slit during that portion of the cycle that surrounds the minimum by about 1 ns. Improved measurements of $R(t)$ should be carried out on a shot-by-shot basis [8]. Signal relative to noise could then be increased by averaging together only those photos that do not display the artifacts discussed above. We also suggest that the index of refraction inside the bubble be measured with an optical probe (such as Thomson scattering) that is sensitive to ionization.

It is good news that the action of an audible sound field on water has led to a debate about experimental techniques on the scale of 100–700 ps. A determination of $R(t)$ will help resolve the existence of shock waves or other energy focusing effects inside the bubble [13–15].

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