Predictions for Upscaling Sonoluminescence

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Only a small fraction of the high-dimensional parameter space that governs the occurrence of stable single-bubble sonoluminescence (SBSL) has been explored so far. We predict that decreasing the acoustic driving frequency $f$ upscales SBSL. More specifically, at $f = 5$ kHz we expect more than 100 times as many photons per flash as at $f = 20$ kHz and a flash width of about 1000 ps. The application of lower frequencies has to be assisted by reducing the partial inert gas pressure of the dissolved gas (e.g., stronger degassing) to maintain diffusive stability of the bubbles.

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The astonishing energy focusing capabilities of a bubble in single-bubble sonoluminescence (SBSL) [1,2] have intrigued many researchers and triggered numerous experimental [2,3] and theoretical [4] studies aimed at determining the central temperatures and emitted light intensity of SBSL bubbles. Most experiments and theories indicate that the temperatures (or equivalent energy densities) inside the bubble can reach at least several $10^4$ K and the short (80–300 ps [5,6]) light pulses consist of up to $10^7$ photons in the visible range [2]. The question now arises if the experimental parameters can be altered such that these figures can be pushed to even more extreme values, i.e., can SBSL be upscaled?

Experimentally [2] it was found that an efficient way to achieve more intense light is to cool the water. Decreasing the water temperature from 33 to 2.5 °C gives nearly 1000 times more photons per pulse. Following the hydrodynamical/chemical approach to SBSL [7–10], the water temperature dependence was quantitatively accounted for by considering the temperature dependence of the material constants of water [11].

In this paper we focus on the upsaling of SBSL by reducing the driving frequency $f$, a procedure suggested by Apfel [12]. The effect of reducing the frequency is twofold: (i) In the SL regime the dynamics of the bubble radius $R(t)$ is characterized by a long and relatively slow expansion that occurs during the negative pressure phase of the driving, and by a subsequent violent collapse. Because a smaller frequency allows the bubble more time to expand, it leads to a larger expansion ratio $R_{max}/R_0$ (maximum radius divided by ambient radius) and a stronger collapse. Indeed, the example of Fig. 1 shows that, in comparison to frequencies commonly used today in experiment (e.g., $f = 20–40$ kHz), more violent collapses can be reached (at fixed $R_0$) with smaller $f$. (ii) For smaller $f$ the threshold of shape instability which limits the SBSL regime [8] is shifted towards larger bubbles which potentially emit more sonoluminescence light. Indeed, experiments with smaller $f$ by Barber and Putterman [13] and by Cordry [14] found brighter SL bubbles. But what is the maximum light intensity which can be expected, and how should one choose the forcing pressure $P_d$ and the gas concentration in the liquid to achieve an optimal photon yield? The present study will make quantitative predictions for answers to these questions.

Our analysis of upscaled SBSL is based on the hydrodynamical approach to SBSL [7–10], augmented by an approximate description of the thermal bremsstrahlung of the partially ionized gas inside the bubble [15,16].

The bubble is driven by the harmonic driving $P(t) = -P_d \cos 2 \pi ft$; it responds according to the Rayleigh-Plesset (RP) dynamics [2,8,17,18]. The pressure $p(R(t))$ inside the bubble is approximated by a spatially homogeneous van der Waals pressure. This approximation is well...
justified for inert gases \cite{19,20} which are the only relevant

gas species for stable SBSL as all molecular compounds

e.g., \( \text{N}_2, \text{O}_2 \) dissociate and their reaction products dis-
solve in water \cite{10}. As an example we take argon and
describe its concentration in the liquid by the ratio of its
partial pressure \( p_{\text{Ar}} \) to the ambient pressure \( P_0 = 1 \) atm.
The gas temperature follows from a polytropic law \cite{16}
based on results by Prosperetti \cite{21}. The material con-
stants are those for argon dissolved in water at 20 °C. We
supplement the RP equation by a corresponding ordinary
differential equation (ODE) for nonspherical distortions of
the bubble surface derived in \cite{22}. This equation can be
solved with a boundary layer approximation \cite{7,8,23}; see
\cite{8} for details. This approximation slightly overestimates
the shape instabilities due to the simplified treatment of
thermal losses \cite{24,25}; however, the quantitative agree-
ment is still satisfactory \cite{25,26}.

Of the different types of shape instabilities found in
Ref. \cite{8} we focus here on the long time scale parametric
instability, where the perturbations of the spherical surface
can grow from cycle to cycle and finally overwhelm
the bubble. At a given \( f \), bubbles with ambient radii
\( R_0 < R_0^{\text{max}} \) are stable, with \( R_0^{\text{max}} \) only weakly dependent
on \( P_a \) in the SBSL regime. Numerically, we find that
\( R_0^{\text{max}} \) increases with decreasing \( f \); see Figs. 2 and 3. This
finding can be accounted for by the following argument:
The shape instability is driven by a parametric instability
\cite{8} on the time scale of the afterbounces, which is close
to the inverse of the bubble’s eigenfrequency \cite{17}
\( f_e = \sqrt{3P_0/\rho_l/2\pi R_0} \), where \( \rho_l \) is the liquid density.
The exponential growth with time constant \( f_e^{-1} \) is stopped
after a time \( \approx R_0^2/2\nu_l \), due to the damping of the \( R(t) \)
dynamics by the liquid viscosity \( \nu_l \). Over the remaining
afterbounce interval whose length is \( \approx 1/2f \) \cite{9}, the shape
instabilities are themselves damped by viscosity, typically
with half the time constant of the \( R(t) \) damping \cite{8}.

Putting all these facts together finally yields

\[
R_0^{\text{max}} \sim \left( \frac{\rho}{3P_0} \right)^{1/6} \left( \frac{8\pi\nu_l^2}{f} \right)^{1/3}
\]  

(1)

for the maximum ambient radius for which bubbles are
still shape stable in the large forcing SBSL regime, in
decent agreement with the numerical results of Fig. 2.

The instabilities of the Rayleigh-Taylor (RT) type have
not been included here, as for small \( f \) and very intense
collapses they act on too short time scales as to be
described within a uniform bubble model. A correct
description probably requires a detailed modeling of the
pressure and density variations of the bubble interior.
Also, the aforementioned neglect of heat losses affects the
RT instability more severely at the lower \( f \). Qualitatively,
the RT instability sets an upper limit to the driving pressure
(cf. \cite{8}), with this limit decreasing for smaller \( f \).

![FIG. 2. Largest ambient radius \( R_0^{\text{max}} \) for stable argon SBSL bubbles in water at given \( P_a \) = 1.2, 1.3, and 1.4 atm. The dashed line shows the estimate (1).](image)

![FIG. 3. Phase diagram in the \( R_0-P_a \) parameter space for
(a) \( f = 20 \) kHz and (b) \( f = 5 \) kHz. The dashed lines indicate
the \( M_e = 1 \) Mach criterion. The thin solid lines give the
threshold for parametric instability. SL bubbles fulfill both
criteria (shaded region). The thick lines show the diffusive
equilibria for the partial pressure of noble gas \( p_{\text{Ar}}^\infty/P_0 \) indicated
in the graph (branches with positive slope represent stable
equilibria). Note that in (b) \( p_{\text{Ar}}^\infty/P_0 \) has been chosen a factor
of 40 lower than in (a) in order to achieve stable SL at high
driving pressure. For the same gas concentration as in (a), the
5 kHz driven bubble in (b) could not be stable for \( P_a/P_0 > 1.13 \).](image)
Sonoluminescing bubbles must also fulfill the energy focusing condition, which can be estimated by requiring \( M_g = |R/c_g| \geq 1 \) [2,7,8]; \( M_g \) is the Mach number with respect to the speed of sound in the gas \( c_g \). The \( M_g = 1 \) curve is also given in Fig. 3; it undergoes a slight shift towards smaller \( P_a \) and \( R_0 \) when lowering \( f \) because of the enhanced collapse strength.

To summarize, because of the enhancement of shape stability and collapse strength, the SBSL regime where both the energy focusing condition and shape stability are fulfilled (shaded region in Fig. 3) is enlarged for smaller \( f \), allowing for larger bubbles and stronger collapses.

Next we discuss the diffusive stability of the bubbles [2,8,17,27,28]. Up to now we have implicitly assumed that the ambient radius \( R_0 \) can be changed directly in experiment. However, it can only be altered indirectly by adjusting the gas concentration [5,8,29]. Here we are interested in diffusively stable bubbles, as they exhibit the brightest SBSL; unstable bubbles suffer from diffusive growth and shedding of microbubbles; they are less long lived and less bright [2,26].

The diffusive equilibria in the SBSL regime are characterized by the condition [8,28],

\[
p^\infty_{\text{Ar}}/P_0 = \langle p(R,t) \rangle_4 / P_0, \tag{2}
\]

where \( \langle p(R,t) \rangle_4 = \int_0^T p(R(t)) R^4 \, dt / \int_0^T R^4 \, dt \) is a weighted mean of the gas pressure inside the bubble. The right-hand side of Eq. (2) can easily be calculated from the RP dynamics. To very good approximation \( \langle p(R,t) \rangle_4 / P_0 = (R_0/R_{\text{max}})^3 \) [2.9]. As \( R_{\text{max}}/R_0 \) gets very large for the strong collapses at low \( f \) (Fig. 1), the (inert) gas concentration \( p^\infty_{\text{Ar}} / P_0 \) in the liquid must be very small for (2) to be fulfilled. Therefore, much stronger degassing is necessary to get stable bubbles at high driving pressures. In Fig. 3 we plot diffusive equilibria resulting from Eq. (2); only those with positive slope are stable. For \( f = 5 \) kHz (Fig. 3b) the required inert gas concentration to have stable bubbles at, say, \( P_a/P_0 = 1.3 \) is well below \( p^\infty_{\text{Ar}}/P_0 = 0.01\% \). From an experimental point of view a controlled degassing to such tiny concentrations may be difficult. Therefore we recommend to make use of argon rectification by mixing a small amount of argon with pure nitrogen, which is burned off, thus adjusting \( p^\infty_{\text{Ar}} \) to the desired value.

For the experimenter it is convenient to have predictions for phase diagrams in the space of the directly adjustable parameters \( p^\infty_{\text{Ar}} / P_0 \) and \( P_a/P_0 \). These diagrams can easily be extracted from the graphs in \( R_0-P_a \) space (Fig. 3). The results for three different frequencies are shown in Fig. 4. From this figure it can again be seen that lower \( f \) requires lower inert gas concentrations \( p^\infty_{\text{Ar}} / P_0 \).

The larger bubbles and the stronger collapses made us expect more light using smaller frequencies. But how much more? We can quantify the light emission using an extension of the hydrodynamic theory [16]. In this approach, an ODE for the gas temperature inside the (uniform) bubble is coupled to the RP equation. Subsequently, the opacity of the heated gas—whose importance was first recognized in the pioneering work of Moss et al. [15]—and the resulting light emission (primarily from thermal bremsstrahlung [15,16]) are computed, showing good agreement with the available experimental data. Lowering the driving frequency, we predict that, e.g., at \( f = 5 \) kHz more than 100 times as many photons are emitted as at \( f = 20 \) kHz; see Fig. 5a. The maximum temperatures in the bubble reach about 60 kK at \( f = 20 \) kHz. The widths of the light pulses can exceed 1000 ps, much longer than hitherto observed [5] for the standard frequencies (Fig. 5b).

We make the explicit prediction that for these long and intense light pulses at 5 kHz the width in the red spectral regime should be nearly twice as long as in the blue spectral regime, in contrast to what has been observed at larger \( f \) [5].

We suggest to experimentally check the predictions for the light intensities and widths of the light pulses in the proposed parameter regimes, i.e., for low frequencies and the correspondingly low inert gas concentrations (cf. Fig. 4). Such experiments will validate the present understanding of SBSL and may lead to refinements of the model. In particular, one will be able to better judge the role of water vapor. Clearly, the lower \( f \), the more water molecules will enter the bubble upon expansion and will modify the light emission process upon collapse [30]. Likewise, the gas composition may become different at the center of the
bubble and near its wall [31]. The neglect of thermal
damping [24] will furthermore become more and more
questionable for increasing peak temperatures, i.e., for
decreasing \( f \). Also, the electronic degrees of freedom
of argon will eventually limit the compressional heating
of the gas.

The good agreement between experiment and theory
[16] suggests that the processes mentioned in the previ-
ous paragraph are of minor importance in the hitherto ex-
plorated parameter range of \( f = 20-40 \) kHz. Therefore,
for the time being, we neglected them also in the neigh-
boring regime \( f = 2-20 \) kHz. Eventually only the com-
parison to experiment will tell down to which frequencies
these approximations are justified and reveal how much
further one can push the limits of SBSL towards the re-
region of extreme and extravagant states of matter.

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