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Comparison of Xe single bubble sonoluminescence in water and sulfuric acid*

An Yu(安 宇)

Physics Department, Tsinghua University, Beijing 100084 *and*
Institute of Acoustics, Chinese Academy of Science, Beijing 100080, *China*

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Using the equations of fluid mechanics with proper boundary conditions and taking account of the gas properties, we can numerically simulate the process of single bubble sonoluminescence, in which electron–neutral atom bremsstrahlung, electron–ion bremsstrahlung and recombination radiation, and the radiative attachment of electrons to atoms and molecules contribute to the light emission. The calculation can quantitatively or qualitatively interpret the experimental results. We find that the accumulated heat energy inside the compressed gas bubble is mostly consumed by the chemical reaction, therefore, the maximum degree of ionization inside Xe bubble in water is much lower than that in sulfuric acid, of which the vapour pressure is very low. In addition, in sulfuric acid much larger p_a and R_0 are allowed which makes the bubbles in it much brighter than that in water.

Keywords: sonoluminescence, ionization, Xe bubble
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1. Introduction

It used to be considered that water is the favourite liquid for single bubble sonoluminescence (SBSL) since this phenomenon was observed.[1−3] It was not until 2000 that Suslick's group observed a brighter SBSL in many other low vapour pressure liquids,[4] and further in 2005 they observed a moving SBSL in sulfuric acid thousands of times brighter than that in water.[5] Chen's group reported that in a recent experiment they observed a steady SBSL in sulfuric acid solution as bright as the moving SBSL.[6] Besides the brightness of SBSL in sulfuric acid, the atomic line spectrum is visible,[5] which used to be submerged in the background of the featureless spectrum of SBSL in water. What makes such differences of SBSL in sulfuric acid and in water? It is easy to notice that the vapour pressure of sulfuric acid is much lower than that of water, therefore, different from SBSL in water, the vapour effects of SBSL[7−10] are negligible in sulfuric acid, and so are the contributions from the radiative attachment. Another important difference lies in the viscosity. Sulfuric acid is much more viscous than water. The calculation of the shape instability and Rayleigh–Taylor (RT) instability of SBSL bubbles shows that the parameter area of stable SBSL bubbles in the more viscous liquid is much wider. For example, driven by ultrasonic wave of frequency ∼30 kHz, a stable SBSL bubble in 85 wt.% sulfuric acid the radius of the bubble R_0 can be larger than 15 μm and the amplitude of the driving acoustic pressure p_a can extend to 6 atm,[11] while in water the maximum $R_0 \sim 6\,\mu$m and the maximum p_a ∼1.6 atm (1 atm = 1.01×10^5 Pa).[12] On the other hand, for Xe bubble under a lower amplitude of driving acoustic pressure of ∼1.5 atm, the experiment shows that sometimes the continuum part of spectrum of SBSL in sulfuric acid at room temperature is similar to that in water at 1°C.[13] Thus we may expect that the mechanism of SBSL in sulfuric acid at room temperature and in water at low temperature should have some similarities. In order to understand more deeply the phenomena of SBSL, we need to make careful calculation and comparison of their differences in sulfuric acid and in water in detail.

2. Calculation model

The present calculation model is almost the same as that sketched in Ref.[14], where the equations of fluid mechanics of two kinds of gas components, inert gas and water vapour, are employed to describe gas dynamics inside the bubble; the mass action law[15] is applied to evaluate the production of

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chemical reaction and the ionization; the phase transition of vapour and heat exchange at the boundary of bubble are considered; electron–neutral atom bremsstrahlung,[16] electron–ion bremsstrahlung and recombination radiation,[17] and the radiative attachment of electrons to atoms and molecules[14,18] are included in the light emission calculation.

One of the differences lies in the RP equation. The different forms of the equation lead to a small difference in the bubble motion, but a relatively large difference in the gas dynamics results.[19] Yet, this problem has not been solved, and we will not repeat the discussion here. We simply adopt a relatively prevalent form, the Keller–Miksis formulation,[20] and make the small improvement:

$$\begin{aligned} &\mathrm{e}^{-M} R\ddot{R} + \frac{3}{2}\mathrm{e}^{-M/3}\dot{R}^2 \\ &= \mathrm{e}^{M}\frac{1}{\rho_{\mathrm{l}\infty}}[p_{\mathrm{l}} - p_\infty - p_{\mathrm{s}}(t+t_R)] + \frac{t_R}{\rho_{\mathrm{l}\infty}}\dot{p}_{\mathrm{l}}, \end{aligned} \quad (1)$$

where t is the time, $R(t)$ is the radius of the bubble, $\rho_{\mathrm{l}\infty}$ the ambient liquid density, p_∞ the ambient pressure, $p_{\mathrm{s}}(t) = -p_{\mathrm{a}}\sin(\omega t)$ the driving acoustic pressure, $t_R \equiv \dfrac{R}{c_{\mathrm{l}\infty}}$, $c_{\mathrm{l}\infty}$ being the sound speed in the liquid at the ambient temperature and pressure of 1 atm, $p_{\mathrm{l}} = p_{\mathrm{g}}(R,t) - \dfrac{4\eta\dot{R}}{R} - \dfrac{2\sigma}{R}$, which is the pressure on the liquid side of the bubble wall, $p_{\mathrm{g}}(R,t)$ the pressure on the gas side of the bubble wall, η the dynamic viscosity, σ the surface tension coefficient of the liquids. The parameter $M \equiv \dfrac{\dot{R}}{c_{\mathrm{l}\infty}}$ is the bubble-wall Mach number which in fact embodies the effect of the liquid compressibility. The improvement is just to replace $1 \pm M$ by $\mathrm{e}^{\pm M}$ and $1 - M/3$ by $\mathrm{e}^{-M/3}$, while the displacement makes almost no difference when $|M| \ll 1$, but when $|M| > 1$, it keeps the sign of right term in (1) unchanged and offsets the drawback of the Keller–Miksis formulation to some extent. As for the numerical results, the improvement has only little effect on them.

The formula of the radiation power is slightly different from that given in Ref.[14]. In opaque media, the more appropriate formula of the total emitted power from the bubble per wavelength interval at wavelength λ should be

$$\begin{aligned} P_\lambda(t) = 8\pi^2 &\int_0^R \int_{-1}^1 \kappa_\lambda(r) P_\lambda^{\mathrm{Pl}}(r) \\ &\times \mathrm{e}^{-\int_{rx}^{\sqrt{R^2 - r^2(1-x^2)}} \kappa_\lambda \mathrm{d}s} r^2 \mathrm{d}r\mathrm{d}x, \end{aligned} \quad (2)$$

where r is the radial coordinate, P_λ^{Pl} the Planck radiation intensity, κ_λ the absorption coefficient.

3. Results and discussion

With the present model, we may calculate those cases of experiment reported in Ref.[13], where the SBSL of Xe bubble in water and in sulfuric acid is compared. Figure 1 shows the spectrum of SBSL of Xe bubble in water (WA) at 1°C and in sulfuric acid (SA) at room temperature. The concentrations of gas

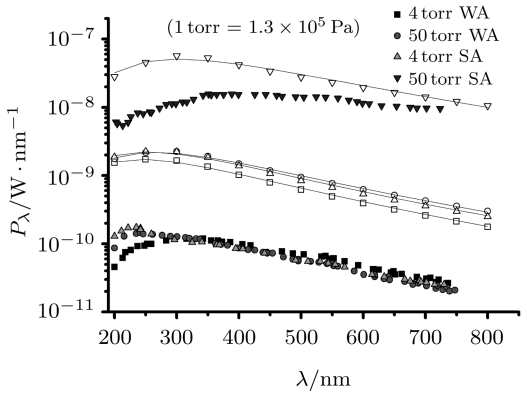


Fig.1. Spectra of SBSL of Xe bubble in water (WA) and in sulfuric acid (SA), in which the gas concentrations are 4 torr and 50 torr respectively. The filled symbols are the experimental data digitized from Ref.[13], and the open symbols are calculated with the present model. Solid lines are the best fit to the black body radiation formula.

in the liquids are 4 torr and 50 torr respectively. It seems that there are systematic errors existing between the calculation and the observation. We notice that all values of the radiances of the brightest Xe bubbles in water near freezing point reported in Ref.[3] are $\sim 10^{-9}$ W/nm, while in Ref.[13] the observed values of radiance of bright Xe bubbles in water near freezing point are $\sim 10^{-10}$W/nm, see Fig.1. This discrepancy may lead to the difference between the experiment and the calculation. We do not know the exact reason yet. However, the calculation still shows the essential feature that the spectra of SBSL of Xe bubble in 4 torr and 50 torr water at 1°C and that of 4 torr sulfuric acid at room temperature are similar and much lower than those in 50 torr sulfuric acid at room temperature. Also, the calculated spectrum can be well fitted by the black body radiation formula, though the fitted temperatures are a little different from that of the experimental data. Therefore, we are certain that the present calculation to a certain extent reflects the real processes. We must point out that in the calculation for the WA bubbles, the radius R_0 and the amplitude of the driving acoustic pressure p_{a} are the same as that provided by Ref.[13], which makes the calculated

maximum radius of the bubble consistent with the observed one. For SA bubbles, R_0 is still the same as in Ref.[13], but p_a has to be adjusted to fit the maximum radius of the bubble observed. In the present calculation, p_a is adjusted to 1.76 atm for 4 torr SA bubble and 2.1 atm for 50 torr SA bubble respectively.

The fitted Black body radiation temperatures for four cases in Fig.1, namely 4 torr and 50 torr WA and 4 torr and 50 torr SA, are respectively 11,965 K, 11,072 K, 11,580 K and 9,651 K. As we have discussed in the earlier paper,[14] the fitted temperature has no connection with the real temperature inside the bubble. The good fit may be due to the narrow range of the spectrum, with wavelength ranging from 200 nm to 800 nm. Figure 2 shows the spatial distribution of temperature inside the bubble when the radiation from the bubble is the strongest. With similar spectrum (see Fig.1), the three cases, 4 torr and 50 torr WA and 4 torr SA bubbles, have similar maximum temperature ∼20,000 K, which is much higher than the temperature fitted by the black body radiation formula. Does this mean they have the similar mechanism of SBSL? We know the ambient vapour pressure of the 4 torr SA at room temperature is over 100 times less than that in water at 1°C, therefore, there must be much more residual water vapour remaining inside the WA bubble as the bubble is at its flashing phase. We find the contribution of the bremsstrahlung and recombination radiation is almost 100% for the SA bubble, while for 4 torr and 50 torr WA bubbles this contribution is 77% and 83%, respectively, and the rest parts are contributed by the radiative attachment.

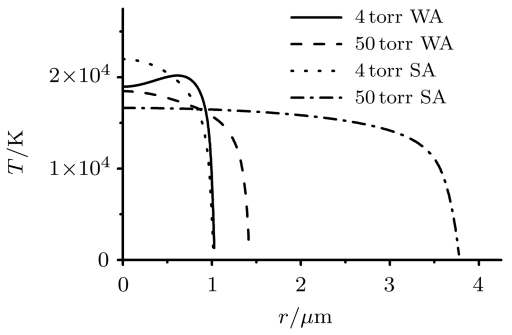


Fig.2. Spatial distributions of maximum temperature inside the bubbles.

A contribution of about 20% from the radiative attachment is typical for Xe bubble in water, which means the water vapour effects are very important for the WA bubble. We must notice why those three cases discussed above have the similar spectrum and the similar maximum temperature (see Figs.1 and 2). It is just the values of R_0 and p_a that accidentally gave rise to this happen. The 50 torr SA bubble has the largest R_0 and Fig.1 shows that it is the brightest. It is unexpected that the maximum temperature of the 50 torr SA bubble shown in Fig.2 seems the lowest. In fact, its largest size makes it the brightest, since there should be more heated atoms and molecules inside the bubble then, which makes greater contributions to the light emission. Brightness is correlated with both the maximum temperature and the bubble size.

On the other hand, our calculation shows the pulse width of those bright SBSL bubbles used to be large. The measured pulse width of the 4 torr SA bubble is < 0.5 ns but that of the 50 torr SA bubble is ∼10 ns,[13] while the present calculation gives the full width at half maximum (FWHM) of the SBSL pulse ∼0.7 ns and ∼5.5 ns, respectively. Different from the Ar bubble, the FWHM of SBSL Xe bubbles is wavelength-dependent. Figure 3 shows that the FWHM values always increase with wavelength, whether for SA or for WA bubble. The possible reason is that the Xe bubbles are some what optically

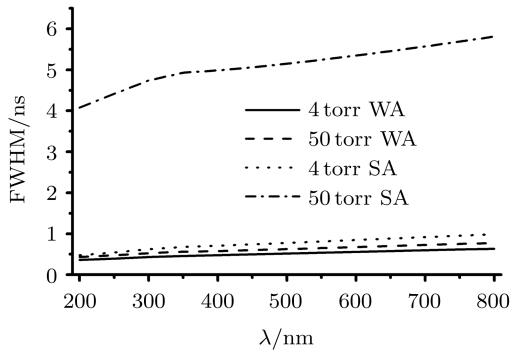


Fig.3. FWHM values vs. wavelength of SBSL.

thick.[17] We may consider the relative change of FWHM, i.e. $(t_{red} - t_{UV})/t_{ave}$, where t_{red}, t_{UV} and t_{ave} are the FWHM's of red, UV and the average of the total, respectively. The UV range is set as wavelengths 300–400 nm and red range 590–650 nm in the calculation. From Fig.3 we can easily evaluate those relative changes of FWHM, which are similar for 4 torr and 50 torr WA bubbles and 4 torr SA bubble, being ∼0.26, but for 50 torr SA bubble it is ∼0.13. That may imply that the 50 torr SA bubble is optically thinner than the other three bubbles. In order to make things

clearer, we may calculate the coefficient of absorption length of SBSL bubble. Figure 4 shows the product of the coefficient of absorption length and the bubble radius $R\kappa_\lambda$ for various wavelengths. If $R\kappa_\lambda \gg 1$, it means the bubble is opaque, and if $R\kappa_\lambda \ll 1$, it means the bubble is transparent. We can see that for visible light and infrared, 4 torr and 50 torr WA bubbles and 4 torr SA bubble are opaque, but 50 torr SA bubble is neither opaque nor transparent, which is consistent with the result presented in Fig.3.

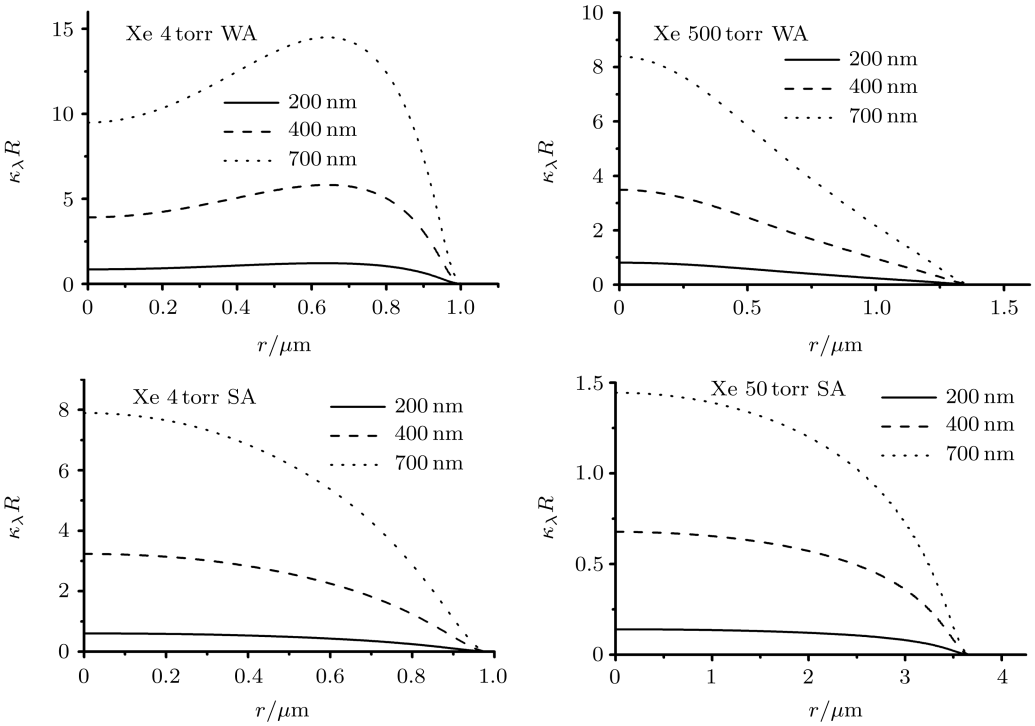


Fig.4. Spatial distribution of coefficient of absorption length (κ_λ) inside the bubbles for various wavelengths multiplied by the bubble radius R corresponding to the cases of Fig.2.

Another interesting phenomenon of SBSL in sulfuric acid is the appearance of atomic line spectrum[5] which used to disappear in water. Due to the limit of the shape instability and Rayleigh–Taylor (RT) instability, there are upper limits of R_0 and p_a.[21,22] In the present cases, parameters of 4 torr WA and 50 torr WA are almost at the threshold. The larger p_a is, the higher the maximum temperature is, which leads to the larger ionization in general. We find the degree of ionization α_e(= electron number/total atom and molecule number) is roughly less than 10% when the bubble in water is at its maximum temperature, which may be larger when the bubble in SA, see Fig.5. On the other hand, upper limits of R_0 and p_a are much larger in 85% wt SA.[11] We simply choose $R_0 = 13\,\mu$m and $p_a = 4$ atm for Xe bubble in SA, which may be close to the circumstance when the observed line spectrum of SBSL is notable. For contrast, an Ar bubble under the same conditions is calculated too. The calculated results show that the degree of ionization α_e can be increased to > 50% for Ar and > 70% for Xe

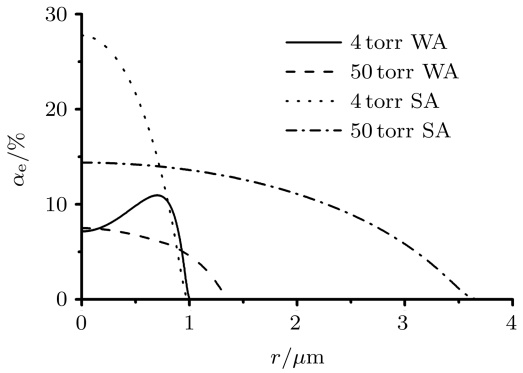


Fig.5. Distribution of the degree of ionization α_e inside the bubble corresponding to the cases in Fig.2.

bubble, see Fig.6. If we choose larger p_a or smaller R_0, the degree of ionization α_e will be much larger. Since there is a large number of atoms and molecules ionized which consume much heat energy, the maximum temperature only increases to ∼40,000 K. The large

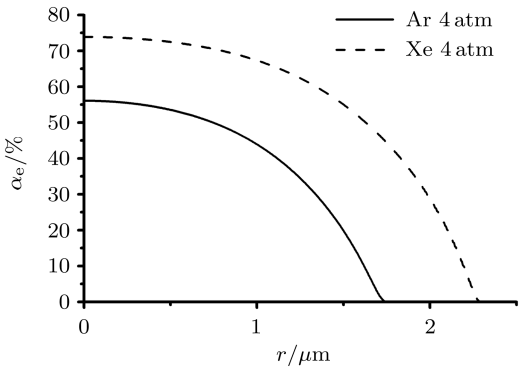


Fig.6. Distribution of the degree of ionization α_e inside Ar and Xe bubbles in SA with $R_0 = 13\,\mu$m and $p_a = 4$ atm as the bubble at its maximum temperature.

amount of ionization events makes more atoms to be stimulated to its excitation states through the process of the combination radiation and leads to the emergence of atomic line spectrum on the continuum background. In Ref.[5], the appearance of atomic line spectrum of those SBSL bubbles in SA is attributed to the transient existence of the hot plasma core in the bubble. The present calculation verifies their prediction.

4. Conclusion

The present calculation of SBSL can not accurately reproduce the whole experimental data, however, it can reveal the most interesting features qualitatively. According to the calculation, we find the greatest distinction between WA bubble and SA bubble lies in the degree of ionization inside the collapsing bubble. Due to the water vapour effects, most of the heat energy is consumed by the chemical reaction of decomposition and the maximum degree of ionization inside WA bubble is only ∼10%. However, for SA bubble, since the vapour effects are negligible, even if its maximum temperature is similar to that of the WA bubble, the maximum degree of ionization can be much higher than 10%. In addition, in SA p_a can be much larger than that in WA, therefore, the maximum degree of ionization even gets to ∼80%, and R_0 in SA can be larger, which makes the SA bubble much brighter than the WA bubble.

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