Theory of the Origin of Brief Sonoluminescence Light Pulses

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Abstract

An explanation of the origin of the ultrafast light pulses in sonoluminescence experiments is presented. The pulses are due to black-body radiation emitted under the conditions of high temperature and negative dielectric constant. A simulation of the bubble content as a hard sphere gas was employed in order to reach this conclusion. A discussion of the spatial profile of the density and the temperature in the bubble is given.

Sonoluminescence (SL), the production of light from an ultrasonically modulated bubbles, has been the object of study for over sixty years[1]. Until recently [2-4], all experiments were performed with bubble fields, so that precise data about the length of the SL pulse and the power emitted from the single SL event could not have been obtained. In [5], the explanation of SL occurring during transient cavitation based on the chemical process going on in the bubbles was given. This approach could not explain experimental results given in [2-4]: Very short duration of the pulses (below $50\,ps$), peak power (lower limit is $30\,mW$), and the spectrum, which is black-body like and extends into the ultraviolet.

The time scale of the flashes was found to be sufficiently short to rule out conventional radiative relaxation processes, which requires time scale of the order of nanoseconds, or more. On the other hand, the energy spectrum is broad enough to rule out collective effects like superradiance or superfluorescence or laser type mechanism. The explanation of the phenomenon as a thermal radiation can not be understood in a simple way, taking into account that the radius of the bubbles in question is just a few times larger than the wavelength of the emitted light. Detailed measurements of the radius of the bubble as a function of time [4] offer the possibility that the wall of the bubble achieves supersonic speeds prior to the onset of the luminescence. It has been proposed that SL is associated with the production of shock waves in the air in the bubble [4].

In this paper we show our analysis of the dynamics of the bubble and how it leads to the possible explanation of the experimental data about SL.

From the experimental data, especially from Fig. 4b in [4] it was not clear to us that the conditions for launching the shock waves inside the bubble are really satisfied, especially considering the dependence of the speed of sound c_s on the local parameters (pressure, density), $c_s = \sqrt{\gamma p/\rho}$. Actually, assuming adiabatic conditions in the bubble, it follows that the speed of sound in the bubble close to the point of maximum contraction, there SL emission actually happens, is even higher than its standard value.

Our way to resolve this question was to try to simulate the conditions in the bubble and the dynamical evolution of the system in the sound field. The motivation for this project was also the long lasting discussion in the literature [6] about the commonly assumed approximation that macroscopical variables (pressure, density, temperature) in the bubble are independent of position.

We model our bubble as follows: a certain number of spheres are randomly distributed inside a pulsating spherical cavity. These spheres (simulating atoms/molecules) are given random velocities according to the Maxwellian distribution and are allowed to specularly collide with each other and with the bubble wall. Between the collisions, free flight is assumed. The rotational degrees of freedom are not included, so we are able to simulate a monoatomic gas. The collisions are assumed to be elastic and no correction for the loss of the kinetic energy due to ionization is included. The parameters for the simulation are chosen in such a way that the internal energy, the mean free path for the collisions and the mass density inside the bubble are the same as in experiment [4]. The temperature in the bubble is obtained by examining the local velocity fluctuations away from the mean flow velocity and extracting the kinetic energy fluctuations. The pressure is obtained as the ratio of the average force (which acts on the bubble wall because of the collision with the spheres) to the area of the bubble.

The dynamics of the bubble wall is assumed to follow from Rayleigh-Plesset (R.P.) equation

$$R\ddot{R} + \frac{3}{2}\dot{R}^2 = \frac{1}{\rho_l}\left[P_g(R) - P_a(t) - P_0 - \frac{2\sigma}{R}\right] + \frac{R}{\rho_l c_l}\frac{d}{dt}[P_g(R) - P_a(t)] - 4\nu_l\frac{\dot{R}}{R} \tag{1}$$

Here $P_g(t)$ is the pressure in the gas, $P_a(t)$ is the acoustic pressure given by $P_a(t) = P_a \sin(\omega_a t)$, P_0 is the hydrostatic pressure, ρ_l is the density of the liquid, c_l is the speed of sound in the liquid, ν_l is the kinematic viscosity of the liquid, and σ is the surface tension. The bubble content enters into this equation through $P_g(t)$ term. Assuming the adiabatic conditions in the bubble, $P_g(t)$ follows from the van der Walls equation of state

$$P_g = P_0 \, \frac{(R_0^3 - a^3)^\gamma}{(R^3 - a^3)^\gamma} \tag{2}$$

where R_0 is the equilibrium radius of the bubble and a is the hard core radius. Equation (1) was solved numerically (Runge - Kutta method with repeated Richardson extrapolation was used), and the results are shown in Fig. 1. The maximum radius, for the choice of parameters like in [4] is smaller ($27\,\mu m$, compared to $40\,\mu m$). Since all other parameters are the same, we attributed this difference to the effect of surface tension, which was neglected in [4][1] Equation (1) is valid assuming that the speed of the bubble wall is much smaller than the speed of sound in the liquid and that the effects like mass diffusion, rectified diffusion and thermal conductivity can be neglected.

Let us go back to our simulation. In order to simplify the problem, we started the evolution of the system from the maximum radius, assuming that all terms on the right hand side of R.P. equation, Eq. (**??**), can be safely neglected. (The effect of these terms grow with time, but the time scale for the collapse of the bubble from the maximum radius to the minimum is much shorter than the acoustic period.) At the point where the pressure developed in the bubble was high enough to influence the motion of the wall, the term $P_g(R)/\rho_l$ was included[2]. In order to keep the computing time at a reasonable level, the number of spheres was limited to 1,000.

Our results for the average pressure and the temperature versus radius of the bubble are shown in Fig. 2 and Fig. 3. It can be seen that the results of our simulation and the van der Walls fit are in very good agreement. By

[1] For the time being, the role of the surface tension in determining the bubble dynamics is not very clear, because of the small bubble size.

[2] The other terms on the right hand side of Eq. (**??**) should not be very important compared to the pressure in the gas, which gets to be very high.

fitting the average temperature in the bubble to the form

$$T(R^3 - a^3)^{\gamma-1} = C \tag{3}$$

where R is the bubble radius, we obtained the values for the fitting parameters $\gamma = 1.678$, $a = 4.413 \times 10^{-5}\,cm$ and $C = 4.77 \times 10^{-5}$. These results are consistent with those obtained from the analogous pressure fit. The fitting value of γ is very close to the value for monoatomic gases (5/3), as might be expected. The excluded radius a gives the result that the ratio of the excluded volume to the volume actually occupied by the spheres is equal to 5.3 ± 0.7, close to the value of 4 which is given, for example, in [7]. The excluded volume is of crucial importance, giving a high temperature close to the point of the maximum contraction. We also noted a well defined spatial dependence of the gas density in the bubble. The results for the collapse part of the cycle are shown in Fig. 4. We see that the density is higher close to the wall, as it is expected on the basis of simple dynamical arguments. During the expansion part, the density is higher close to the bubble center. Temperature has the similar behavior, with the surface temperature slightly higher than inside. The time profile of the average and the surface temperature is shown in Fig. 5. The fluctuations because of the statistical effects are high, so, at this point, our conclusions about space dependence of the density and the temperature in the bubble are more of qualitative nature. Anyway, this spatial dependence, for the variety of the initial conditions could not be connected with the jump conditions, characteristic for the strong shock waves. So, our conclusion is that production of a strong shock waves in the bubble, under the conditions reported in [4], should not be expected. We hope that these results can be useful for a deeper understanding of the dynamics of the bubbles in a sound field.

Let us now go back to the main question of the SL emission. Using our results from the simulation for the temperature profile, it is easy to see that the percentage of ionized atoms (molecules) is very high close to the point of maximum contraction, where temperature gets to be very high. In order to calculate the number of electrons produced, we use Saha equation with the criteria that maximum number of electrons can not be bigger than number of atoms (single ionization was assumed):

$$n_e = -\frac{x}{2}\left(1 - \sqrt{1 + \frac{4\rho}{x}}\right) \tag{4}$$

where n_e is the density of electrons, ρ is the atomic (molecular) density, and x is the usual Saha expression for the density of electrons[3]. This equation have the correct limits for the $x \gg \rho$ (saturation) and $x \ll \rho$. Furthermore, the effect of the screening was taken into account, what leads to the correction to the ionization energy of the form $I_{cor} = e^2/l_D$, where e is the electron charge and l_D is the Debye length, which is dependent on the electron density, so self-consistent calculations are required. As parameters we used ionization energy of $16\,eV$ and the ratio of the statistical weights of ionized and neutral states, entering the Saha equation, was taken to be 1.

Knowing the concentration of electrons, we were able to calculate the plasma frequency $\omega_p = \sqrt{4\pi n_e e^2/m_e}$, which turned out to be very high, of the order of $10^{16}\,Hz$. This says that the dielectric constant for the radiation of frequencies lower than plasma frequency is negative, as it follows from the

[3]This electrons should be expected to radiate according to the bremsstrahlung formula. We explored this possibility and concluded that the intensity of bremsstrahlung emission is much lower than the SL emission. Also, the spectrum does not correspond to the one reported in [3].

expression for dielectric constant

$$\epsilon(\omega) = 1 - \frac{\omega_p^2}{\omega^2} \tag{5}$$

effectively leading to the attenuation of the radiation from the inside of the bubble, and to the emission of the black-body like spectrum from the surface layer of the bubble, as observed in the experiment [3]. Our results for the power emitted versus time are shown in Fig. 6. It is characterized with the width of $60\,ps$ (full width at half maximum), what is consistent with the experimental results [4]. The slight asymmetry should also be noticed (as well as in Fig. 5). This asymmetry gives natural explanation of the results for the shape of the SL pulse reported in [2], which show fast rise and slower decay of the pulse. In Fig. 7 we presented the results for the spectrum of emitted radiation. Together with the results which we obtained using the approximation that ionization has no effect on the kinetic energy of the molecules, we also present the results following from the assumption that black-body emission will start at the point where the ionization is almost complete. The critical temperature for the complete ionization is gas and density dependent - for the ionization potential of nitrogen of $16\,eV$, and for the densities of $10^{22}\,g/cm^3$, it is about $23,000K$, what closely corresponds to the black-body fit in [3].

Finally, we are able to show that the length of the pulse is comparable with $50\,ps$, the number of photons (10^6) and the power emitted (peak power of a few hundreds mW) are consistent with the lower limits given in [4]. Our approach simulates the monoatomic gases, so on purely hydrodynamic reasons (the ratio of the specific heats is higher), it is expectable to get higher intensity of radiation [1].

We have to mention that negative dielectric constant (imaginary index of refraction) is going to have influence on the results for Mie scattering of light from the bubble. Since this is the way how bubble radius versus time diagram was obtained [4], it might happened that this effect leads to the not absolutely correct values for the bubble radius. Our simulation shows that the minimum bubble radius can be as small as $0.6\,\mu m$, compared to the values $0.8-1.0\,\mu m$ as reported in [4][4].

Our approach is capable of explaining all major characteristics of SL radiation: short time of the flashes, the high concentration of energy and the spectral distribution. We are looking forward for the future experiments, done with the different gases and liquids, which will help to determine the correctness of our approach. It would be especially useful to have experiments performed with rare gases, to verify if the intensity of radiation is much higher, as it will be expected from our theory. Our approach could be improved as well; Next step would be to try to simulate diatomic molecules, with the hope that this approach could be successfully used to explain number of other physical phenomena connected with the bubbles in liquids.

[4]This discrepancy is explained there assuming the launching of the shock wave which should slow down the contraction of the bubble wall.

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Figure Captions

Fig. 1. The bubble radius as a function of time. $R_0 = 4.5\,\mu m$, $P_a = 1.325\,atm$, $c_s = 1,481\,m/s$, frequency of acoustic field $f_a = 26.5\,K\,Hz$, $\nu_l = 0.01 cm^2/s$, $\sigma = 72.5\,dyn/cm$.

Fig. 2. The pressure of the gas versus radius of the bubble (log - log scale). Simulation with 1000 spheres, $R_0 = 4.5\,\mu m$, 3×10^9 molecules in the bubble, maximum radius $R_m = 50\,\mu m$, ambient temperature $T = 300\,K$. The points are the result of simulation and the line is a least square fit to $P = C(r^3 - a^3)^\gamma$, fitting parameters are $\gamma = 1.69$, $a = 2.67 \times 10^{-5}\,cm$, $C = 2.45 \times 10^{-11}$.

Fig. 3. The average temperature versus radius (log - log scale). The parameters are the same as for Fig. 2. The points are the result of the simulation, the solid line is a fit to $T(r^3 - a^3)^{\gamma-1} = C$; resulting parameters are given in text.

Fig. 4. The density of the gas in the bubble versus radial coordinate for a few different bubble radii. Apscisa is radial coordinate and ordinate is $\rho^{1/3}$. Here R_0 is the current size of the bubble. The parameters are the same as before and what is shown is a fit to $\rho(r) = A + B\ r^2$;

Fig. 5. The temperature of the gas in the bubble versus time. The solid line is the average temperature and points represent the surface temperature. The parameters are the same as before.

Fig. 6. The power emitted versus time. The parameters are the same.

Fig. 7. The spectral energy of a single flash. The points are the experimental data reported in [3], the solid line is the output from the black-body formula assuming that radiation takes place for all temperatures and the doted line represent the radiation for temperatures higher than $30,000K$.