Radiation from a Sonoluminescencing Bubble

L. Kondić, Joel I. Gersten and Chi Yuan*
City College of The City University of New York New York, NY 10031
(September 29, 1994)

A solution to the coupled gas dynamic and radiation transfer equations, as it applies to a sonoluminescencing bubble, is presented. Important modifications to the thermodynamic variables arise from radiation, ionization and dissociation processes. The theory is compared with experiment and provides a possible explanation for sonoluminescence.

78.60Mq, 44.40+a, 47.55Bx, 47.40-x

Sonoluminescence (SL), the emission of light from an ultrasonically driven bubble in a liquid, has been the object of considerable study for more than sixty years [1]. Recently [2] it was found that the brief flashes of light are of very short duration (about $50\,ps$) and possess a broad spectral range, spanning the visible spectrum and extending into the ultraviolet, consistent with that of a black body. Furthermore, measurements have shown that the wall of the bubble can achieve supersonic speeds prior to the onset of luminescence. It has been proposed that SL is associated with the production of shock waves in the air inside the bubble [2]. Theoretical calculations, based on the gas dynamic equations coupled to the bubble wall dynamics, have found shock waves [3]. The predicted high temperatures, coupled with a bremsstrahlung mechanism, were used to explain SL. In this letter we find that important modifications of this theory occur if these equations are solved self-consistently with the radiative transport equation. In particular the shock core temperature is lowered by an order of magnitude. We also focus on the spectral properties of the emitted radiation and find that pure bremsstrahlung alone cannot account for the observed spectrum [3].

The physical problem is an air (diatomic gas) bubble in water, of equilibrium radius R_0 of about $4\,\mu m$, subjected to a sound field of typical ultrasound frequency ($20\,KHz$) and acoustic amplitude P_a of about $1.3\,atm$. The motion of the bubble wall follows from the Rayleigh-Plasset (R.P.) equation [4]. It is necessary to solve the R.P. equation coupled with the spherically symmetric gas dynamics equations (cf. [3] for the Eulerian sourceless equations)

$$\frac{\partial V}{\partial t} - \frac{\partial}{\partial \xi}(r^2 u) = V^2\ L, \qquad \frac{\partial u}{\partial t} + r^2\ \frac{\partial p}{\partial \xi} = F \qquad \text{(1a,b)}$$

$$\frac{\partial E}{\partial t} + \frac{\partial}{\partial \xi}(r^2\ u\ p) = -\Lambda, \qquad \frac{\partial r}{\partial t} - u = 0 \qquad \text{(1c,d)}$$

where $V = 1/\rho$ is the specific volume, $E = \epsilon(p,\rho) + u^2/2$ is the energy per unit mass and $d\xi = (r^2/V)\ dr = \rho r^2 dr$ connects the Lagrangian coordinate ξ with r. The variables p, ρ, u are the pressure, density and velocity of the gas, respectively. L, F and Λ are mass loss, body force and energy loss per unit mass. Discounting dissociation and ionization, the equation for the internal energy per unit mass is $\epsilon(T) = Nk_BT/(\gamma-1)$ which, together with van der Waals equation $p(1-b\rho)/\rho = Nk_BT$ gives $\epsilon(p,\rho)$. Here b is the inverse of the maximum allowed density, N is the number of particles per unit mass, k_B is the Boltzmann constant and γ is the ratio of specific heats. Equations (1) were solved using a first order Godunov scheme [5] which is known to work well for problems involving shock waves. Previously [3] a modified Lax scheme had been utilized.

The results for the pressure, temperature and velocity of the gas without inclusion of loss terms are presented in Fig. 1. They are similar to those given in [3]. Consider the space-time dependence of the gas temperature. After formation, the shock collapses to the center (left-forward peak), rebounds, collides with the bubble wall and partially reflects from it. The reflected wave converges to the origin again and, as a weaker disturbance, rebounds outwards. Near the point of complete collapse of the shock wave p, u and T follow the expected self-similar behaviour [6] and the temperature reaches a few million degrees. These results imply that some very energetic processes, including nuclear fusion could occur [2]. But, they also imply that the loss terms entering Eqs. (1) will have considerable importance. These loss terms account for the following: 1) Inclusion of vibrational degrees of freedom ; 2) Dissociation; 3) Ionization; 4) Radiation losses.

The temperature at which vibrational degrees of freedom unfreeze is typically a few thousand degrees. At high enough temperatures [7], we included this effect simply by changing the ratio of heat capacities from 7/5 to 9/7. The dissociation of air starts at temperatures of the order of $10^4\,K$. The net result is that the number of particles per volume increases from N to $N[1+\alpha(T)]$ (where $\alpha(T)$ is the degree of dissociation) and there is kinetic energy loss. Inclusion of ionization is more complicated. Already at temperatures of few times $10^4\,K$ there are a considerable number of double and triply ionized atoms. We used a continuous approximation to the Saha equation which gives the average degree of ionization $\overline{m}$ (number of electrons per initial molecule). The equation of state which allows for ionization is

$$\epsilon(T) = \frac{N_a}{\gamma_a - 1}k_BT + \frac{N_m}{\gamma_m - 1}k_BT + \frac{N_a + N_m}{\gamma_e - 1}\overline{m}k_BT + (N_a + N_m)Q(\overline{m}) \qquad (2)$$

where N_a, N_m are the numbers of atoms and molecules per unit volume, respectively, $\gamma_m, \gamma_a, \gamma_e$ are the heat capacity ratios for molecules, atoms and electrons. $Q(\overline{m})$ is the energy required to remove an electron with ionization potential corresponding to $\overline{m}$. The final term in Eq. (2) was included in Eq. (1c) as energy loss.

The radiation process produces SL and strongly affects the values of the thermodynamic quantities. We will present two different approaches to the problem. The first uses the radiative transfer equation [8]. We refer to this as the "thermal approach". We also developed a model (bremsstrahlung (BRS) model) for comparison purposes.

a) *Thermal approach;* Inclusion of radiation losses involves coupling the radiative transfer equation:

$$\frac{1}{c}\frac{\partial I_\nu}{\partial t} + \mathbf{\Omega} \cdot \nabla I_\nu = \kappa_\nu' (I_{\nu p} - I_\nu) \tag{3}$$

with the gas dynamics equations. I_ν is the frequency-dependent radiative intensity of the body, $I_{\nu p}$ is the Planck distribution and κ_ν' is the attenuation coefficient corrected for induced emission. The unit vector $\mathbf{\Omega}$ specifies the direction of photon propagation. The time t_p required for radiation-matter equilibrium to be established is of the order of $1/c\kappa_\nu'$ and is short compared to the time the bubble spends near its minimum radius. Thus Eq. (3) can be employed without the time derivative term. Solving this simplified equation and calculating the energy flux $S_\nu(r,t)$ through a spherical surface of radius r [9], we obtain the result for the energy loss per unit mass in time interval Δt

$$\Lambda(r,t) = \frac{\Delta t}{m(\Delta r)} \int_0^\infty (S_\nu(r,t) - S_\nu(r-\Delta r, t))d\nu \tag{4}$$

where $m(\Delta r)$ is the mass in a shell of width Δr. The properties of matter enter Eqs. (3) and (4) through the attenuation coefficient κ_ν', which is composed of the absorption coefficient $\kappa_{\nu a}$ and scattering coefficient $\kappa_{\nu s}$. Scattering is usually only important in a very rarefied gas, so it can be neglected. The calculation of the absorption coefficient requires a detailed knowledge of the atomic (and molecular) structure of the gas. The contribution of bound-free (photoelectric effect) and free-free (bremsstrahlung absorption) transitions to the total absorption coefficient are in the ratio $(e^x - 1) : 1$, where $x = h\nu/k_BT$ [8]. Since there is a wide range of frequencies and temperatures, both kinds of transitions are important in absorption [10]. Finally, the energy loss given by Eq. (4) with I_ν calculated from Eq. (3) was included in the loss term on the right hand side of Eq. (1c).

b) *Bremsstrahlung (BRS) model;* The radiation cross section $d\chi/d\omega$ for BRS follows from the Born approximation (Bethe-Heitler formula, [11]). The energy radiated per unit time/frequency/volume is $dE/(d\omega dV dt) =< v\, d\chi/d\omega > n_e n_{ion}$, where n_e and n_{ion} are the concentrations of electrons and ions, and $< v\, d\chi/d\omega >$ is the thermal average of the electron velocity v and the radiation cross section. We also include corrections to individual radiative scattering events due to the rest of the plasma [12]. One finds

$$\frac{dE}{dt\ dV} = \frac{64\pi}{3}\frac{e^2 c}{m_e}\left(\frac{e^2}{m_e c^2}\right)^2 \left(\frac{\beta m_e}{2\pi}\right)^{3/2} \frac{\omega_p}{\beta} n_e n_{ion}\ \mathcal{F} \tag{5}$$

where e and m_e are the electron charge and mass, respectively, and $\beta = 1/(k_BT)$. The function $\mathcal{F}$ is given by:

$$\mathcal{F} = \frac{1}{\omega_p}\int_0^\infty d\omega \int_1^\infty dx \frac{e^{-\beta\hbar\omega x}}{\sqrt{x(x-1)}} \left|\frac{3}{2+\epsilon(\omega)}\right|^2 \tag{6}$$

and was calculated numerically. Finally, the energy loss per unit mass was calculated using Eq. (5) and included in the loss term at the right hand side of Eq. (1c).

Consider the relative importance of the various mechanisms for energy loss. While the inclusion of vibrational degrees of freedom, dissociation and ionization has some effect, the primary corrections come from radiation. The profiles in space and time of the thermodynamic variables of the gas are similar to the uncorrected solution (see Fig. 2). The most drastic changes occur in the temperature (Fig. 3), especially close to the origin. The maximum temperature is about an order of magnitude smaller than the results without corrections. Also, radiation plays an important role in transferring energy from hot regions to those with moderate temperatures. The net result is that the hot bubble center is cooled more rapidly.

The primary mechanism for producing the SL pulse is presumably bremsstrahlung, but it is not the radiation that is seen. The spectrum of BRS falls with increasing frequency opposite to the trend seen experimentally [2]. The surrounding plasma plays a crucial role in determining the spectrum. We have treated its effect in two limiting cases. The BRS model modifies the bremsstrahlung spectrum by the filtering action of an optically thin plasma shell surrounding the high temperature core. In the thermal model primary radiation never gets out. What is seen is secondary radiation stemming from processes such as radiative recombination, secondary bremsstrahlung, radiative deexcitation, etc., which get quenched as the expanding shell gets rapidly cooled.

In the thermal approach the absorption coefficient determines if a body radiates a black body spectrum (surface radiation) or whether it will be a volume radiator. It could not be assumed that the bubble content behaves as any of these two limiting cases. The bubble, in the present approach, can be described as having three separate regions with quantitatively different temperatures and qualitatively different optical properties. The first one consists of the hot bubble center. In this region the absorption coefficient is large for optical radiation which is rapidly absorbed. The medium is opaque and this radiation never leaves the bubble. The intermediate region has a sharp temperature gradient and an opacity which changes from optically thick to optically thin across a distance comparable in size to the mean free path of optical radiation. The outer layer of the bubble (close to the bubble wall) is typically at temperatures of

$10,000 - 15,000\,K$. There the gas is optically thin, so this layer is non-absorbing. The origin of SL stems from the intermediate zone where the brightness temperature is not very frequency dependent. Correspondingly, the SL radiation emitted from the bubble has spectral properties similar to black body radiation with a temperature characteristic of the intermediate zone. This could explain the fit of the observed SL pulse [2]) to the Planck spectrum with a temperature of $25,000\,K$. The actual calculations of emitted radiation involved using the results for the energy loss Eq. (4), where the limits for the frequency integration had to be modified due to the absorption of radiation in water. The length of the pulse (full width at half maximum) is less than $50\,ps$, which is consistent with experiment. The peak power is about $100\,mW$, consistent with the experimental value of $30\,mW$ (lower limit) [2]. Absorption of radiation in water and container walls was not taken into account in our results. We also noticed a sharp rise and slower decay of the pulse as mentioned in [2].

In the BRS model the absorption coefficient strongly influences the spectral energy and power output due to the radiative absorption around the plasma region. The calculations were performed using Eq. (5), corrected for the absorption in region around the plasma. The length of the pulse is longer than in the thermal approach and the peak power is about the same.

The theoretical emission spectrum is presented in Fig. 4 in comparison with experimental results from [2]. The agreement between the experimental results and the thermal approach is very good. The results of the BRS model depend on choice of cutoff m_c, but show the common characteristic of becoming saturated at angular frequencies of about $5 \times 10^{15}\,rad/s$. In summary, the "thermal model" seems to be in very good agreement with experimental results and possibly explains the origin of sonoluminescence. The "bremsstrahlung" model does not fit the experimental data as well as the thermal approach.

We conclude that the self-consistent inclusion of loss terms, especially radiation (which is neglected in the previous work in the subject), is necessary in order to correctly understand sonoluminescence. The results of our theory are very sensitive to the experimental parameters, such as the acoustic pressure amplitude and equilibrium bubble radius. A study of this parameter dependence will be presented elsewhere [13]. Aspects of out theory not yet considered involve how these parameters relate to the temperature of the liquid or the precise chemical nature of the gas.

After completing this work the paper by Moss at al. [14] was published. They include dissociation and ionization but did not allow the radiation to modify the dynamics of the gas self-consistently, as is done in the present letter.

We would like to thank Prof. S. Putterman for a helpful discussion.

---

* Also at: Institute of Astronomy and Astrophysics, Academia Sinica, P. O. Box. 1-87, Taipei, Taiwan, R. O. C.

[1] L.A. Chambers, J.Chem. Phys. **5**, 290, 1937.

[2] B.P. Barber, S.J. Putterman, Nature, **352**, 318, 1991; B.P. Barber, R. Hiller, K. Arisaka, H. Fetterman and S.J. Putterman, J. Acoust. Soc. Am **91**, 3061, 1992; R. Hiller, S.J. Putterman, B.P. Barber, Phys. Rev. Letters **69**, 1182, 1992; B.P. Barber, S.J. Putterman, Phys. Rev. Letters **69**, 3839, 1992.; B.P. Barber, C.C. Wu, R. Lofstedt, P.H. Roberts, S.J.Putterman, Phys. Rev. Lett. **72**, 1380, 1994.

[3] C.C. Wu, P.H. Roberts, Phys. Rev. Lett.**70**, 3424, 1992; C.C. Wu, P.H. Roberts, Proc. Acad. Soc. **A445**, 323, 1994.

[4] A.J. Walton, G.T. Reynolds, Adv. in Phys. **33**, 595, 1984.

[5] S.K. Godunov, Mathematicheskii Sbornik **47**, 271, 1959.

[6] G. Guderley, Luftfahrtforschung **19**, 302, 1942.

[7] The results are insensitive to the transition temperature ($4,000\ K$) or the ionization or dissociation temperatures.

[8] Ya. B. Zeldovich, "Physics of shock waves and high temperature hydrodynamic phenomena", Academic Press, New York, 1966.

[9] D. Mihalas, "Stellar atmospheres", V.H.Freeman and Company, San Francisco, 1978. The energy flux per unit area is given by $\mathbf{S}_\nu(\mathbf{r},t) = \int I_\nu(\mathbf{r},\mathbf{\Omega},t)\ \mathbf{\Omega}\ d\mathbf{\Omega}$.

[10] For T in the range of $10^5 - 10^6\,K$ and frequencies near the maximum of the Planck spectrum of $10^{16}\ Hz$, κ'_ν is typically $10^5 - 10^6\,cm^{-1}$. Thus t_p is on the order of $10^{-15}\ s$, which is much shorter than the time the bubble spends close to the minimum radius. Then κ'_ν is on the order of $10^6\,cm^{-1}$, i.e. the bubble center is optically thick.

[11] J.D. Jackson, "Classical electrodynamics", John Wiley & Sons, New York, 1975.

[12] Note that the electric dipole $\mu(\omega)$ in a small cavity with dielectric constant $\epsilon(\omega)$ emits radiation as if it had an effective dipole moment $p(\omega) = 3\mu(\omega)/[2+\epsilon(\omega)]$ (independent of its location or orientation)!

[13] L. Kondic, J.I. Gersten, C. Yuan, (to be published).

[14] W.C. Moss, D.B. Clark, J.W. White, D.A. Young, Phys. Fluids **6**, 2979, 1994.

FIG. 1. Space-time dependence of the gas pressure, temperature and velocity without loss terms. Time $t = 0$ is defined as when bubble wall velocity changes sign. Parameters are $P_a = 1.3\,atm$, $R_0 = 4.5\,\mu m$, $f_a = 26.5\,KHz$, $1/b = 0.794\,g/cm^3$, $\rho_l = 1.0\,g/cm^3$, $c_l = 1.481\,km/s$, $\nu_l = 0.01\,cm^2/s$ (for water at $20^\circ C$).

FIG. 2. Space-time dependence of the gas pressure, temperature and velocity following from the thermal approach. The parameters are the same as before. The small oscillations are artifacts of the discrete procedure for including the energy losses.

FIG. 3. Space profile of the gas temperature at the time immediately after the first shock rebounds from the origin, $P_a = 1.3\,atm$.

FIG. 4. Spectral energy of the SL radiation. Theoretical results were scaled down to experimental values. Acoustic pressure amplitude is $1.325\,atm$ both in theoretical and experimental results.