Modern Physics Letters B, Vol. 19, Nos. 28 & 29 (2005) 1711–1714

INFLUENCE OF SONOCHEMISTRY ON SINGLE-BUBBLE SONOLUMINESCENCE*

LI YUAN† and PING HE‡

LSEC and Institute of Computational Mathematics,
Academy of Mathematics and Systems Science,
Chinese Academy of Sciences, Beijing, 100080, People's Republic of China
† *lyuan@lsec.cc.ac.cn*
‡ *peace@lsec.cc.ac.cn*

Received 1 June 2005

Spherical oscillation of an acoustically levitated gas bubble in water was simulated numerically to elucidate the phenomenon of single-bubble sonoluminescence (SBSL). A refined hydro-chemical model was used, which took into account the processes of water vapor evaporation and condensation, mass diffusion, and chemical reactions. The numerical results show significant water vapor dissociations but rather low degrees of ionizations. A widely accepted weakly ionized gas model is then used to compute the light emission. Contrary to earlier predictions without chemical reactions, the present calculated light spectra are generally too small and the pulses are too short to fit to recent experimental results within stable SBSL range. To solve this contradiction, the *electrostatic interactions* of the ionized gases are included, which is shown to *lower* the ionization potentials of gas species in the bubble significantly.

Keywords: Single-bubble sonoluminescence; electrostatic interactions; lowering of ionization potential.

1. Introduction

Acoustic waves in liquids can cause cavitation. Cavitating bubbles undergo repeated cycles of growth and collapse in response to the acoustic waves. Under certain conditions, a single bubble trapped at the pressure antinode of an acoustic wave can emit a brief flash of light during the violent collapse of the bubble. This phenomenon is known as single-bubble sonoluminescence[1,2]. SBSL has fascinated scientists from various fields. Intriguing and extreme conditions were observed[2,3]. Many models were proposed to explain the mechanism of the light emission. The model of thermal bremsstrahlung and recombination radiation from an optically thin bubble was most successful as it predicted the widths, shapes and spectra of the emitted light fairly well under certain hydrodynamic frameworks[4-7]. However, chemical reactions were ignored in these predictions, whose influence was found to reduce the temperature in the bubble significantly[8]. In this paper,

* This work is supported by National Natural Science Foundation of China (G10172089, G10476032) and State Key Program for Basic Research (G1999032801).

we use a refined hydrodynamic model that accounts for the chemical reactions of water vapor mixture in a bubble irradiated by acoustic waves (related to *sonochemistry*). The electrostatic (Coulomb) interactions inside the bubble are considered. Numerical simulations indicated that the interactions cause significant increase in ion percentages. The light emissions computed with and without considering the Coulomb interactions are compared with a calibrated experiment[9].

2. Model

The bubble is assumed to be spherically symmetric and is composed of mixture of noble gas, water vapor and reaction products. The equations to be solved are the Navier-Stokes (NS) equations coupled with the Rayleigh-Plesset (RP) equation for the bubble radius $R(t)$, the water temperature equation and the mass concentration equation of the dissolved noble gas in the surrounding water. Detailed formulations are given in Refs.10 and 11. A brief list of the equations is as follows. The compressible NS equations in the spherical coordinates are:

$$\frac{\partial \mathbf{Q}}{\partial t}+\frac{\partial \mathbf{F}}{\partial r}=\mathbf{H}+\frac{1}{r^2}\frac{\partial r^2\mathbf{F}_v}{\partial r}+\mathbf{M}_v+\mathbf{S}\,, \tag{1}$$

where S is the chemical source term. The weakly compressible liquid flow outside the spherical bubble is accounted for by the RP equation[12]:

$$(1-M)R\ddot{R}+\frac{3}{2}(1-\frac{1}{3}M)\dot{R}^2=(1+M)[H_b-\frac{1}{\rho_{l\infty}}P_s(t+\frac{R}{C_{l\infty}})]+\frac{R}{C_{lb}}\dot{H}_b\,. \tag{2}$$

The equations for the water temperature T and for the mass concentration of dissolved noble gas c take similar forms:

$$\frac{\partial T_l}{\partial t}+u_l\frac{\partial T_l}{\partial r}=\frac{\lambda_l}{\rho_l C_{P_l}}\frac{1}{r^2}\frac{\partial}{\partial r}(r^2\frac{\partial T_l}{\partial r})\,, \tag{3}$$

$$\frac{\partial c}{\partial t}+u_l\frac{\partial c}{\partial r}=D_l\frac{1}{r^2}\frac{\partial}{\partial r}(r^2\frac{\partial c}{\partial r})\,. \tag{4}$$

Eqs. (1)-(4) constitute the hydrodynamic-chemical model, whose numerical solutions are sought for by time marching. Since the light energy is very small compared with the kinetic energy, the light emission is postprocessed. This is done by using the weakly ionized gas model of Hilgenfeldt *et al*[4]. This model accounts for the absorptions due to the free-free interaction of electrons and ions, free-free interactions of electrons and neutral atoms, and bound-free ionization of already excited atoms[4,13]. The bound-bound absorption is ignored in this paper.

The well-known Coulomb interactions exist for charged systems. A sonoluminescing bubble is thought to contain trace amounts of plasma. Therefore, it is meaningful to see how large the effect of the interactions is for such constituents. According to the Debye-Hückel theory[14], the electrostatic free energy F_{Coub} is

$$F_{Coub} = -\frac{2e^3}{3}\left(\frac{\pi}{k_B TV}\right)^{1/2}\left(\sum N_j z_j^2\right)^{3/2}. \tag{5}$$

The total free energy of the system is obtained by adding F_{Coub} to the ideal gas free energy. From the total free energy one can obtain a modified Saha equation:

$$\frac{N_{j+1}N_e}{N_j} = \frac{Q_{j+1}Q_e}{Q_j}\exp\left(\frac{\Delta I_{j+1}}{k_B T}\right) \propto \exp\left(-\frac{I_{j+1} - \Delta I_{j+1}}{k_B T}\right). \tag{6}$$

where $\Delta I_{j+1} = 2(j+1)e^3(\pi/k_B T)^{1/2}(n_e + \sum i^2 n_i)^{1/2}$ describes a decrease in the ionization potential due to the Coulomb interactions.

3. Numerical Results

The model has following controllable parameters: the driving pressure amplitude P_a and frequency f, the water temperature T_∞, and the gas concentration dissolved in the water c_∞. The ambient bubble radius R_0 depends on above parameters. For comparison, we use the same set of parameters as in the experiment[9]: R_0=4.5 μm (He) or 5.5 μm (Xe), f=42 kHz, T_∞=296.15 K, P_{diss}=150 Torr (He) or 3 Torr (Xe), while is P_a adjustable.

Figure 1 shows one snapshot of the number density distributions and the degrees of ionization. In Fig.1(a), it can be seen that the number densities of chemical products are considerable. In Fig. 1(b) it is seen that the degrees of ionization for cases with the Coulomb interactions are much larger than those without the Coulomb interactions. A threshold is set to prevent the computed ionization potential from becoming negative, which is reflected in the sudden levelling of the curves in the inner zone for cases with the Coulomb interactions.

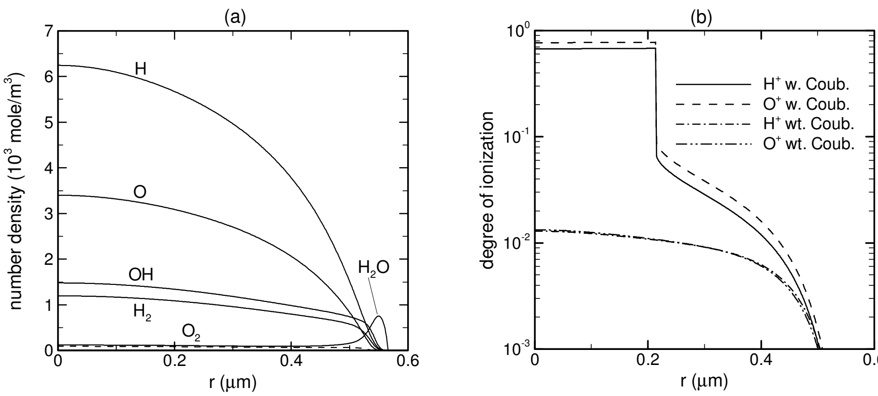

Fig. 1. The spatial profiles of number densities for molecular species (a) and degrees of ionizations (b) at the time of minimum bubble radius t=0($t_{\min}$=14.936791μs) for He bubble at P_a=1.45atm, R_0=4.5μm. The degree of ionization is computed using the Saha equation.

Figure 2 shows the time history of the effective ionization potential of atomic species. Note that the ionization potentials are reduced significantly around t=0. Figure 3 shows comparison of the spectral radiances. It is seen that calculated spectra with the Coulomb interactions are much larger than those without them, and the experimental data[9] are better fitted using driving pressure within stable SBSL range when the Coulomb interactions are taken into account.

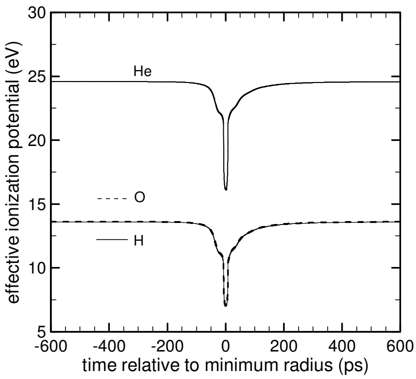

Fig. 2. Time history for the effective ionization potentials of atomic species in the He bubble. Each ionization potential is averaged over the whole bubble. P_a=1.45 atm, R_0=4.5 µm. t=0 ps corresponds to the time of minimum radius t_{min}=14.836791 µs.

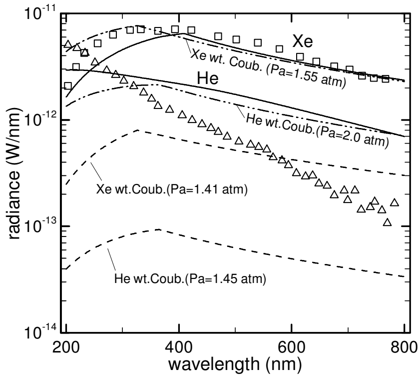

Fig. 3. Spectral radiance of the SL light from bubbles of Xe and He in water. The squares and triangles are experimental spectra of Xe and He bubbles. The solid lines are fittings with the Coulomb interactions at P_a =1.41 atm (Xe), 1.45 atm (He), the dashed lines are fittings without the Coulomb interactions, and the dash-dotted lines are for P_a above SBSL range.

References

1. B. Barber, R. Hiller, R. Löfstedt, S. Putterman, and K. Weninger, *Phys. Rep*. **281**, 65 (1997).
2. M. Brenner, S. Hilgenfeldt, and D. Lohse, Rev. Mod. Phys. **74**(2), 425 (2002).
3. R. Taleyarkhan *et al.*, *Science* **295**, 1868 (2002).
4. S. Hilgenfeldt, S. Grossmann, and D. Lohse, *Phys. Fluids* **11**, 1318 (1999).
5. W. Moss *et al.*, *Phys. Rev.* **E 59**, 2986 (1999).
6. D. Hammer and L. Frommhold, *J. Mod. Opt.* **48**, 239 (2001).
7. P. Burnnet et al., J. Phys. B: At. Mol. Opt. Phys. **34**(16), L511 (2001).
8. B. Storey and A. Szeri, *Proc. Roy. Soc. London A* **456**, 1685 (2000).
9. G. Vazquez, C. Camara, S. Putterman, and K. Weninger, *Opt. Lett.* **26**, 575 (2001).
10. C. Ho, L. Yuan, M. Chu, P. Leung, and W. Wei, *Phys. Rev.* **E 65**, 041201 (2002).
11. L. Yuan, *Phys.Rev*. **E72**, 046309 (2005).
12. A. Prosperetti and A. Lezzi, *J. Fluid Mech*. **168**, 457 (1986).
13. Y. Zeldovich and Y. Raizer, Vol. I, ed. W. D. Hayes and R. F. Probstein (Academic Press, New York, 1966).
14. S. Eliezer, A. Ghatak and E. Teller, *An Intorduction to Equations of State: Theory and Applications* (Cambridge University Press, Cambridge, 1986).