Modelling single-bubble sonoluminescence with chemical reactions and Coulomb interactions*

Ping He, Li Yuan
Institute of Computational Mathematics and LSEC
Academy of Mathematics and Systems Science
Chinese Academy of Sciences, Beijing 100080, China

session: cavitation field and sonoluminescence

September 6, 2005

Abstract

A refined hydro-chemical model was applied to numerical simulation of single-bubble sonoluminescence (SBSL) by taking into account the processes of water vapor evaporation and condensation, heat and mass diffusion, and chemical reactions. The numerical results show significant water vapor dissociations but rather low degrees of ionization. A widely accepted weakly ionized gas model is then used to compute the light emission. Contrary to earlier predictions without chemical reactions, the calculated light spectra and pulses are generally too small to fit to experimental data within stable SBSL range. To alleviate this contradiction, the *electrostatic interactions* of the ionized gas particles are taken into account, which are shown to *lower* the ionization potentials of gas species in the bubble significantly, hence resulting in dramatic increase of ion populations.

Key words: single-bubble sonoluminescence; Coulomb 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, a phenomenon 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, 5, 6, 7]. However, chemical reactions were ignored in these predictions, whose influence was found to reduce the temperature in the bubble significantly [8]. A modification to the reaction rates was found to suppress vapor dissociations and

*Presented at World Congress on Ultrasonics/Ultrasonics International 05, Aug.29-Sept.1, Beijing, China.

raise the bubble temperature to such an extent that the above light emission model recovered its plausibility [9]. Yet a very recent study using a spatially uniform pressure model with chemical reactions predicted that the popular thermal bremsstrahlung and recombination radiation mechanisms gave insufficient light intensities compared with experimental observations [10]. Other researchers found that thermal bremsstrahlung from a very small emitting interior could fit to the experimental spectra more robustly [11].

In this paper, we use a refined full hydrodynamic model that accounts for the chemical reactions of water vapor mixture. As an initial step, the electrostatic (Coulomb) interactions inside the bubble are considered in a posteriori way. Numerical simulations indicated that the Coulomb interactions reduce ionization potentials considerably, resulting in significant increase in ion percentage. This result is quite different from remarked in [10]. The light spectra computed with and without the Coulomb interactions are compared with a calibrated experiment [12].

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, the water temperature equation and the mass concentration equation of the dissolved noble gas in the surrounding water. Detailed formulations are given in Ref.[13, 14]. A brief list of the equations is as follows.

The compressible NS equations are written in the spherical coordinates:

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

where $\mathbf{S}$ is the chemical source terms. The weakly compressible liquid flow outside the spherical bubble is accounted for by a form of the RP equation that includes first order terms in the Mach number $M = \dot{R}/C_{lb}$ and allows for variable speed of sound in the water [15]:

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

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

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

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

Eqs.(1)-(4), together with appropriate boundary conditions, constitute the hydrodynamic-chemical model, whose numerical solutions are sought for by time marching. The term $\mathbf{S}$ is computed by chemical kinetics, where only a subset consisting of 8 elementary reactions is used, corresponding to the first eight ones used by Yasui [16]. The first 19 reactions of [16] with additional species (HO_2, H_2O_2) were also tried but the resulting temperature was found only a little lower than that from the 8 reaction scheme. Nonequilibrium ionizations are not taken into account. Notice that the modification to the chemical equilibrium constant for a van der Waals gas as suggested by Toegel *et al.* [9] is also used in this study, see detail in [14].

Since the light energy is very small compared to the kinetic energy, the light emission is post-processed. This is done by using the light emission model of Hilgenfeldt *et al.* [4]. This model

accounts for the absorptions due to the free-free interaction of electron and ions, free-free interactions of electrons and neutral atoms, and bound-free ionization of already excited atoms [4, 17]. The bound-bound absorption is ignored in this study.

The well-known Coulomb interactions exist for charged systems. A sonoluminescing bubble is thought to contain trace amounts of plasma, probably at liquid-like densities. Under such conditions, the effective ionization potentials tend to be lowered [6]. Therefore, it is meaningful to see how large the effect of the Coulomb interactions is for such constituents. According to the Debye-Hückel theory [18], the electrostatic free energy $F_{\rm Coub}$ is

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

The total free energy of the system is obtained by adding $F_{\rm Coub}$ 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 $\Delta I_{j+1} = 2(j+1)e^3(\pi/k_B T)^{1/2}(n_e + \sum_i i^2 n_i)^{1/2}$ describes a decrease in the ionization potential due to the Coulomb interactions. We remark that the modified Saha equation (6) is used in a posteriori way from known number densities of atoms and other chemical radicals as calculated from the hydrodynamic-chemical solutions, whose feedback influence on the equation of state, hence on the hydrodynamics, is not considered for the time being.

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 nearly the same set of parameters as in the experiment [12]: $R_0 = 4.5$ μm (He) or 5.5 μm (Xe), $f = 42$ kHz, $T_\infty = 296.15$ K, $P_{\rm dissolve} = 150$ torr (He) or 3 torr (Xe), but P_a is adjustable.

Fig. 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 in considerable amounts. In Fig. 1(b) it is seen that the degrees of ionization for the case with the Coulomb interactions are much larger than those without the Coulomb interactions. A threshold is set to prevent the ionization potential from reducing to negative, which is reflected in the sudden levelling of the curves in the inner zone for cases with the Coulomb interactions. The reason that the ionization potential tends to zero is probably due to lack of a feedback mechanism to the hydrodynamics, as remarked in the end of Section 2. This will be modified in future work.

Fig. 2 shows the time history of the effective ionization potential of atomic species. Note that the ionization potentials are reduced significantly around $t = 0$. Fig. 3 shows comparison of the spectral radiances. It is seen that calculated spectrum intensities with the Coulomb interactions are closer to the experimental data [12] under the same driving pressure within stable SBSL range than without the Coulomb interactions. However, the long-wavelength light intensities are still larger than the experimental data for He bubble. Similar discrepancy also existed for earlier models [6].

4 Conclusions

The full hydrodynamic simulation of single-bubble sonoluminescence indicates that the light intensities are too small to fit to experimental data within stable SBSL range. This result is in agreement with recent one [10]. Furthermore, we studied the Coulomb interactions of the ionized particles. We show that the ionization potentials of gas species are reduced significantly during a brief stage of the bubble collapse, which leads to dramatic increase of ion percentage. This new result awaits confirmation from further studies.

Acknowledgements

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

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] Toegel R., Hilgenfeldt S., and Lohse D., Phys. Rev. Lett. **88**, 034301 (2002).

[10] C. Ying, Y An and C.Xie, J. Phys. D: Apply. Phys. **38**, 2489-2496 (2005).

[11] C. Camara, S. Putterman, and E. Kirilov, Phys. Rev. Letts. **92**, 124301 (2004).

[12] G. Vazquez, C. Camara, S. Putterman, and K. Weninger, Opt. Lett. **26**, 575 (2001).

[13] C. Ho, L. Yuan, M. Chu, P. Leung, and W. Wei, Phys. Rev. E **65**, 041201 (2002).

[14] L. Yuan, "Sonochemical effects on single-bubble sonoluminescence ", phyics/0501041.

[15] A. Prosperetti and A. Lezzi, J. Fluid Mech. **168**, 457 (1986).

[16] Yasui K., Phys. Rev. E **56**, 6750 (1997).

[17] Y. Zeldovich and Y. Raizer, *Physics of shock waves and high-temperature hydrodynamic phenomena*, Vol. I, ed. W. D. Hayes and R. F. Probstein (Academic Press, New York, 1966).

[18] S. Eliezer, A. Ghatak and E. Teller, *An Intorduction to Equations of State: Theory and Applications* (Cambridge University Press, Cambridge, 1986).

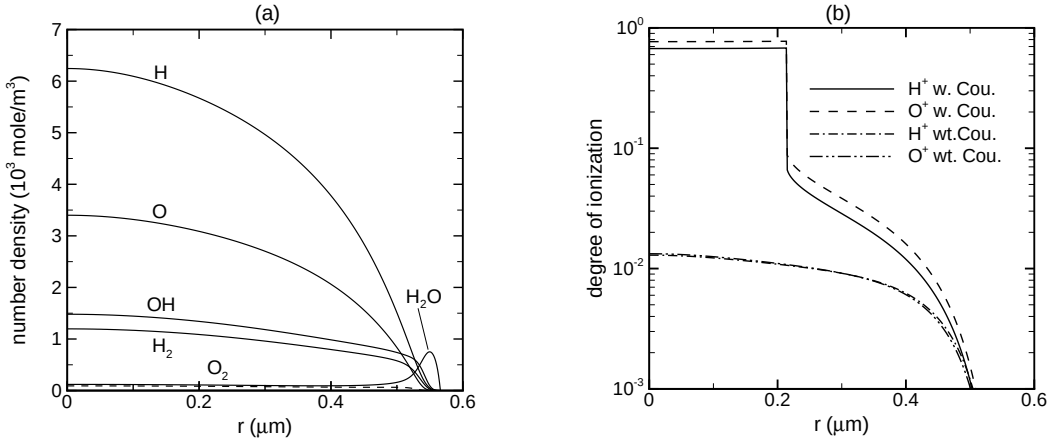

Figure 1: The spatial profiles of number densities for molecular species (a) and degrees of ionizations (b) at the time of minimum bubble radius $t = 0$ ($t_{\min} = 14.836791$ μs) for He bubble at $P_a = 1.45$ atm, $R_0 = 4.5$ μm. The degree of ionization is computed using the Saha equation.

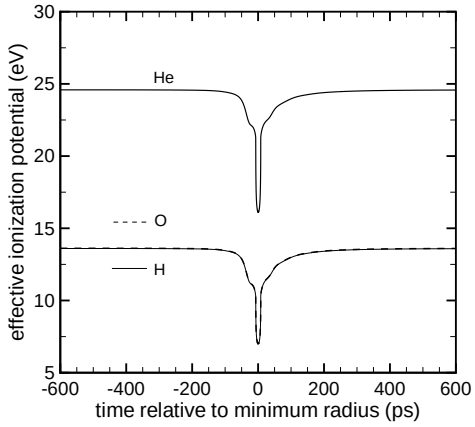

Figure 2: Time history for the effective ionization potentials of atomic species in He bubble. Each ionization potential is averaged over the whole bubble. $P_a = 1.45$ atm, $R_0 = 4.5$ μm. $t = 0$ ps corresponds to the time of minimum radius ($t_{\min} = 14.836791$ μs).

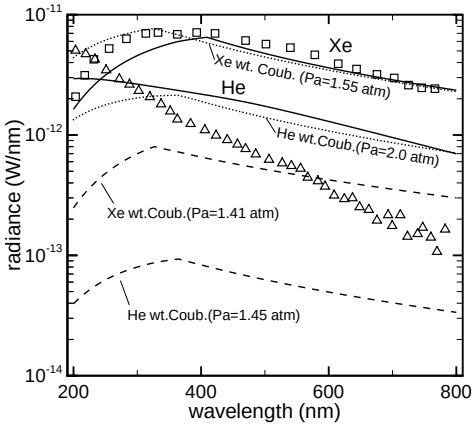

Figure 3: Spectral radiance of the SL light from bubbles of Xe and He in water. The squares and triangles are experimental spectra of Xe and He bubbles [12]. The solid lines are fittings with the Coulomb interactions at $P_a = 1.41$ atm (Xe), 1.45 atm (He), the dashed are without them for the same P_a, and the dotted lines are without them for P_a out of stable SBSL range.