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Computational Simulation of Sodium Doublet Line Intensities in Multibubble Sonoluminescence *

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We perform a computational simulation of the fluid dynamics of sodium doublet (Na-D) line emissions from one sonoluminescing bubble among the cavitation bubbles in argon-saturated Na hydroxide (NaOH) aqueous solutions. Our simulation includes the distributions of acoustic pressures and the dynamics of cavitation bubbles by numerically solving the cavitation dynamic equation and bubble-pulsation equation. The simulation results demonstrate that when the maximum temperature inside a luminescing bubble is relatively low, two emission peaks from excited Na are prominent within the emission spectra, at wavelengths of 589.0 and 589.6 nm. As the maximum temperature of the bubble increases, the two peaks merge into one peak and the full width at half maximum of this peak increases. These calculations match with the observations of Na-D line emissions from MBSL occurring in aqueous solutions of NaOH under an argon gas.

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Multi-bubble sonoluminescence (MBSL) is a phenomenon in which light is emitted during the intense collapse of clouds of bubbles in liquids irradiated by high-intensity ultrasound.[1] It is generally realized that MBSL results from the extremely high temperature and pressure that occurs during the intense compressional heating of gas and vapor in a collapsing bubble, which is similar to that of single bubble sonoluminescence (SBSL).[2] To date, however, there is no equipment available for directly measuring the temperature and pressure inside sonoluminescing bubbles experimentally because the bubble is only several microns in diameter when it luminesces.

Spectra of MBSL and SBSL are generally used as a probe to estimate the temperature and pressure in these micro-bubbles. In 1999, Hilgenfeldt *et al.* proposed a simple model to quantitatively interpret these spectral data for SBSL. They assumed that the temperature and pressure inside the bubble were uniform and vapor effects were neglected.[3] In 2001, Yasui performed computer simulations of SBSL by considering the vapor effect inside a sonoluminescing bubble.[4] He demonstrated that the temperature at the collapse of the bubble was higher in a colder liquid because a smaller amount of water vapor was trapped inside a bubble at the point of collapse due to the lower-saturated vapor pressure. This demonstrated that the vapor pressure can prevent further increases in temperature inside a bubble. In 2008, An and Li improved upon this analysis utilizing computational fluid dynamics (CFD) simulations that included appropriate moving boundary conditions and vapor effects.[5] Their calculations suggested a possible mechanism of how the Na emission peak near 589 nm changes as the total radiation power increases. In 2009, Xu *et al.* investigated the line emissions from non-volatile species during MBSL.[6] They observed the spatial separation of MBSL in 0.1 M Na_2SO_4 solution in 95 wt% H_2SO_4 saturated with argon (Ar), and 0.1 M Na_3PO_4 solution in 85 wt% phosphoric acid. The excited Na atom (Na^*) emission cannot be detected at the top of the bubble cloud (near the horn tip), and the emission was strictly a broad continuum. Strong Na^* emission near the wavelength of 589 nm on a continuum was observed at the bottom of the bubble cloud. Recently, Zhao *et al.* experimentally measured Na doublet (Na-D) lines from MBSL in Na hydroxide (NaOH) aqueous solution saturated with Ar.[7] However, there is no theoretical computational simulation available to demonstrate the variation in the intensity of Na-D lines during MBSL as a function of acoustic pressure.

MBSL is caused by the intense collapse of cavitation bubbles, which is similar to the SBSL. However, the variation in the radius of the bubble with time exhibits higher fluctuation in MBSL compared to SBSL because of a complicated sound field arising from interactions among bubbles. The effect of these interactions among bubbles on MBSL spectra remains unknown. Therefore, in this work, we further develop the theoretical model of SBSL to explore the mechanism of Na-D lines emission in MBSL, using some reasonable approximations of MBSL phenomena.

The theoretical model employed herein includes the equation of cavitation bubble dynamics,[8] the equation of acoustic waves in a cavitation bubble cloud[9] and the spectral radiation formula of a single sonoluminescing bubble, which is similar to the model previously reported.[10]

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To simplify the dynamics of one bubble among a cloud of bubbles, we assume that the number density (N) of the bubble with radius R remains unchanged and we only consider one kind of bubbles, with an ambient radius R_0. Ignoring the flow of the bubble, R reads[8]

$$(1-M)R\ddot{R}+\frac{3}{2}\Big(1-\frac{M}{3}\Big)\dot{R}^2 = (1+M)\frac{1}{\rho_{\rm l}}[p_{\rm l}-p]+\frac{t_{\rm R}}{\rho_{\rm l}}\dot{p}_{\rm l}, \quad (1)$$

where $M = R/c_{\rm l}$ is the bubble-wall Mach number, R is the radius of a cavitating bubble, $\rho_{\rm l}$ is the liquid density, p determined by Eq. (2) is the pressure at the location of the bubble in liquid, $p_{\rm l} = p_{\rm g}(R,t) - 4\eta\dot{R}/R - 2\sigma/R$ is the pressure on the liquid side of bubble wall, $p_{\rm g}(R,t)$ is the pressure on the gas side of the bubble wall, η is the shear viscosity of liquid, σ the surface tension coefficient of the liquid, and $t_{\rm R} \equiv R/c_{\rm l}$. When p is computed by Eqs. (1) and (2), the bubble is assumed in an isothermal approximation, $p_{\rm g}(R,t) = \mu\mathcal{R}T/(V-b)+p_{\rm v}$, where μ is the gas mole number, $\mathcal{R}$ is the gas constant, T is the temperature of the surround liquid, V is the bubble volume, b is the van der Waals hard core volume, and $p_{\rm v}$ is the vapor pressure of the liquid. When the spectrum from a bubble is computed, the isothermal approximation of the bubble is invalid.

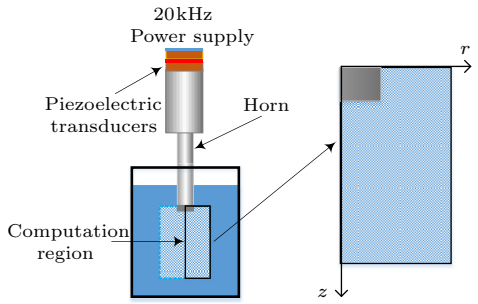


Fig. 1. Sketch of the experiment for the MBSL: (a) stereogram and (b) axisymmetric view of computation region.

To determine the pressure at the location of the bubble in liquid and to simplify the calculation, we take a mean-field approximation to yield the acoustical wave equation for a cavitation cloud,[9]

$$\nabla^2 p - \frac{1}{c_{\rm l}^2}\frac{\partial^2 p}{\partial t^2} = -4\pi\rho_{\rm l}N(2R\dot{R}^2+R^2\ddot{R}), \quad (2)$$

where $c_{\rm l}$ is the speed of sound in the liquid at ambient temperature and pressure. In this work, because the horn tip (i.e., the ultrasonic source) is cylindrical (see Fig. 1(a)), we use an axisymmetric approximation to simplify the calculation. A homogenous distribution of bubbles is assumed in the liquid, which forms a pressure field in the r–z plane, as shown in Fig. 1(b). From Eqs. (1) and (2), we can obtain the pressure distribution in the r–z plane by numerical calculation. The pressure driving the bubble at a location in liquid is derived approximatively.

To simulate the Na-D line emission spectra from MBSL, we employ the theoretical model described in Ref. [10]. In this model, the boundary at the moving bubble wall is still described by Eq. (1). However, the driving bubble pressure is the pressure at the location of the bubble that is obtained by numerically solving Eq. (2), based on Eq. (1).

The function for radiation power is taken from Ref. [10],

$$P(\lambda,t) = 8\pi^2\int_0^R\int_{-1}^1 \kappa_\lambda(r)P_\lambda^{\rm PI}(r) \cdot \exp\Big(-\int_{r_x}^{\sqrt{R^2-r^2(1-x^2)}}\kappa_\lambda ds\Big)r^2 dr dx, \quad (3)$$

which describes the total power emitted from the bubble for each acoustic cycle at wavelength λ, κ_λ is the absorption coefficient, $P_\lambda^{\rm PI}$ is the Planck radiation intensity, and r is the radial distance from the center of the bubble.

For the line emission spectra

$$\kappa_\lambda = \frac{\lambda^5}{2hc^2}(e^{hc/\lambda kT}-1)P_\lambda, \quad (4)$$

where P_λ denotes the radiation power per unit volume per unit wavelength interval of the line spectrum. Herein, we mainly consider the $3P_{3/2} \to 3S_{1/2}$ and $3P_{1/2} \to 3S_{1/2}$ transitions of Na.[11]

For line emissions from a Na atom, we assume that thermal excitation dominates the formation of excited Na atom (Na*) during an individual luminescing bubble among MBSL bubbles. For the transition of Na*, the radiation power per unit volume may be calculated by

$$P_{i,j} = \frac{n_{\rm a}g_i e^{-h\nu_{i,j}/kT}}{\sum_k g_k e^{E_k/kT}}A_{i,j}h\nu_{i,j}, \quad (5)$$

where $n_{\rm a}$ is the number density of Na atoms inside the bubble, $A_{i,j}$ is the transition probability, $h\nu_{i,j}$ is the photoenergy, and g_i is the Lande factor of the ith energy level E_i.

The distribution of the intensities of line spectra emitted by MBSL is assumed to be even. The radiation power from Na* per unit volume per wavelength interval is given by

$$P_\lambda d\lambda = P_{i,j}g_{i,j}\Big(\nu=\frac{c}{\lambda}\Big)\frac{c}{\lambda^2}d\lambda, \quad (6)$$

where ν is the emission frequency, and $g_{i,j}$ is the following Lorentzian function

$$g_{i,j}(\nu) = \pi\frac{\Delta\nu/2}{(\nu-\nu_{i,j})^2+(\Delta\nu/2)^2}. \quad (7)$$

For collision and resonance broadening[12]

$$\Delta\nu = \frac{\sigma_0\nu_0 n}{\pi}+50\frac{n_{\rm a}f_{\rm a}}{\nu_{i,j}}, \quad (8)$$

where σ_0 is the collision cross section, ν_0 is the average relative speed of molecules in the gas, n is the

number of particles, and f_a is the absorption oscillator strength. We determine the cross sections for collision from the diameters of the gas molecules.

The total radiation power is the integral over the relevant wavelengths λ. To calculate the detail of Na-D line emissions, we reduce the wavelength interval of the integral

$$P(t) = \int_{584\,\mathrm{nm}}^{596\,\mathrm{nm}} P(\lambda, t) d\lambda. \tag{9}$$

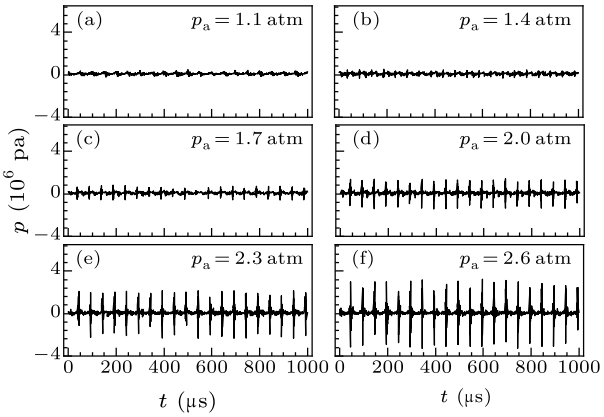


Fig. 2. Variations in acoustic pressures versus time t at location ($r = 0$, $z = 4$ mm) in the sound field (a) with a driving $p_a = 1.1$ atm, (b) with a driving $p_a = 1.4$ atm, (c) with a driving $p_a = 1.7$ atm, (d) with a driving $p_a = 2.0$ atm, (e) with a driving $p_a = 2.3$ atm, and (f) with a driving $p_a = 2.6$ atm.

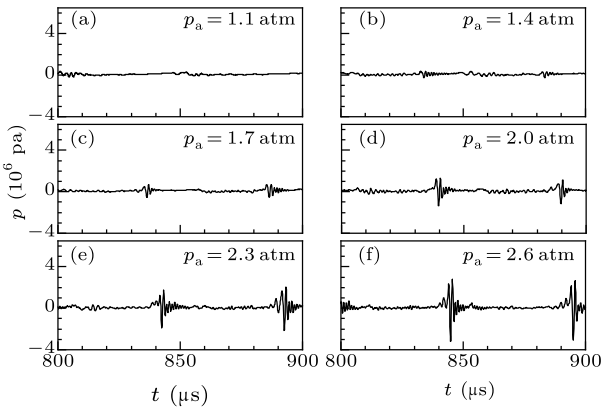


Fig. 3. Variations in acoustic pressures in a period of 800 to 950 μs at location ($r = 0$, $z = 4$ mm) in the sound field (a) with a driving $p_a = 1.1$ atm, (b) with a driving $p_a = 1.4$ atm, (c) with a driving $p_a = 1.7$ atm, (d) with a driving $p_a = 2.0$ atm, (e) with a driving $p_a = 2.3$ atm, and (f) with a driving $p_a = 2.6$ atm.

We also evaluate the cumulative radiation energy emitted from an MBSL bubble over the time 0 to t within each acoustic period

$$E_\lambda(t) = \int_0^t P(\lambda, t')dt', \quad 0 \le t \le \frac{2\pi}{\omega}. \tag{10}$$

Because MBSL occurs for only a few hundreds of picoseconds, or even less, in each acoustic cycle, only in the time interval, short duration does $P(\lambda, t)$ not vanish.

To obtain the distribution of the sound field in the cavitation cloud and to reduce the time of computation, Eq. (2) is solved numerically to exploit the axi-symmetry of the setup (see Fig. 1(b)), that is, p is as functions of radial distance r and height z. In addition, we consider only one bubble type, that is, $R_0 = 4.5\,\mu$m. The number density of bubbles is homogenous in liquid, that is, $N = 3.0\,\mathrm{mm}^{-3}$. Some parameters are derived from experimental data. The height of the liquid is 8 cm, the diameter of the horn tip is 1.3 cm and the horn is inserted into the host liquid. The region of calculation is set to be 1.2 cm at the r azimuth, and 1.2 cm at the z azimuth, because the cavitation region is about 1 cm near the horn. In fact, Eq. (2) is from three dimensions to two dimensions.

Figure 2 presents the evolution of pressure at the location $(r, z) = (0, 4\,\mathrm{mm})$ with time for six different acoustic pressures. This location is defined by the horizontal alignment of the slit of the spectrometer and the center of the flat quartz window in the cell.[7] The pressure is periodically pulsed with an ultrasound frequency (20 kHz) (Fig. 2). The peak of each pulse corresponds to the minimum radii of collapsing bubbles, which demonstrates that the same kind of bubbles are in synchronous resonance near the horn tip (about 1 cm).

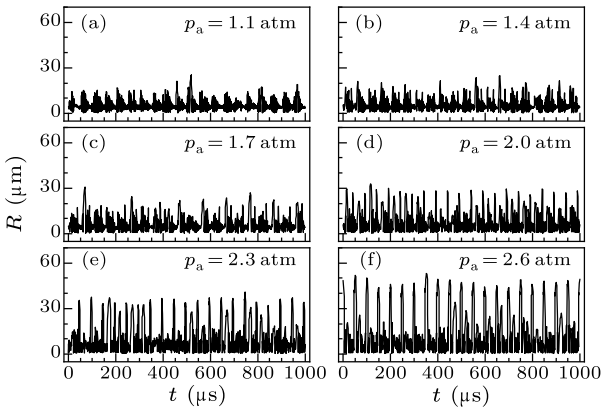


Fig. 4. Evolutions of bubble radius with time t at location ($r = 0$, $z = 4$ mm) in the sound field (a) with a driving $p_a = 1.1$ atm, (b) with a driving $p_a = 1.4$ atm, (c) with a driving $p_a = 1.7$ atm, (d) with a driving $p_a = 2.0$ atm, (e) with a driving $p_a = 2.3$ atm, and (f) with a driving $p_a = 2.6$ atm.

Because Eqs. (1) and (2) include information about a bubble's radius, we can calculate the evolution of bubble radius with time (Fig. 4). Figure 4 shows that the period of bubble radius in MBSL is less than that in SBSL, even though a bubble expands, intensely collapses, and bounces, similar to an SBSL bubble. This bubble pulsation is strongly non-periodic because of the strong non-linear interactions, which generates a complicated sound field, between bubbles and the acoustic field. This is exhibited in the fluctuations of the pulsation of bubbles.

Previous studies have collected the MBSL spec-

tra from NaOH aqueous solutions with different driving acoustic pressures.[7] These studies demonstrate that as the driving power increases, the full width at half maximum (FWHM) of the Na-D lines increases. To obtain an approximate quantitative prediction of the peaks in the MBSL emission spectrum, we assume that the Na-D lines originate from Na* within the gas bubble, and simulate the Ar bubble in NaOH aqueous solutions for six values of driving acoustic pressure.

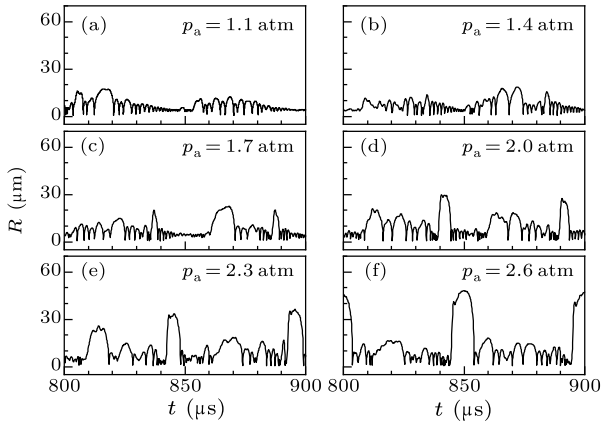


Fig. 5. Evolutions of bubble radius in a period of 800 to 950 μs at location ($r = 0$, $z = 4$ mm) in the sound field (a) with a driving $p_a = 1.1$ atm, (b) with a driving $p_a = 1.4$ atm, (c) with a driving $p_a = 1.7$ atm, (d) by driving $p_a = 2.0$ atm, (e) with a driving $p_a = 2.3$ atm, and (f) with a driving $p_a = 2.6$ atm.

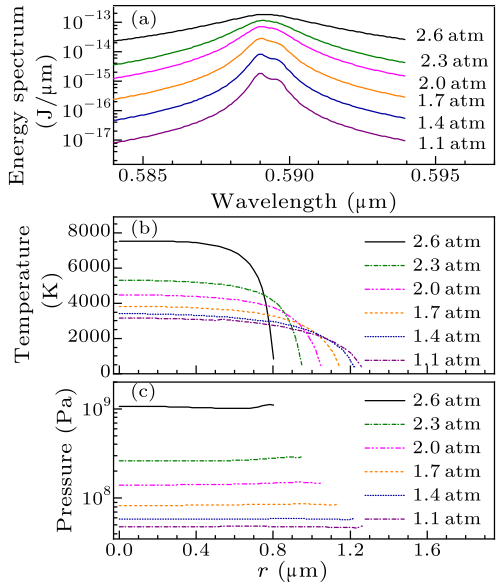


Fig. 6. Simulation results of a sonoluminescing Ar bubble at the location ($r = 0$, $z = 4$ mm) during the 17th acoustic cycle (800–850 μs) shown in Fig. 5 for different amplitudes (p_a) of the driving acoustic pressure: (a) energy spectra, (b) temperature when the bubble reaches its minimum size, (c) corresponding temperature.

We calculate the spectra of an Ar bubble at location ($r = 0$, $z = 4$ mm) in the sound field (Fig. 6). The Na-D spectral line intensities in the regions of 0.584–0.594 μm are low at the smallest amplitude of acoustic pressure, $p_a = 0.11$ MPa; the peaks of Na-D lines at wavelengths of approximately 0.5890 and 0.5896 μm are distinct (Fig. 6(a)). As the driving pressure increases, the two peaks of the Na-D lines merge into a single peak, and the FWHM of this peak increases.

Even though an exact comparison between the simulation results in Fig. 6(a) and the observed data is not possible, the simulation results follow a similar trend to that of previously published experimental data.[7] Therefore, we judge that our simulation models the MBSL system reasonably well. Figures 6(b) and 6(c) plot the distributions of temperature and pressure inside a bubble at its minimum radius. However, the core temperature remains below approximately 3829 K (corresponding to a pressure of approximately 8.77×10^7 Pa), the two peaks of the Na-D lines are observed. As the temperature increases, the two peaks of the Na-D lines finally merge into a single peak, and the FWHM of this peak increases. This trend agrees with the experimental data.

In summary, we have obtained the pressure distribution in a NaOH aqueous solution containing cavitation bubbles, and the dynamics of an individual gas bubble, by numerically solving the nonlinear acoustic wave equation with the KM equation. We also calculate the spectra of an individual bubble in a cavitation bubble cloud using the spectral formula. The results show that as the amplitude of acoustic pressure (p_a) increases, the intensities of emission increase, the two peaks of the Na-D lines will finally merge into one peak, and the FWHM of this peak increases. This trend agrees with the experimental data. It should be noted that certain parameters in our calculation, such as the number density of bubbles (N), and the number density of Na atom, are assumed, rather than being derived from experimental measurement. This may lead to differences between the theoretical results and experimental data.

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