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Temperature and Pressure inside Sonoluminescencing Bubbles Based on Asymmetric Overlapping Sodium Doublet *

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We experimentally measure the sodium D-lines from the multibubble sonoluminescence in sodium hydroxide aqueous solution. The asymmetric overlapping D-lines are successfully decomposed based on the Fourier transform analysis. The line broadening of the decomposed sodium D-lines shows the effective temperature of 3600–4500 K and the pressure of 560–1000 atm during sonoluminescence.

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Sonoluminescence was first discovered at the University of Cologne in 1934 by Frenzel and Schultes. This phenomenon is referred to as multi-bubble sonoluminescence (MBSL) to differentiate it from the single bubble sonoluminescence (SBSL). After the discovery of MBSL, researchers tried to understand the mechanism of sonoluminescence, but no substantial progress has been achieved in the dynamics of the cavitation bubbles because of a large number of short-lived interacting bubbles during MBSL. In this circumstance, spectroscopic diagnosis is used as an effective approach to study the extreme conditions inside the sonoluminescencing bubbles based on the detection of the spectrum. Kappus *et al.*[1] fitted the continuous spectra and obtained ∼8000 K SL temperature in a xenon bubble in water on the basis of ideal Planck black-body assumption. Flannigan and Suslick obtained the temperature of ∼15200 K and the pressure of ∼1500 atm by fitting the atomic and ionic characteristic spectra in an argon bubble in H_2SO_4 solutions.[2,3] However, at higher driving acoustic power, accurate fittings to characteristic spectra become difficult owing to the unresolved line profiles among many fine lines and the development of line asymmetries. Thus a method to decompose these unresolved fine lines is needed.

In this Letter, based on the classical Fourier transform theory, we successfully decomposed the asymmetric overlapped doublet of sodium (Na-D lines), then obtained their line broadening, and then obtained the temperature and pressure during MBSL in argon-saturated sodium hydroxide aqueous solution.

In the potential model of Düren and Gröger,[4] it was assumed that there is no difference between the $A^2\Pi_{1/2}$ and the $A^2\Pi_{2/3}$ interaction potentials, which leads to an equal shape but different intensities for Na-D lines. Considering this, we can write the observed spectrum intensity at wavelength λ as a sum of N fine lines as

$$X(\lambda) = \sum_{j=1}^{N} X_j(\lambda), \tag{1}$$

and each line is the convolution of its profile and the isolated line which can be abstracted as a Dirac-function having no width. Thus the intensity distribution of the line centered at wavelength λ_j is written as

$$X_j(\lambda) = A_j \int_{-\infty}^{\infty} F(\tau)\delta(\lambda - \lambda_j - \tau)d\tau, \tag{2}$$

where A_j is the intensity of the jth component, $F(\tau)$ is the mutual line profile of Na-D lines, and $\delta(\lambda - \lambda_j)$ is the intensity of the line centered at λ_j. According to the convolution theorem,[5] the inverse Fourier transform of Eq. (1) leads to

$$x(k) = f(k)\sum_{j=1}^{\mathrm{N}} A_j e^{ik\lambda_j}, \tag{3}$$

where k is the wave number. Thus we can reconstruct the jth component as

$$X_j(\lambda) = \mathcal{F}\Big\{A_j e^{ik\lambda_j}\frac{x(k)}{\sum A_n e^{ik\lambda_n}}\Big\}, \tag{4}$$

where $\mathcal{F}(\bullet)$ represents the Fourier transform. To decompose the Na-D lines, the intensity ratio of D_2(589.0 nm) and D_1(589.6 nm) is needed. From the work of Volz *et al.*,[6] we obtained this ratio of 2:1, thus the Na-D lines can be decomposed by

$$X_{D_1}(\lambda) = \mathcal{F}\Big\{\frac{e^{ik\Delta\lambda}x(k)}{2 + e^{ik\Delta\lambda}}\Big\}, \tag{5}$$

$$X_{D_2}(\lambda) = \mathcal{F}\Big\{\frac{2x(k)}{2 + e^{ik\Delta\lambda}}\Big\}, \tag{6}$$

where $\Delta\lambda$ is the distance between D_1 and D_2 lines.

We experimentally studied the MBSL spectrum in argon-saturated NaOH aqueous solutions of 3 mol/L concentration. A cuboid quartz glass cell with a flat optical pass band in visible wavelengths was used as a solution container. The size of the cell is 50 ×

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$50 \times 60\,\text{mm}^3$ and this 150 mL volume solution was maintained at 286.15 K by circulating temperature-controlled water around the cell. The spectra of MBSL were collected using a system of a monochromator (Acton Research SpectraPro 300i) having a 0.3 m focal length, a cooled CCD detector (Princeton Instruments, model PIXIS) matrix consisting of 1340 pixel×400 pixel of 20×20 per pixel size, and a diffraction grating of 1800 groove/mm blazed at 500 nm. High resolution spectra around the sodium emission fine lines were measured at a variable electric driving power in the range of 150–600 W by using a Ti horn ultrasonic transducer (Sonics & Materials VCX750) with a fundamental frequency of 20 kHz. The entrance slit width is 0.2 mm and the instrument bandwidth was estimated to be 0.08 nm from the measured spectral lines emitted from a low pressure sodium lamp. The obtained spectra of different resolutions are shown in Fig. 1.

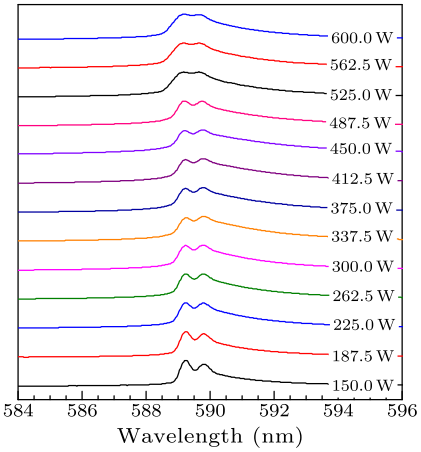

Fig. 1. (Color online) Spectra measured by using different driving powers of 150–600 W.

With the application of the decomposition method, we separated the asymmetric overlapping doublet of the Na-D lines as shown in Fig. 2.

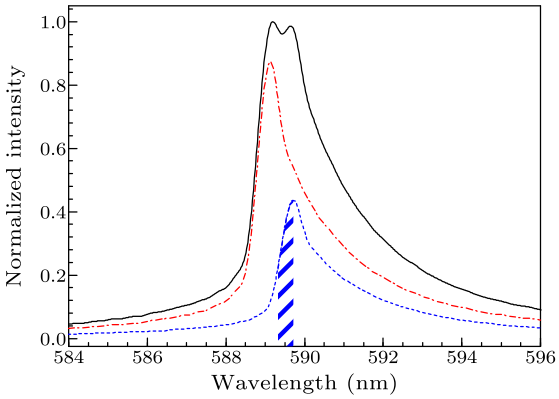

Fig. 2. (Color online) The decomposition of Na-D lines.

The major theories about the atomic line profile are impact theories. Theories in which the Fourier series was obviously asserted, i.e., generally so-called phase-shift theories, predicted a Lorentzian line core but less about the line wing.[7] This Lorentzian profile is

$$I(v) = \frac{(\Gamma/2)/\pi}{(v - v_0)^2 + (\Gamma/2)^2}. \tag{7}$$

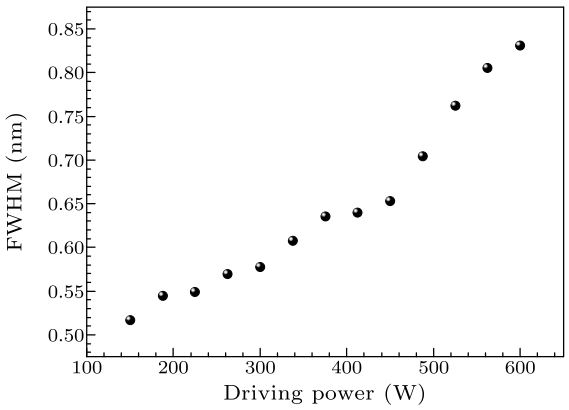

Fig. 3. The FWHM of Na-D lines as a function of the driving power.

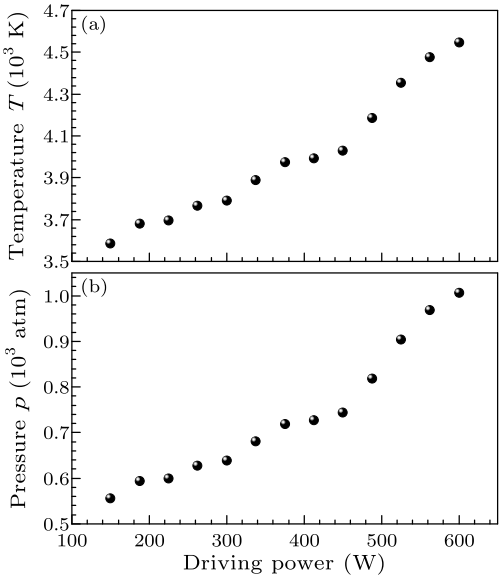

Fig. 4. (a) The temperature and (b) pressure of collapsing bubbles as a function of the driving power.

According to the phase shift theory of line broadening,[8] the full width at half maximum (FWHM) can be expressed as

$$\Gamma = N_{\text{A}} \sqrt{\frac{8}{\pi^3 R\mu}} \frac{\sigma p}{\sqrt{T}}, \tag{8}$$

where N_{A} is the Avogadro constant, R is the mole gas constant, μ is the reduced mass of the radiator Na and the perturber Ar, σ is the collision cross section defined as $\pi(r_{\text{Na}} + r_{\text{Ar}})^2$ with r_{Na} and r_{Ar} being the van der Waals radii of atoms Na and Ar, respectively, T and p are the temperature and pressure of the perturbers, respectively. Thus the line width can be a probe to the temperature and pressure inside the sonoluminescencing bubbles. One phenomenon which

can be easily observed is that the red wing of the profiles is asymmetric apparently. This phenomenon may be due to the generation of dense plasma which causes the decline of the ionic potential.[9] Considering the asymmetric factor and the Lorentzian line profile being effective in the line core which locates in the range of FWHM, we fit the separated line profile only in the range of the line center to the half maximum at the blue wing as shown in the shadow area in Fig. 2 for clarification. After the deconvolution of the instrument width portion, the fitted FWHM of the Na-D lines are obtained as shown in Fig. 3.

To estimate the temperature and the pressure, another constraint condition is needed in addition to the width of the characteristic line. At the collapsing phase of the bubbles, the dynamic time scale is far less than the diffusion time scale, thus we assume that the gas inside the bubbles moves adiabatically, then we have the adiabatic equations as

$$T = T_0 C^{\gamma-1}, \tag{9}$$

$$p = p_0 C^{\gamma}, \tag{10}$$

where T_0 is the initial temperature inside the bubble, p_0 is the initial partial pressure of the perturber particles inside the bubble, C is the compression ratio of initial volume to collapsing volume of the bubble, and γ is the adiabatic coefficient. With the union of Eqs. (8)–(10), the expressions of temperature and pressure can be written as

$$T = \left(\frac{\Gamma}{N_{\rm A}\sigma p_0}\sqrt{\frac{\pi^3 R\mu}{8}}T_0^{\frac{\gamma}{\gamma-1}}\right)^{\frac{2(\gamma-1)}{\gamma+1}}, \tag{11}$$

$$p = \frac{\Gamma}{N_{\rm A}\sigma}\sqrt{\frac{\pi^3 R\mu}{8}T}. \tag{12}$$

Under the conditions $T_0 = 286.15\,{\rm K}$, $p_0 = 1.013 \times 10^5\,{\rm Pa}$, $\gamma = 1.67$, the calculated temperature and the pressure have been plotted in Fig. 4. It is easy to see that the temperature and pressure are ranged in 3600–4500 K and 560–1000 atm, respectively.

In summary, we have proposed a method that decomposes the asymmetric overlapping doublet of the Na-D lines, and based on this method, we have investigated the extreme conditions inside the bubbles during MBSL. It is shown that as the driving power increases, the FWHM of the Na-D lines increases, which indicates a higher temperature and pressure of the collapsing bubbles. The calculated temperature from the Na-D lines reflects the conditions in the spatiotemporal region occupied by the emitting Na atoms, and the calculated pressure only reflects the conditions in the spatiotemporal region occupied by the perturbing particles. Finally, the spectroscopic method provides a probe of the extreme conditions during MBSL.

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