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Spectroscopic characteristic of conical bubble luminescence*

Chen Qi-Dai(陈岐岱)[a], Fu Li-Min(付立民)[b], Ai Xi-Cheng(艾希成)[b], Zhang Jian-Ping(张建平)[b], and Wang Long(王 龙)[a]†

[a] *Institute of Physics, Chinese Academy of Sciences, Beijing* 100080, *China*
[b] *State Key Laboratory for Structural Chemistry of Unstable and Stable Species, Institute of Chemistry, Chinese Academy of Sciences, Beijing* 100080, *China*

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The conical bubble sonoluminescence (CBSL) from the collapse of the bubble was observed in an improved U-tube apparatus. The emitted light energy of a single CBSL flash was measured to be ~ 1.4mJ. The pulse width was about 100μs. The spectra of luminescence were continuum superimposed with the spectral bands from the excited-state C_2, CN and CH. The CBSL provides a link between the light emission of the single-bubble and the multi-bubble sonoluminescence (SBSL and MBSL).

Keywords: sonoluminescence, transient cavitation, bubble
PACC: 7860M, 4325

1. Introduction

Acoustically-driven gas bubbles in liquids can emit short bursts of light (sonoluminescence: SL) as they collapse. This luminescence is generally thought to be accompanied with the creation of extremely high temperature and pressure at the centre of the bubbles. In the case of an air-filled bubble in water at ambient pressure, it is known that SL only occurs when the water contains dissolved argon and the impressed acoustic field has a pressure amplitude in excess of 1×10^5Pa.[1] SL can also occur when a liquid flows through a Venturi tube,[2] when a liquid jet impacts a stationary plate,[3] a superheated steam bubble is injected into a liquid,[4] a pulsed laser beam is focused into a liquid[5] or a spark discharge occurs in a liquid.[6] When Leighton *et al*[7] studied the dynamics of the Rayleigh-like collapse of a conical bubble, they observed the luminescence from the conical bubble with a high-speed camera. However, no detailed luminescence features, such as the pulse shape and emitted spectrum were reported. In this paper we present our improvement of Leighton's apparatus and the temporal and spectroscopic features of the luminescence.

2. Apparatus

We adopt and improve the U-tube conical bubble device that was originally employed by Leighton *et al.*[7–9] The excitation method used in this experiment is quite different from the conventional one to obtain SL; hereafter, we call it conical bubble luminescence (CBL). As shown in Fig.1, a U-tube is partially filled with a volume of 1,2-propanediol that is 20cm in height, empty space in both arms of the tube is charged with argon at a pressure of 1×10^5Pa. A gas inlet valve is attached to the end of the left arm, whereas the other end of the tube is terminated with a hollow cone structure, having a half angle of 30 degree. For optical detection a 1.5mm diameter hole is opened at the apex, and sealed by a quartz window. To ignite a SL flash, argon inside the arms is first evacuated to a desired pressure (the initial pressure P_0, e.g. 300Pa or 1000Pa) before closing the regulation valves, external argon at 1×10^5Pa is then introduced through the gas inlet valve, exerting a pressure on the liquid and moving it rapidly to the right. Gas in the right arm is thus compressed and collapsed violently into the apex

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†E-mail address: wanglong@aphy.iphy.ac.cn

of the cone to generate SL. The single SL flash is intense enough to be visible by naked eye in a bright environment, and can even act as the flash to illuminate an object for taking an ordinary photograph.

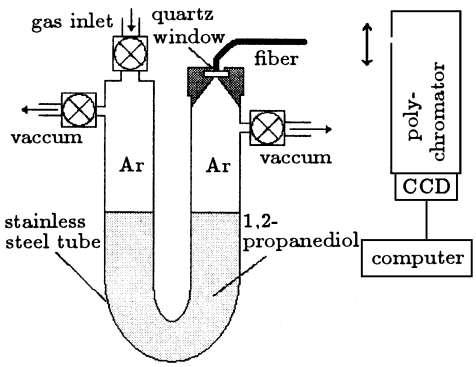


Fig.1. Experimental setup. The stainless steel tube in U-shape, with 40 cm in height and 2.6 cm in inner diameter.

The light emission from the bubble was monitored by a photomultiplier tube (R212, Hamamatsu photonics), whose output was measured across the 50Ω input of an Tektronics 620B oscilloscope. For spectroscopic measurement the luminescence was collected through the quartz window into a fibre optic bundle butted up against the window. The bundle consisted of ~800 UV-pass quartz fibre with 200μm in diameter distributed to fill a circular area. The light from the output end of the fibre was focused by a 25mm diameter objective lens onto the entrance slit of the polychromater (Spectra 500i) equipped with a 150 groove grating blazed at 500nm and a slit set at 50μm. The incident light fell upon a liquid-N_2 cooled charge-coupled device (CCD) array. The resolution was just under 1nm.

In the present study, the U-tube and the conical head are made of stainless steel to avoid being broken by the strong liquid impact. Therefore, it is practically difficult for us to image the bubble dynamics as Leighton *et al* did. However, if we assume that only the 1×10^5Pa ingoing gas works on the liquid column and ignore the pressure of the bubble, the gravity and other dissipation effects, with the following formula we can make a simple estimation about the linear speed of the liquid column with reference to the wall of the U-tube:

$$P_{\rm atm} S \frac{L}{2} = \frac{1}{2}\rho S L v^2, \qquad (1)$$

where S represents the cross section area of the U-tube, L is the height of the U-tube, ρ the density of liquid, and v the speed of the interface separating the liquid and gas phases, which is $v = \sqrt{P_{\rm atm}/\rho} \approx 10$m/s. The speed is approximately independent of P_0 and L. The liquid can be accelerated further when it gets into the section of the conical head. As the area of the liquid-gas interface to be 1% of the U-tube cross section, the speed would possibly reach 1000m/s which is comparable to the speed of sound.

3. Results and discussion

When the apparatus was operated at an initial argon pressure of P_0=1000Pa, a precise optical power meter (Yakowa 3292, Japan), whose detector is 10mm in effective diameter and sensitive to the spectral range of 400–1100nm, measured a single flash energy of 21 nanojoule. To avoid overexposure, the detector is 65cm away from the observed window. Assuming a uniform distribution of the optical radiation in the solid angle of 4π, the light energy of a single flash is estimated to be 1.4mJ. Taking an average photon energy of 2.0eV, the overall photon flux is then estimated to be $\sim5\times10^{15}$ photons/flash, a value that is nine to ten orders of magnitude higher than that of a typical SBSL pulse, i.e. 10^5–10^7 photons/flash.[1]

Figures 2(a) and 2(b) show two representative spectra in the spectral range from 300 to 655nm for two individual luminescence events at the initial pressure P_0=1000 Pa (a) and P_0=300Pa (b), respectively, recorded with a CCD spectrometer. Contrary to other kinds of SL, the continuum part of the spectrum shows an increase in intensity from short wavelengths to long wavelengths. The peaks with band heads at 438, 473, 516 and 563nm are assigned to $\Delta\nu$=+2, +1, 0, and –1 of the $d^3\Pi_g \rightarrow a^3\Pi_u$ Swan transition of C_2, respectively. In addition, Fig.2 shows the CN* peaks at 386 and 416nm, and a weak peak at 431nm, which we assign to the $A^2\Delta \rightarrow A^2\Pi_u$ transition of CH. Comparing the two cases of different initial pressure, it is clear that a higher initial gas pressure results in a stronger continuum and also a stronger total light intensity, while the molecular emission dominates the spectrum at a low initial pressure.

Although the nature of the light radiation mechanism is unknown, a useful means of characterizing the spectrum is to fit it with a blackbody spectrum.[10] The dotted line in Fig.2 indicates the fit to the spectrum radiated by a blackbody. The fit is especially good for wavelengths greater than approximately 400nm. The intense absorption of 1,2-propanediol at wavelengths below 400nm would be possible to answer for the large off-track fit spectral features in the ultraviolet. The best-fit temperature for our CBL spec-

trum is 3000K. For single bubble sonoluminescence (SBSL), Barber *et al*[1] found the best-fit temperature for their data to be 25000K and significantly larger than for the CBL data. This would imply that the collapse in SBSL would be much violent than that from CBL.

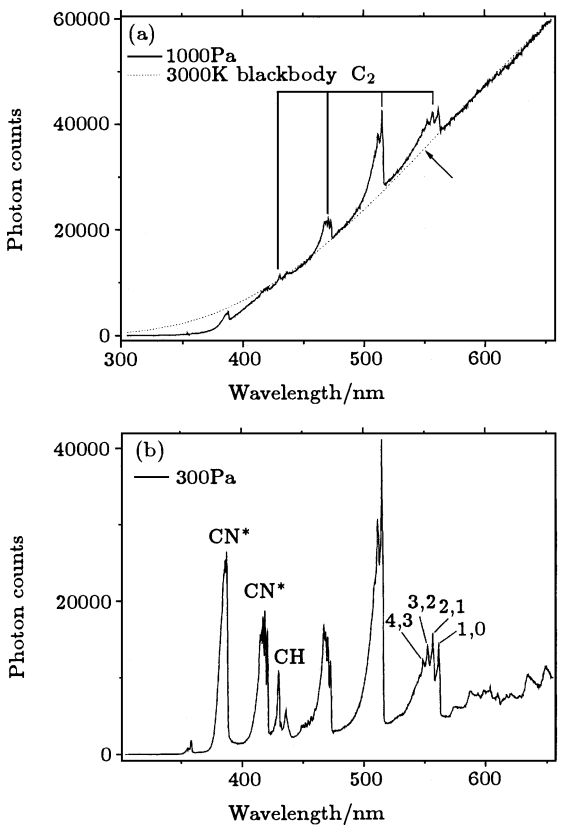


Fig.2. Spectrum at P_0= 1000Pa (a) and 300Pa (b) recorded by a conventional spectrometer equipped with a CCD detector (solid line) and its blackbody fit (dotted line).

We can make a simple estimate about the maximum temperature that CBL can reach. Considering an adiabatic compression, and assuming that the whole kinetic energy of the liquid is converted to the internal energy of the gas, we obtain the following three equations:

$$P_0 V_0 = nkT_0, \tag{2}$$

$$\frac{3}{2} nkT_{\max} = P_{\rm atm} V_0, \tag{3}$$

$$T_{\max} V_{\min}^{\gamma-1} = T_0 V_0^{\gamma-1}. \tag{4}$$

Here P_0, V_0, T_0 are, respectively, the initial pressure, volume and temperature of the gas, $P_{\rm atm}$ is the pressure of atmosphere, $V_{\min}$ the minimal volume to which the bubble can be compressed, $T_{\max}$ the gas temperature at the minimal volume, and γ the ratio of the specific heat of the inert gas at constant pressure to that at constant volume. The maximum temperature $T_{\max}$ and the compression ratio $V_0/V_{\max}$ can be obtained as follows:

$$T_{\max} = \frac{2}{3} \frac{P_{\rm atm}}{P_0} T_0, \tag{5}$$

$$\frac{V_0}{V_{\min}} = \left(\frac{2}{3} \frac{P_{\rm atm}}{P_0} \right)^{3/2}. \tag{6}$$

For bubbles with P_0=1000Pa and T_0=300K, the compression ratio obtained from Eq.(5) is about 500, which is much less than that of the SBSL (typically, 100, 000), and the maximum temperature is estimated to be about 20000K (Eq.(5)) which is significantly larger than the actually measured spectroscopic temperature (~3000K). For SBSL, a maximum temperature of 10^4K during the collapse was widely acknowledged by theoretical researchers.[1] However, if all the dissipation effects, such as the viscosity, surface tension, heat conduction and liquid vaporization during the collapse were ignored, a high temperature of 100000K can be easily attained through numerical simulation. The bubbles in conventional SL were compressed and collapsed in three-dimensional space, while in a sense, the bubbles in CBL are compressed in a more than one-dimensional space, depending on the coning. A lower compression dimension leads to a smaller compression ratio.

A distinct feature of the multibubble sonoluminescence (MBSL) spectra is the appearance of molecular emission,[11] which is absent in SBSL. One possible explanation for this difference is the relative lower temperature in MBSL than that in SBSL. As shown in Fig.2, the molecular bands are also present in the case of CBL. Furthermore, both the spectroscopic temperature and that from the simple theory estimation (Eq.(5)) are much lower than that of SBSL. Therefore, the mechanism to generate CBL might have some similarities to that for MBSL.

In 1987, Suslick *et al*[12] discovered the Swan bands emission in MBSL for the first time with dodecane. Swan bands chemiluminescence is seen from hydrocarbons under a wide variety of conditions, including flames, shock tubes, shock pyrolysis, plasmas, laser ablation, and low-temperature discharge. The appearance of Swan bands suggests a thermal mechanism of luminescence. The vibration structure of 563nm peaks can be clearly seen in Fig.2(b). The peaks, going from high to low wavelength, correspond to the ($\upsilon' \to \upsilon''$) transition (1–0), (2–1), (3–2), (4–3). These indicate

that the emission is from the gas phase, since resolution of the vibrational bands is not expected from liquid-phase emission. In addition, the observation of CN^* bands discard the electrical discharge hypothesis, which would lead to the N_2^* emission otherwise.

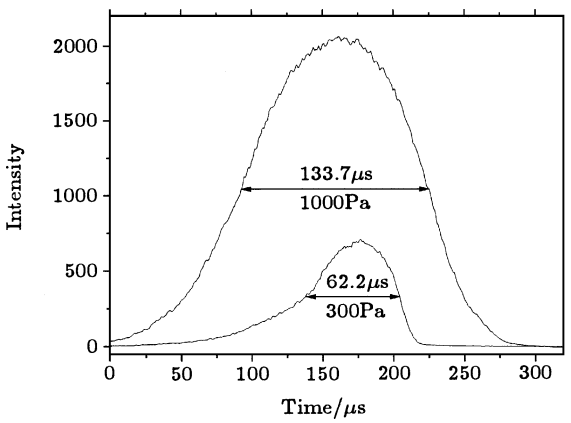


Fig.3. SL pulses at different initial pressure.

The pulse width of the flash is now considered to be a key parameter in understanding the physics of luminescence. The temporal evolutions of a single SL pulse at different initial pressures (1000 and 300Pa) are displayed in Fig.3. The full widths at half maximum for 1000 and 300Pa are 130 and 62μs, respectively, which are significantly larger than those from MBSL and SBSL, whose durations are of subnanosecond or nanosecond scales. We are concerned with whether the presently measured relatively long CBL flash durations are related to the fact that our observations are in multibubble environment, since under these conditions it is possible that the observed pulse shapes are the results of convolution or superposition of optical signals from a large number of SL events. However, if it is from a multibubble environment, it is hard to warrant the irregular luminescence pulses to be superposed together to form such a smooth luminescence curve.

The luminescence intensity at an initial pressure of 300Pa is significantly lower than that at 1000Pa initial pressure. However, according to Eq.(5), a lower initial pressure should result in a higher temperature and then a stronger luminescence. A possible explanation for this inconsistency is that a higher initial pressure implies more gas emitters, and therefore results in a stronger luminescence.

Compared with SBSL, the present conical bubble exhibits lack of the clock-like regularity, the subnanosecond short time, and a higher inner temperature. But due to the robustness of this method, it will also be of interest to apply it to other liquids or gases where SBSL is difficult or impossible to study. We can make certain that for the inner reaction gas, while in SBSL, the gas builds up in many thousands of acoustic cycles. Furthermore, all the bubbles collapse under the liquid inertia. However, experimental control of the liquid inertia in SL has not readily been achievable without changing the liquid density and, simultaneously, changing other liquid properties. In this apparatus the inertia at the early stages of the collapse of a conical bubble can easily be controlled. Perhaps this method will yield a means of improving industrial processes that rely on cavitation.

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