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Time-resolved spectra of single-bubble sonoluminescence in sulfuric acid with a streak camera

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The time-resolved spectra of single-bubble sonoluminescence (SBSL) in sulfuric acid have been observed with a streak camera after a spectrograph. The spectral center evolves from infrared to ultraviolet gradually within a SBSL duration, which corresponds to an increase of temperature. The peak temperature within one sonoluminescence (SL) duration is 5–9 times higher than the average temperature based on the average spectrum in our experiment. Furthermore, the ratio of the peak temperature to average temperature increases with the increase of driving pressure. The SBSL flash dies out after a dramatic heating-up, and there is no cooling procedure observed at the time resolution of 1/10 SL duration, which is incompatible with the radius-related adiabatic heating model as the mechanism of SBSL.

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A nonlinearly oscillating spherical bubble possesses the best focusing ability of energy in (3+1)-dimensional Minkowski space. A cavitation bubble can focus sound energy by 12 orders of magnitude and emit light during bubble collapse [1]. The temperature inside a collapsing cavitation bubble had been estimated as high as several thousand kelvins in early multiple-bubble sonoluminescence [2] and tens of thousands of kelvins in single-bubble sonoluminescence (SBSL) [3]. The driving sound pressure in SBSL is usually of the order of 10^5 Pa [3]. In theory the temperature can be expected to reach the threshold of thermonuclear fusion if stronger driving sound is used, such as 1.5 MPa, although observations of sonofusion [4,5] have been disputed [6,7] since proposed in 2002. Recently, scientists [7] have repeated the sonofusion experiments and found that the neutron signal from thermonuclear fusion was much lower than that observed previously [5]. However, the new experimental results could not rule out the possibility of sonofusion, because even a small increase in bubble temperature would lead to a huge increase in the rate of *D-D* fusion [7]. Therefore, it is the temperature that is the most crucial variable of the bubble interior for which direct measurement is not possible. The light emission is usually expected to be sensitive to the temperature [8–10]. One can get a blackbody temperature $\overline{T_b}$ to describe the spectrum by fitting the luminescing spectrum to the blackbody radiation curve [8], although the cavitation bubble is not an idea blackbody. The experimental value of $\overline{T_b}$ is at least 25 000 K [11]. Recently, some atomic and ironic lines have been observed in SBSL in sulfuric acid (SA), and the internal temperature can be estimated by calculating the widths of the lines [9,10]. The estimated temperature calculated from the spectral line is in agreement with that from blackbody fits [9,10]. Obviously, these temperatures are the average ones over the whole luminescing pulse because the spectra used to calculate have been integrated and lose the time correlation. There is an evolution of the SBSL spectrum within each pulse, although the SBSL pulse duration is measured on the order of hundreds of picoseconds in water-air systems [12,13]. The instant temperature together with its evolution remains unknown due to the lack of time-resolved spectra (TRS) within a SBSL pulse. In 1998 Hiller *et al.* used the time-correlated single-photon counting (TCSPC) [14] to get the TRS of SBSL and obtained some important information about the emission mechanism [12]. Although the TCSPC is good at detecting dim light, it is not capable of capturing a continuum spectrum in a single measurement. As a result, Ref. [12] reported the TRS of the sonoluminescence in water with wavelength step of 50 nm, which is quite sparse to resolve the spectrum. On the other hand, the TRS can be obtained by using the streak camera combined with a spectrograph. However, the sonoluminescing light passing through a spectrometer becomes too dim to expose the streak camera, which makes the measurement impossible based on the streak camera up to date. Fortunately, the bright SBSL in SA dissolved with argon [9] enables one to observe its TRS with a streak camera. The early SBSL in SA is taken place in a moving bubble [15]. For both measurements of bubble dynamics and SBSL spectrum, the stable levitation of the luminescing bubble is quite important. Today one has levitated a luminescing bubble with strong stability in SA as reported in Refs. [16,17]. In this paper, the TSR of sonoluminescence in SA is reported by using a combination of a streak camera and a spectrometer.

The experimental apparatus consisted of a spherical acoustic resonator and the spectral acquisition system (see Fig. 1). The acoustic resonator, a 100-ml glass flask with a quartz window, was driven by ultrasound at its resonant frequency of about 26 kHz. The host liquids used here were aqueous SA with different concentrations. Krypton gas was dissolved into the host liquids under pressure head 40×133.3 Pa in our experiments.

The TRS are observed by a combination of a streak camera (Hamamatsu C7700) and an spectrograph (Acton Research SP-300i). The C7700 streak camera is a specialized model for dim light. It consists of two microchannel plates (MCPs), so that it is more sensitive to dim light than other models, such as C5600 used for the measurement of the unprismatic SBSL pulse [13]. Furthermore, MCPs have no weakening effect to the dim light before light passes into the streak tube, because both MCPs are behind the streak tube. The time resolution of C7700 is 5 ps, which is enough for resolving the light pulse of several hundred picoseconds. In

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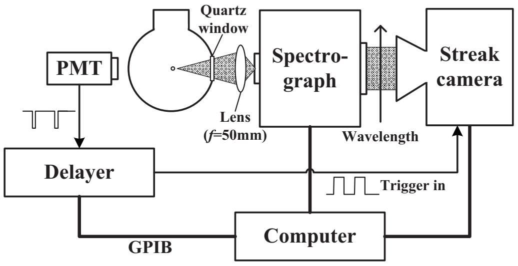

FIG. 1. Sketch of the experimental setup for measurements of the time-resolved spectra of SBSL. The light pulse is focused by a lens ($f=50$ mm) and spreads in wavelength by a spectrograph. The output of the spectrograph injects the streak camera. All of them are controlled by a computer via GPIB (IEEE 488).

fact, the SBSL pulse duration is several nanoseconds in our SA-Kr system. The spectrograph SP-300i uses two 150 rulings/mm gratings blazed at 300 and 500 nm, respectively. The output of the spectrograph injects into the entrance slit of the streak camera. This combination will output the information on the photon's arriving time and its wavelength.

The streak camera was triggered by a previous SBSL pulse through a photomultiplier (PMT, Hamamatsu R212) and a time delayer (Stanford DG535). The block diagram of the experimental setup is plotted in Fig. 1. The catching time window of the streak camera should be set slight larger than the pulse duration. Before the measurement of the TRS we measured the unprismatic SBSL pulse duration. The result showed that the pulse duration was about 10 ns, which is much wider than that in water-air systems [12,13]. Finally the time window was set 20 ns.

Although the SBSL in SA [9,16] is much brighter than that in a water system, the pulse after the grating of the spectrograph (say, the incident light of the streak camera) is much weaker than the original one before passing into the spectrograph. As a result, only a few photoelectrons could arrive to the streak tube in a single SBSL pulse. It is necessary to take many streak images to integrate. In a measurement we took 2000 streak images in which about 98% of them could be used to integrate and the other 2% were not full images of a SBSL pulse due to the nonstrict periodicity in the time scale of SBSL duration. The superposition of the mass center of the streak image can eliminate the error generated by the nonstrict periodicity of SBSL. Figure 2 shows the integrated streak image of all of effective single images, where the horizonal axis is wavelength and the vertical axis is time.

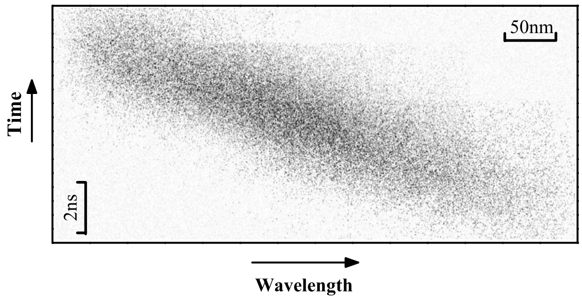

FIG. 2. The streak image of SBSL in 85 wt % sulfuric acid dissolved with 40×133.3 Pa krypton gas. The driving parameters are 26.3 kHz and 1.40×10^5 Pa, respectively.

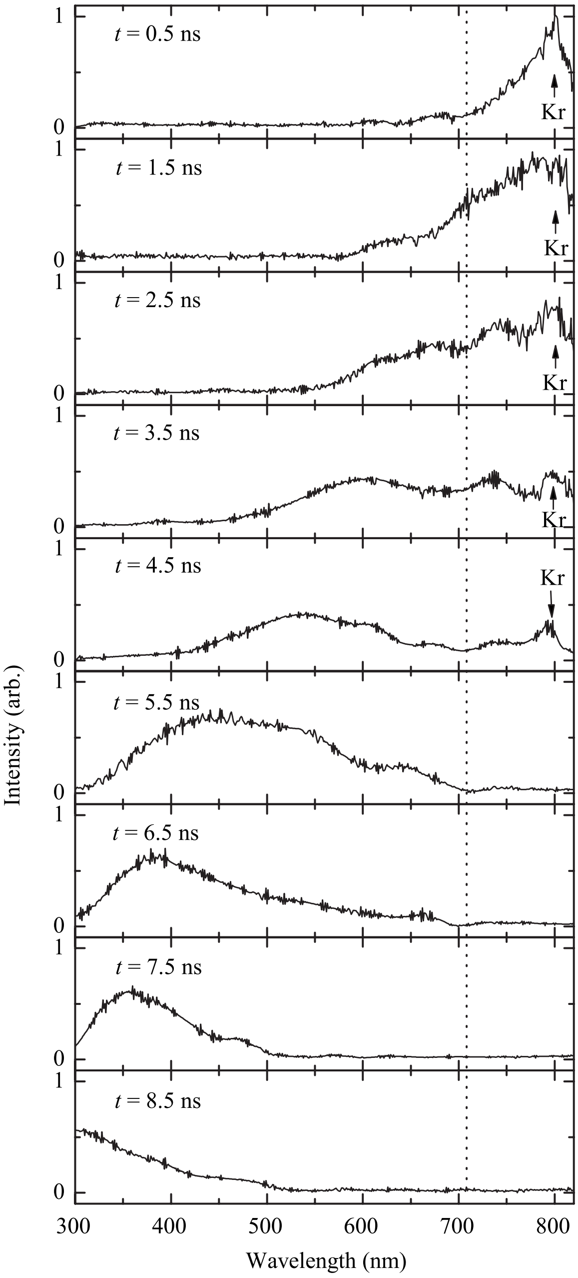

FIG. 3. The time-resolved spectra from Fig. 2. The binning interval is 1 ns. The central time of each binning interval has been labeled. The arrows denote the atomic lines of krypton of $5s$-$5p$. The spectral lines are mainly in the red and infrared, and are confined to the wavelengths to the right of the dashed line. The spectra were calibrated for the absorption of the host liquid and the quartz window against standard lamps.

It is clear that the spectrum evolves from long wavelengths to short wavelengths. Binning Fig. 2 along time (vertical) axis with binning interval 1 ns, we can obtain nine spectra at different time (see Fig. 3). It means that Fig. 2 was cut horizontally into nine parts. Each part represents the spectrum during 1 ns. The spectra in different moment

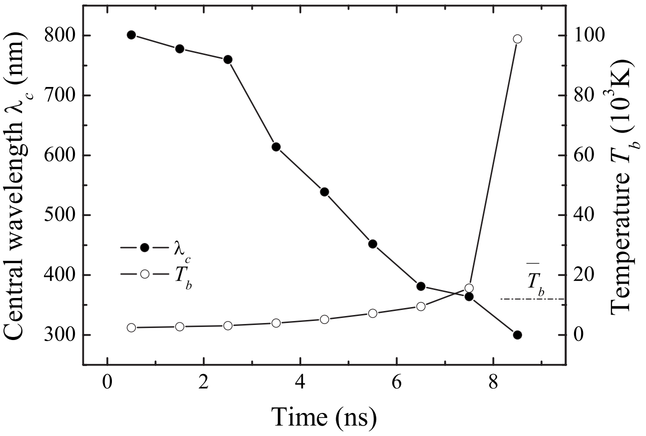

FIG. 4. The evolution of the central wavelength λ_c (●) and the blackbody temperature (○) in an SBSL pulse in a SA-Kr system. With time, the spectral center moves towards the violet and the temperature increases.

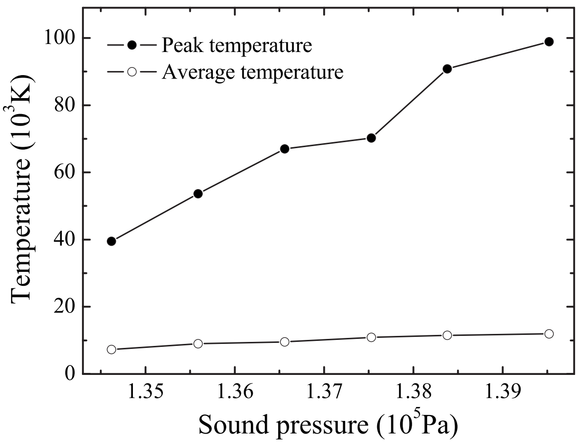

FIG. 5. The peak and average temperatures inside one SL duration driven by different pressure. The host liquid is the same as that in Fig. 2.

within a pulse duration are very different each other. At the beginning of the luminescing, the first 1 ns labeled central time $t=0.5$ ns in Fig. 3, the light is composed of some long wavelengths centered at $\lambda_c \geqslant 800$ nm. Then λ_c shifts toward violet gradually. Finally the central wavelength arrives at <300 nm. Figure 3 shows the evolution of λ_c as solid dots.

In order to parametrize the evolution of the spectra, we fit the continuum waves (the left-hand side of the vertical dashed line in Fig. 3) of the TRS to the blackbody curve and get the corresponding blackbody temperatures T_b. In Fig. 4 both the central wavelength and blackbody temperature are plotted as functions of time within a SBSL duration. Within a sonoluminescing pulse the luminant inside the bubble is heated slowly at the beginning, and it is extinguished suddenly after a rapid calefaction (see open dots in Fig. 4). This evolution of the temperature T_b is consistent with the diminishing of the krypton atomic emission (the right-hand side of the dashed line in Fig. 3). As reported in Refs. [9,16,18] the atomic lines are always observed in SBSL driven by relatively low pressure, which shows that atomic lines indicate the relatively low temperature inside the sonoluminescing bubble. In Fig. 3 the atomic lines of krypton centered at 810 nm from the transition of $5s$-$5p$ [9] can be clearly identified at the beginning of the luminescing. As the gas trapped in the bubble is further heated, the intensity of the krypton lines decreases relatively to the continuum emission, and they broaden gradually into completely unresolved peaks in the later stage.

It is interesting that the peak temperature $T_{b,\mathrm{peak}}$ in sonoluminescence of our SA-Kr system reaches 100 000 K, about 9 times higher than the $\overline{T}_b=11\,800$ K (see open dots in Fig. 4). Although the peak and the average temperature have been studied by others [19], the time scales are different. The peak temperature and the average temperature discussed here are in one SBSL pulse duration. We measured the TRS for SBSL in SA driven by different sound pressures. The peak temperatures range from 5 to 9 times higher than the average one (see Fig. 5). The ratio of $T_{b,\mathrm{peak}}/\overline{T}_b$ increases with the driving pressure. Considering that the sonofusion experiments [4,6,7] used a pressure of 1.5 MPa, over 10 times higher than what we used, we can expect the $T_{b,\mathrm{peak}}$ can be at least one order of magnitude higher than the average one. Therefore the peak temperature inside a sonoluminescing bubble is much higher than that estimated by the integrated spectrum.

To understand the increasing of sonoluminescing intensity in SA we measured the pulse widths [full widths at half maximum (FWHM)] of SBSL in SA solutions with different concentrations. The result shows that the pulse width increases with SA concentration (see Fig. 6). The sonoluminescing pulse width arrives at its maximum, 4.35 ns, in the concentration 85 wt %, and at its minimum, 0.24 ns, in pure water, respectively. Therefore we attribute the increase of the sonoluminescing intensity to the broadening of the pulse width.

If we bin Fig. 2 along the wavelength, we can arrive at the flash width and emission time at different wavelengths as did in Ref. [12]. From Fig. 2 we can know that the flash width is indeed independent of the wavelength [12]. However, the emission times are closely related to the wavelength, which is different from the observation of SBSL in water based on the TCSPC [12]. With evolution of time, the central wavelength moves from infrared to ultraviolet monotonously (see

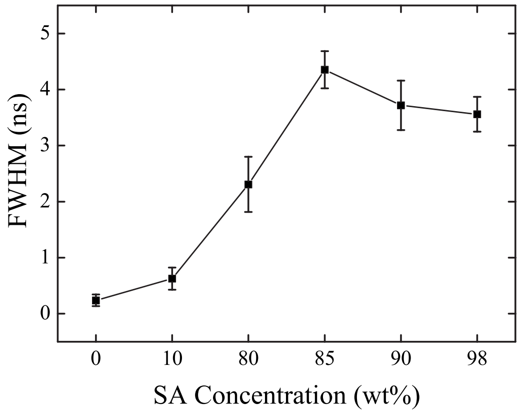

FIG. 6. The pulse durations of typical SBSL in sulfuric acid solutions with different concentrations. The krypton gas has been dissolved into solutions under a pressure of 40 × 133 Pa.

solid dots in Fig. 4). There is no cooling procedure observed at our time resolution (see open dots in Fig. 4). The luminescing dies out after its hottest spark, which provides an important clue to the mechanism of SBSL. We notice that based on Rayleigh-Plesset bubble dynamics [20] the radius rebounds slowly after its minimum value [21]. In other words, the bubble volume collapses more quickly than it rebounds after its minimum. Therefore, a monontonic increase in temperature as suggested by Fig. 4 is incompatible with the adiabatic heating model as the mechanism of SBSL.

In conclusion, the TRS of SBSL in a SA-Kr system have been measured. Both continuum spectra and atomic emissions are observed in the TRS. In continuum spectra, the central wavelength moves from infrared to ultraviolet gradually, which means the bubble is heating up. The heating-up procedure is further validated by the evolution of the krypton atomic emissions. The TRS denote that the peak temperature inside one SBSL duration is usually several times higher than the average temperature based on the integrated spectrum. The ratio of peak to average temperature, furthermore, increases with driving pressure. These results support the conjecture that the instant internal temperature could be at least one order of magnitude higher than that estimated based on the integrated spectrum. Another interesting phenomenon is that there is no cooling procedure observed after a speedy heating-up at the experimental time resolution, which is incompatible with the volume-related adiabatic heating model as a possible mechanism of SBSL.

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