

Temperature dependence of single-bubble sonoluminescence threshold in sulfuric acid: An experimental study

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(Received 21 January 2015; revised manuscript received 11 April 2015; published 30 June 2015)

We experimentally investigated the temperature dependence of intensity of single-bubble sonoluminescence (SBSL) in 85 wt % sulfuric acid. It was found that the intensity increases as temperature increases from 15 °C and 25 °C, confirming what has been predicted by A. Moshaii *et al.* [*Phys. Rev. E* **84**, 046301 (2011)] theoretically. This behavior, however, is completely opposite to what has been observed for water. Above 25 °C, the behavior of intensity of SBSL in sulfuric acid is found to be independent of the liquid temperature. Moreover, it was observed that as the temperature increases, contribution to total intensity from the UV portion of the spectrum increases while contribution from the visible portion decreases, indicating higher bubble temperatures at higher liquid temperatures. Results of this experiment further indicate that the intensity threshold at each temperature is not determined by the shape or the positional stability conditions but by the driving pressure at which the transition from SBSL to multibubble sonoluminescence (MBSL) takes place.

DOI: [10.1103/PhysRevE.91.063015](https://doi.org/10.1103/PhysRevE.91.063015)

PACS number(s): 78.60.Mq

I. INTRODUCTION

In single-bubble sonoluminescence (SBSL), a high-frequency sound wave in the range 15 kHz–1 MHz produces flashes of light with duration of 40–350 ps and a featureless continuous spectrum extends from 200 to 800 nm [1–3]. The intensity of light emitted by the bubble depends on the physical properties of the host liquid as well as the external conditions, such as dissolved gas concentration, ambient pressure, liquid temperature, acoustic driving pressure, and driving frequency [1,2,4,5]. For water-based SBSL, many experimental investigations have been carried out to determine the dependence of light emission on these external conditions. Especially, the temperature dependence of SBSL has been studied both theoretically and experimentally by many [5–7]. They have clearly observed that the total intensity of light emission increases with decreasing temperature, and the light intensity observed at 0 °C was 100 times brighter than at 30 °C. This behavior has been supported by theoretical simulation results [6] and explained in terms of the viscosity of water, the rare gas solubility, and vapor pressure inside the bubble. As water temperature decreases, the shape stability of the bubble increases due to the increase in viscosity of water. This will enable stable bubbles to be achieved at higher acoustic pressures, and the increase of pressure causes the bubbles to attain a smaller minimum radii, higher maximum temperatures, and, consequently, stronger light emissions. Moreover, due to reduction in rare gas concentration and vapor pressure of water at lower temperatures, bubbles will have reduced pressures at the last stage of the collapse and hence have freedom to collapse further into smaller sizes, producing higher internal temperatures. The temperature dependence of SBSL emission in many other liquids has been experimentally explored in literature [4,5]. In almost all tested liquids, the SBSL intensity decreases with the increase in liquid temperature, similar to what has been observed in water.

Among all the liquids tested so far, sulfuric acid produces the most intense SBSL flashes. It has been observed experimentally that at 30 °C, 85% sulfuric acid solutions can produce SBSL flashes, which is up to 2700 times brighter [8] than the same produced by the standard room temperature argon bubble in water. Although at 30 °C SBSL bubbles in sulfuric are very stable and produce very bright light flashes, it has been reported that it is impossible to seed a single pulsating bubble in sulfuric at 0 °C. A few years ago, Moshaii *et al.* [9] theoretically predicted that temperature dependence of light emission in sulfuric is quite opposite to the same observed in water. Through computational simulations, they have found that in fact light intensity decreases as the temperature of sulfuric acid in 85 wt % solution decreases. This remarkable result has never been tested experimentally so far. In this paper we experimentally investigate the temperature dependence of the light emission in sulfuric acid and confirm the results found by Moshaii *et al.* [9]. Further, we found that SBSL spectrum in 85% sulfuric acid solution shows higher spectral density in the visible region at lower temperatures and moves towards UV as the temperature increases. This behavior is completely opposite to what has been observed for water [10].

II. EXPERIMENTAL SETUP

In the present work, a typical experimental setup for obtaining SBSL has been established [2]. Figure 1 shows the basic elements of our experimental setup. The sonoluminescence (SL) bubble was trapped in a 125-mL spherical flask made of quartz glass filled with 85 wt % sulfuric acid–water mixture. The flask system was driven harmonically at the fundamental resonance frequency of the flask, which is about 27.5 kHz, by two piezoelectric transducers attached to the opposite sides of the outer surface of the flask as shown. A small disk-shaped microphone transducer was mounted at the bottom of the flask to monitor the acoustic driving pressure in the flask in a noninvasive way. Ultrasonic sound generated with the signal generator (Agilent 33220) was first amplified with a power amplifier, and then the voltage was enhanced and the

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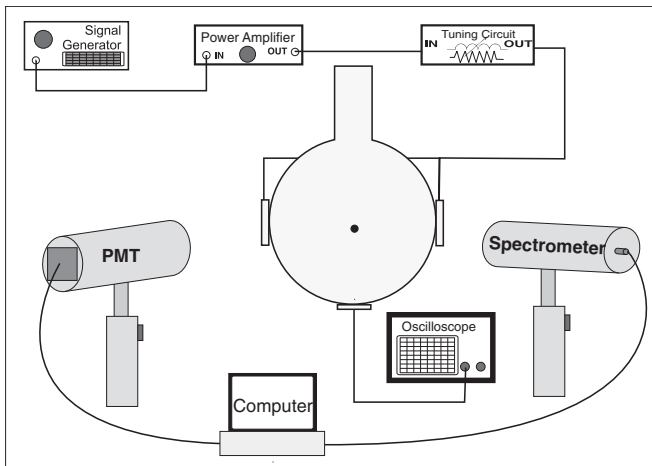


FIG. 1. Experimental setup.

power transfer improved by a tuning resonator circuit. As the temperature of the liquid changed during the experiment, the resonance frequency of the flask changed only slightly (27.45 kHz–27.58 kHz)..

During the experiment, a photomultiplier tube (PMT) (type: Hamamatsu H5784-4) was used to detect the SBSL bubble and measure the relative intensity of the light pulse. A collecting lens (2.5 cm diameter and focal length $f = 4$ cm) was firmly positioned at the end of a tube attached to the PMT housing. The tube was placed such that the bubble was located between f and $2f$ from the collecting lens. Signals from PMT were fed into a computer via a data acquisition board (PCI-DAS 4020/12), and then they were monitored and recorded with TraceDAQ data acquisition software for postaction analysis. Later, the photon count was determined by software developed in our laboratory using the dark reading as the reference. The SBSL spectrum is obtained using an Ocean Optics QE65000 Pro fiber-based spectrometer (wavelength range 200–950 nm) with a homemade lens-collimator system as the front end for collecting light from the bubble. SBSL spectra were displayed and recorded with an Ocean Optics SPECTRASUITE software package. The entire system is calibrated in the 220–920 nm range with an Ocean Optics DH-2000-CAL NIST-traceable fiber-based calibration source. The various components of the system were mounted on an optical bench using translation stages to allow precise positioning and to avoid realignment at each stage of the experiment.

Concentration of sulfuric acid (98% analytical grade) was brought down to 85 wt% by mixing with deionized water. Sulfuric acid (85 wt%) was first degassed by purging with argon under atmospheric pressure for 12 h and then vacuuming under 30 Torr for another 12 h. We repeated this degassing and regassing procedure for five days. During the whole process of gas preparation, a magnetic rotor covered with Teflon was used to stir the solution vigorously to speed up the degassing and regassing processes. Finally, argon was dissolved under 30 Torr for another 2 h. In the experiment, we were able to create a single bubble by just varying the applied driving pressure. At the low acoustic driving level, the bubble gets trapped near the center of the flask. The driving voltage was then gradually increased so that the bubble entered the

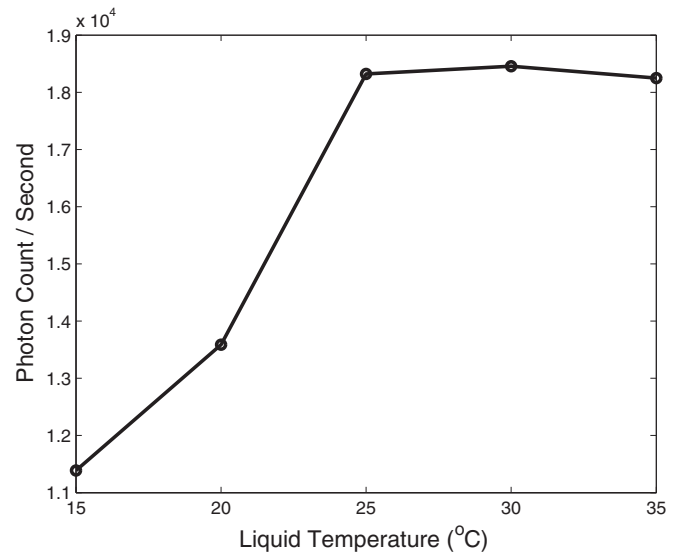


FIG. 2. Number of photons detected per second by the PMT for five different temperatures.

light-emitting region and reached the maximum brightness. Further increase of driving pressure did not make the single bubble disappear due to instability but it became a multibubble system (having between 3 and 15 bubbles).

III. RESULTS AND DISCUSSION

Due to the aforementioned degassing-regassing procedure, it was possible to produce an almost stationary bubble for the temperature range 15 °C to 35 °C. We recorded PMT and spectroscopic data for the temperatures 15 °C, 20 °C, 25 °C, 30 °C, and 35 °C. Figure 2 shows the SBSL intensity as a function of temperature. From the lowest SBSL intensity observed at 15 °C, the intensity substantially increases as temperature increases until 25 °C. However, beyond 25 °C, within experimental error, the intensity seems to remain constant until 35 °C. This behavior is completely opposite to the behavior of SBSL observed in water and agrees well with what has been predicted theoretically by [9].

In [9], Moshaii *et al.* simulated the temperature dependence of SBSL in sulfuric acid for the temperature range 5 °C to 30 °C. They have predicted that the SBSL intensity gradually increases as liquid temperature increases from 5 °C to 25 °C and remains more or less constant between 25 °C and 30 °C. They have predicted similar behavior for the maximum bubble temperature (SL temperature) at the collapse and claimed that the above-mentioned behavior of intensity mainly originates from changes in the SL temperature. Furthermore, they have identified that viscosity and vapor pressure are the only two physical parameters which change considerably with temperature. Since both viscosity and vapor pressure of water and sulfuric acid show similar behavior as the temperature of the liquid varies, a similar temperature dependency of SL emission for both liquids could be expected. However, the results of their simulation showed two opposite temperature dependencies, which seem to originate from the difference in the phase diagrams of the stable SBSL bubble. From the phase diagrams of water and sulfuric acids, they claimed that

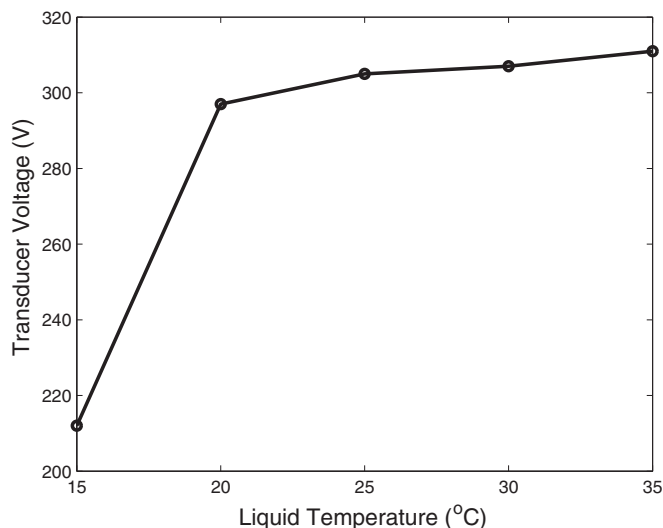


FIG. 3. SBSL threshold (stable SBSL to MBSL) as a function of temperature. The threshold is measured in volts, as it is the driving voltage of the transducers attached to the flask.

the ultimate phase parameters for water are determined by the shape instability, while in sulfuric acid, the phase parameters are restricted by positional instability.

In [11], Urteaga *et al.* have observed that when the acoustic pressure is increased, the bubble moves away from the center of the resonator towards the regions having lower acoustic pressure intensities. Further, they claimed that they have never observed the destruction of the bubble into fragments due to the onset of the Rayleigh-Taylor instability.

In our experiments, when the driving acoustic pressure is larger than a threshold value, we observed the fragmentation of the bubble into a few multiple bubbles which move away from the center of the resonator towards the flask walls, producing multibubble sonoluminescence (MBSL). As a result, unlike SBSL in water, the intensity threshold for sulfuric acid at each temperature is not determined by the stability conditions (shape or positional) but by the driving pressure at which the transition from SBSL to MBSL takes place. In order to understand the relationship between SBSL intensity and the behavior of the SBSL threshold at which SBSL to MBSL transition takes place, the SBSL threshold is plotted against the liquid temperature as shown in Fig. 3. Note that the threshold is measured in volts, as it is essentially the driving voltage of the transducers attached to the flask.

There is a sharp increase in the SBSL threshold from 15 °C to 20 °C and a mild increase for liquid temperatures between 20 °C and 25 °C. This increase in threshold may be one of the reasons behind the behavior of SBSL intensity observed at lower temperatures between 15 °C and 25 °C. Above 25 °C, the threshold voltage shows an almost constant value. This may explain the constant behavior of SBSL intensity between 25 °C and 35 °C.

It is important to note that the SBSL threshold is not determined by the driving pressure at which the bubble becomes unstable due to shape or positional instability. It is entirely governed by the driving pressure at which the transition from stable SBSL to MBSL takes place. Therefore

changes in viscosity do not play an important role in deciding the SBSL threshold. However, viscosity can significantly affect the intensity of SBSL, as it has been observed for MBSL [12,13]. In a theoretical study, Yasui *et al.* [12] showed that up to 20 mPa s, the effect of viscosity on the intensity of bubble collapse is marginal. However, for fluid viscosities > 20 mPa s, the intensity of bubble collapse was found to decrease with increase in viscosity. These predictions are in agreement with our experimental results, as described below.

As the temperature decreases from 30 °C to 25 °C, the change in viscosity of 85 wt % H₂SO₄ is mild (from 17.77 to 19.75 mPa s [9]), and the observed SBSL intensity is almost a constant. On the other hand, when the temperature decreases from 25 °C to 15 °C, the viscosity of 85 wt % H₂SO₄ increases from 19.75 to 35.13 mPa s [9], and SBSL intensity is found to be steadily decreasing with decreasing temperature. For SBSL in water, this reducing effect due to viscosity is compensated by the influence of water vapor [13], and hence the total SBSL intensity increases as liquid temperature decreases. However, the vapor pressure of H₂SO₄ is extremely small (between 10⁻⁵ Pa and 6 × 10⁻⁴ for the above temperature range [14]) compared to that of water (3 × 10³ Pa) and cannot affect the intensity significantly. Therefore the total intensity steadily decreases as viscosity increases.

It has been reported [15] that the observed increase in light emission from a bubble in sulfuric acid at room temperature compared to the same in cold water is correlated with the appearance of rapid translational motion of the bubble. In this experiment, it was observed that at 25 °C, the bubble in sulfuric acid was mostly stationary, while it exhibited some translational motion when the temperature was either increased or decreased from 25 °C. On the other hand, the intensity of the emission increased steadily with increasing temperature from 15 °C to 25 °C and remained constant from 25 °C to 35 °C, indicating an absence of correlation between translational motion of the bubble and the increase in intensity.

We have recorded SBSL spectra for all five temperatures and argon spectral lines have been observed at each temperature. For comparison, spectra at 15 °C and 30 °C are given in Fig. 4. The SBSL spectrum for 30 °C is obtained at a higher driving pressure compared to the same for 15 °C. At 15 °C, argon spectral lines are quite prominent, and raising of the temperature up to 30 °C has produced three main effects: (1) it shifts the spectrum more strongly towards the UV (the peak in the spectrum moved from about 435 nm to about 325 nm), (2) it increases the total intensity of the emitted light, and (3) the intensity of Ar emission lines decreases relative to the continuum emission.

The UV part of the SBSL spectrum is due to blackbody radiation or bremsstrahlung and emission from electronically excited SO. In [16], Flannigan and Suslick have observed very strong atomic emission from neutral Ar from weakly driven bubbles, and as the driving acoustic pressure is increased, the intensity of Ar emission lines decreases relative to the continuum emission. This observation is in complete agreement with the third effect mentioned above. Further, Flannigan and Suslick [17] have found experimental evidence for plasma generation during single-bubble cavitation, and as it is argued in [18], the shrinking of atomic line emission with increasing temperature may be due to rise in the temperature and strength

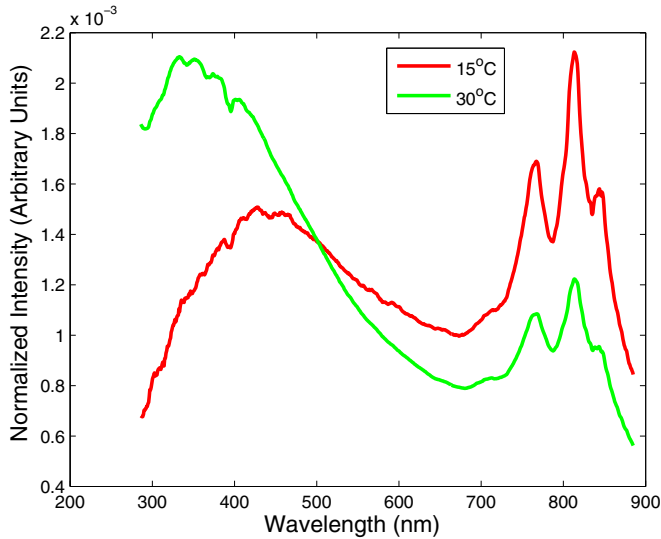


FIG. 4. (Color online) SBSL spectra of 85 wt% sulfuric for 15 °C and 30 °C. Ar emission lines are most prominent in 15 °C. Nevertheless, Ar emission lines are observed at all five temperatures: 15 °C, 20 °C, 25 °C, 30 °C, and 35 °C.

of the inner plasma with increasing acoustic intensity at the threshold.

In our experiment the major parameter affecting the intensity is the driving pressure, and for higher liquid temperatures, higher driving pressures were needed to produce active SBSL bubbles at the threshold (Fig. 3) and the intensity of Ar emission lines became lower, as shown in Fig. 4.

Further, in [16], Flannigan *et al.* used Ar emission lines to determine the bubble temperatures in sulfuric acid and found that the temperature of the Ar bubble increases with increasing acoustic driving pressure. Higher temperature implies higher UV emission from blackbody radiation or bremsstrahlung. This explains Fig. 4. Decrease in intensity of Ar emission lines implies an increase in temperature inside the bubble, which is in agreement with the theoretical predictions of bubble temperatures reported in [9].

In order to analyze variations within the SBSL spectra, as the temperature increases we have calculated total intensities in the intervals 200–400 nm (UV) and 400–800 nm (visible) for the five temperatures mentioned above. The results are shown in Fig. 5.

It is clearly evident that as the temperature of the liquid increases from 15 °C to 30 °C, the light emission in UV (200–400 nm) range steadily increases whereas light intensity in the visible range (400–800 nm) decreases. This behavior is completely opposite to what has been observed from SBSL in water [10]. However, when the temperature is higher than 30 °C, intensity in the UV decreases while the same in the visible range increases. Note that as mentioned earlier, the total intensity of SBSL emission measured with PMT also increases when temperature varies from 15 °C to 25 °C and remains constant above 25 °C. This may be because for higher temperatures more photons are emitted in UV and fewer photons are emitted in the visible frequencies. Since UV photons are more energetic, the total intensity increases with

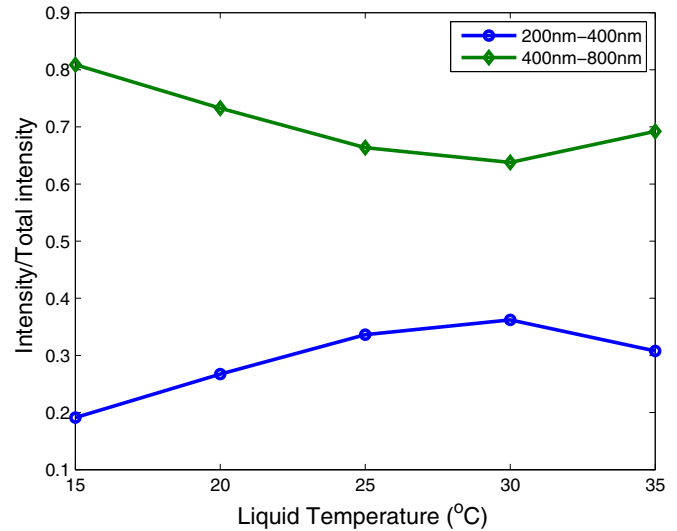


FIG. 5. (Color online) Normalized SBSL intensities in the intervals 200–400 nm (UV) and 400–800 nm (visible) as a function of temperature. Note that the total intensity of SBSL emission increases when temperature varies from 15 °C to 25 °C and remains constant above 25 °C.

the temperature. Then the question is why there are more photons produced when the liquid temperature is high. In order to answer this question, let us first consider SBSL in water. In the case of SBSL in water, contrary to what we have observed for sulfuric acid, the strong increase of light emission has been observed at low water temperatures rather than at higher temperatures [10]. This behavior has been explained in terms of increase in bubble stability [6] and lower vapor pressure [13] at low water temperatures. When the temperature is low, viscosity of water is relatively high and so is the stability. Hence it is possible to apply higher driving pressures and produce SBSL bubbles with higher bubble temperatures. This will result in generating a large number of UV photons, thereby producing strong light intensities.

For SBSL in sulfuric acid, the story is quite different. As the temperature increases, viscosity of sulfuric acid decreases [14]. The viscosity of sulfuric acid is high, and therefore the change in viscosity cannot significantly affect the bubble stability as in the case of water. The deciding factor which prevents us from reaching higher acoustic pressures was that the system becomes multibubble after passing a threshold value. Hence, for 85 wt% sulfuric acid, stability conditions have not played a major role in deciding the maximum intensity.

IV. CONCLUDING REMARKS

In this investigation, we were able to confirm the theoretical predictions made by Moshaii *et al.* regarding the temperature dependence of SBSL intensity in 85 wt% sulfuric acid solutions. Further, we observed that as the temperature increases, more UV photons and less visible photons contribute to the total intensity, indicating higher bubble temperatures at higher temperatures. This behavior is completely different from what has been observed for SBSL in water. Another important observation is that unlike SBSL in water, the

intensity threshold at each temperature is not determined by the stability conditions but by the driving pressure at which the transition from SBSL to MBSL takes place. Beyond 25 °C, SBSL intensity in 85 wt% sulfuric acid solutions became temperature independent.

ACKNOWLEDGMENT

This work was partially supported by the National Research Council of Sri Lanka under Grant No. 11-009. Authors would like to thank referees for their constructive criticisms and bringing their attention to some important references.

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- [1] B. P. Barber, R. A. Hiller, R. Lofstedt, S. J. Putterman, and K. R. Weninger, *Phys. Rep.* **281**, 65 (1997).
 - [2] M. P. Brenner, S. Hilgenfeldt, and D. Lohse, *Rev. Mod. Phys.* **74**, 425 (2002).
 - [3] C. G. Camara, S. J. Putterman, and E. Kirilov, *Phys. Rev. Lett.* **92**, 124301 (2004).
 - [4] K. Weninger, *J. Phys. Chem.* **99**, 14196 (1995).
 - [5] G. E. Vazquez and S. J. Putterman, *Phys. Rev. Lett.* **85**, 3037 (2000).
 - [6] S. Hilgenfeldt, D. Lohse, and W. C. Moss, *Phys. Rev. Lett.* **80**, 1332 (1998).
 - [7] K. Yasui, *Phys. Rev. E* **64**, 016310 (2001).
 - [8] D. J. Flannigan and K. S. Suslick, *Nature (London)* **434**, 52 (2005).
 - [9] A. Moshaii, S. Tajik-Nezhad, and M. Faraji, *Phys. Rev. E* **84**, 046301 (2011).
 - [10] R. Hiller, S. J. Putterman, and B. P. Barber, *Phys. Rev. Lett.* **69**, 1182 (1992).
 - [11] R. Urteaga, D. H. Dellavale, G. F. Puente, and F. J. Bonetto, *Phys. Rev. E* **76**, 056317 (2007).
 - [12] K. Yasui, A. Towata, T. Tuziuti, T. Kozuka, and K. Kato, *J. Acoust. Soc. Am.* **130**, 3233 (2011).
 - [13] M. Bradley, M. Ashokkumar, and F. Grieser, *J. Phys. Chem. B* **118**, 337 (2014).
 - [14] L. Gmelin and R. J. Meyer, *Gmelins Handbuch Der Anorganischen Chemie* (Verlag Chemie GmbH, Leipzig, 1985).
 - [15] S. D. Hopkins, S. J. Putterman, Brian A. Kappus, Kenneth S. Suslick, and C. G. Camara, *Phys. Rev. Lett.* **95**, 254301 (2005).
 - [16] D. J. Flannigan and K. S. Suslick, *Nature (London)* **434**, 52 (2005).
 - [17] D. J. Flannigan and K. S. Suslick, *Phys. Rev. Lett.* **95**, 044301 (2005).
 - [18] N. C. Eddingsaas and K. S. Suslick, *J. Am. Chem. Soc.* **129**, 3838 (2007).