Temperature and Pressure Dependence of Sonoluminescence

G. E. Vazquez and S. J. Putterman

Department of Physics and Astronomy, University of California at Los Angeles, Los Angeles, California 90095

(Received 10 September 1999; revised manuscript received 18 November 1999)

The dependence of sonoluminescence on ambient pressure and temperature is measured. As water is cooled, there occurs a 100-fold increase in light emission which can be accompanied by only slight changes in the ambient radius of the pulsating bubble. This suggests that water vapor trapped in the collapsing bubble is a key parameter for this system. For fixed concentration of gases in water, the maximum intensity of sonoluminescence decreases as the ambient pressure is lowered below 1 atm.

PACS numbers: 78.60.Mq, 42.65.Re, 43.25.+y, 47.40.–x

Sonoluminescence, the transduction of sound into light, occurs as a collapsing bubble of gas concentrates energy to a huge degree [1,2]. Since the energy densities are high enough to create broadband ultraviolet light, they are also high enough to create a plasma [3]. One of the routes to understanding sonoluminescence (SL) consists of determining the key macroscopic variables that control the light emission. These include the amplitude of the sound field that drives the bubble [4–6] and the partial pressure with which a particular gas (e.g., helium, argon, or xenon) has been dissolved into the water [7]. Our observations (Fig. 1) that bubbles with the same radii can have strongly different light emission depending on the ambient temperature T_0 of the surrounding water provides experimental evidence that water vapor content of the bubble is another key quantity controlling SL. This finding disagrees with all-encompassing explanations of SL [10] that are based entirely on the properties of the dissolved gas and ignore the role of water vapor. This finding also disputes explanations of the dramatic increase of SL intensity with decreasing T_0 [11] that invoke bubble shape instabilities [12]. According to these theories, increased light emission is entirely accounted for by increased values of the ambient and maximum bubble radii, and their ratio; $R_0, R_m, R_m/R_0$. At low temperatures, higher values of these parameters can be reached because destructive convolutions are more effectively damped out by the increased viscosity [12]. There exist a number of theories that claim an important role for water vapor [13–17]; this paper provides experimental results relevant to these theories.

Experiments were performed in a resonator with concentric quartz cylinders of different radii mounted between two stainless steel end caps. The central cylinder contained the host liquid, while the outer cavity, between the two pieces of quartz, was evacuated in order to inhibit condensation and improve temperature control. A mixture of ethylene glycol and water was circulated through the top and bottom end caps in order to facilitate cooling. The static pressure P_0 was controlled by placing the host liquid in a pliable gas tight bag (which acts as a volume reservoir) inside of a can which was pressurized to the desired value and connected to the experimental apparatus through a pressure line to form a complete gas tight system. The value of P_0 was recorded with an OMEGA liquid pressure gauge. Otherwise, previously developed techniques were used for (a) seeding a bubble into the sealed system [7], (b) measuring the bubble's radius as a function of time $R(t)$ by use of Mie scattering [6,18], (c) monitoring temperature, and (d) mixing and controlling the liquid gas solution [6]. These solutions are formed by mixing some gas or gas mixture into water under a pressure head measured in Torr (760 Torr = 1 atm). The acoustic mode used in this set of experiments has a velocity node at the center of the central cylinder at a frequency of 33.8 kHz at 20 °C. When this mode was excited at sufficient amplitude by piezoelectric drivers mounted on thinned regions of the top and bottom end caps, a light emitting bubble could be trapped. This frequency also changes by a few percent due to the temperature dependence of the speed of sound in water (1.485×10^5 cm/sec at 20 °C and 1.437×10^5 cm/sec at 0 °C).

The extreme sensitivity of SL to changes in temperature [11,19] and the independence of this effect to changes in bubble parameters are shown in Figs. 1–3. For bubbles formed from a 45 Torr gas-water mixture, where the gas is 1% argon and 99% nitrogen, a 20-fold increase in light emission is accompanied by changes in the ambient radius R_0 that are less than 20% and changes in the expansion ratio R_m/R_0 that are less than 5%. Similarly, at a fixed concentration of 300 Torr, decreasing the temperature results in a 200-fold increase in light emission, while the increase in R_0 is small by comparison; only 60%. In Fig. 3 one notes that a 20 °C bubble at 1.45 atm drive has a larger radius than a 0 °C bubble, yet it gives off 5× less light. Absolute radii have been assigned via a calibration technique discussed in Ref. [6], but it is important to note that the equality of the radii displayed in Fig. 1(a) is a property of the raw data independent of the calibration.

The role of viscosity in damping out bubble shape fluctuations [2,20–22] has been invoked as a basis for understanding the increase in light emission as the ambient temperature is lowered towards 0 °C [12]. Between 35 °C and 0 °C viscosity increases by a factor of 2.2 (note that

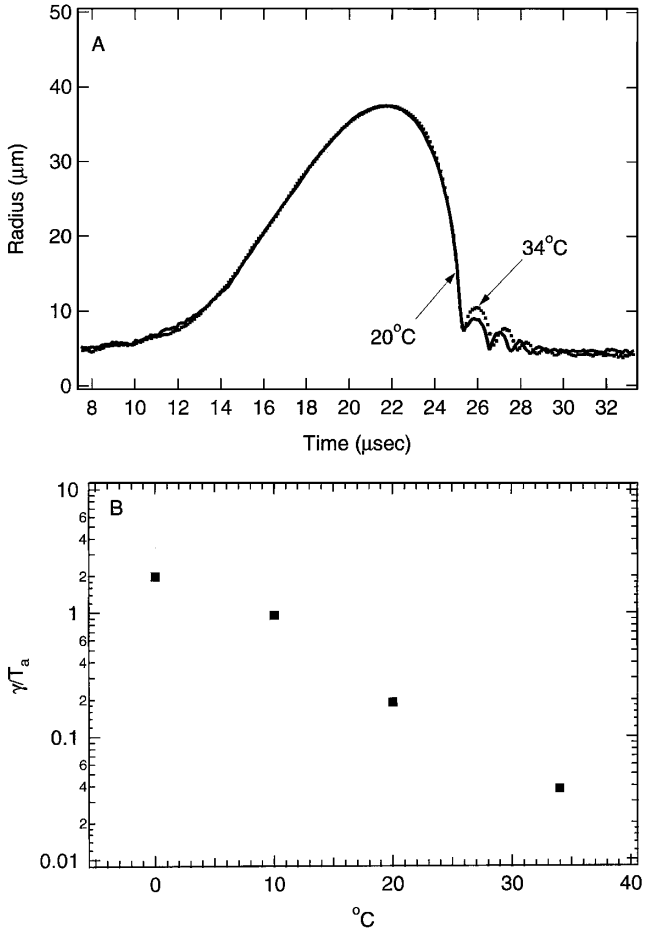

FIG. 1. (A) Radius as a function of time for a bubble formed from a 300 Torr solution of gas (1% argon, 99% nitrogen) in water at 20 and 34 °C. Despite the similar $R(t)$ the 20 °C bubble is 5× brighter. The acoustic drive is 1.30 atm at 20 °C and 1.32 atm at 34 °C, so light intensity increases as T_0 is lowered at fixed P_a. Supplemented with the observation [8,9] that flash width, at fixed concentration, is a monotonically increasing function of intensity leads to the conclusion that flash width increases as T_0 is lowered at fixed P_a and C_∞, in contrast with the conclusion of [9]. (B) Number of photons emitted per acoustic cycle γ/T_a as a function of temperature at fixed ambient radius $R_0 = 5\ \mu$m. The ambient radius is the radius of the bubble when the internal pressure is equal to the static pressure P_0. All graphs have been normalized to a bubble formed from 150 Torr 1% Ar in N_2 at 20 °C and 1 atm, which yields 3.0×10^5 photons per flash.

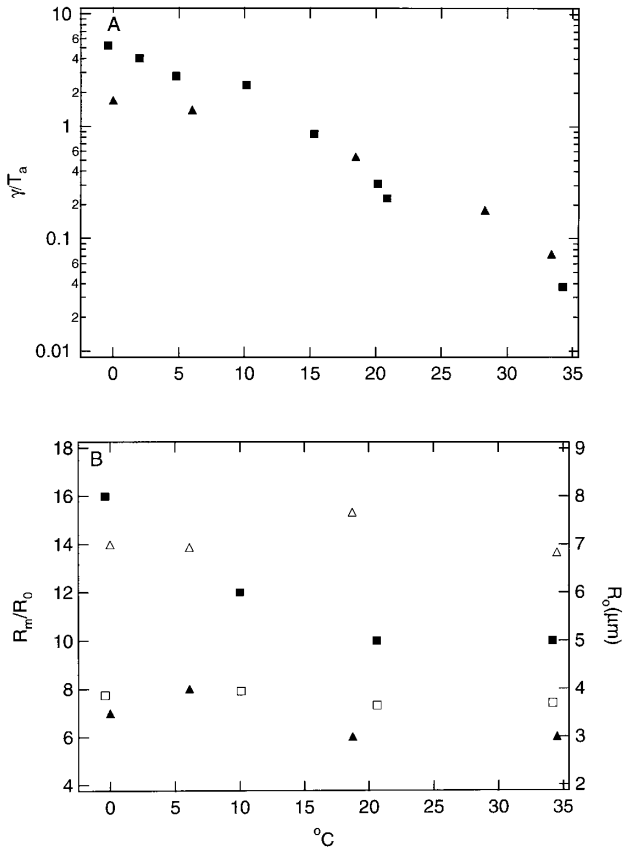

FIG. 2. (A) Maximum intensity of light emitted by a sonoluminescing bubble as a function of temperature for a mixture of 1% argon in nitrogen dissolved into water at room temperature at concentrations of 45 Torr (triangles) and 300 Torr (squares). As the temperature is varied the concentration remains fixed, while the percentage of saturation can change by about 60% since solubility is a function of temperature. Ambient pressure is one atmosphere. The resonant frequency for the centered trapping mode is 34.3 kHz at 35 °C, 33.8 kHz at 20 °C, and 31.9 kHz at 0 °C for pure water as the host liquid. (B) R_0 (filled) and expansion ratio R_m/R_0 (hollow) for the light emitting bubbles displayed in (A). The variations in R_m/R_0 observed in [19] are consistent with the reported factor of 2 variation in gas concentration that characterized different runs.

the water vapor pressure decreases by almost a factor of 9). According to these theories bubble-destroying convolutions are, at low temperature, displaced to a higher drive level where SL is greater. The observation that light emission can be significantly greater at a lower temperature for the same bubble dynamics questions the importance of shape instabilities as the physical phenomena that determine the strength of SL. To gain further insight into this matter, we have measured the properties of a room temperature bubble formed from a mixture of glycerin and water chosen to have the same viscosity as pure water at 0 °C. Figure 4 shows an overlay of the radius time curves for the glycerin mixture at its maximum drive level and a pure water bubble at 0 °C with a very similar $R(t)$, but below its maximum drive level. Although these bubbles have similar parameters, the glycerin bubble is dimmer by about a factor of 10. The inability of glycerin to improve SL from a single trapped bubble in a resonant sound field was first noted by Temple [23]; however, as shown in Fig. 3, glycerin does indeed increase the acoustic level at which a bubble can be driven.

According to Fig. 2b higher expansion ratios are observed at lower concentrations. This fact (along with the observation that the expansion ratio R_m/R_0 varies only slightly with temperature) is consistent with the perspective

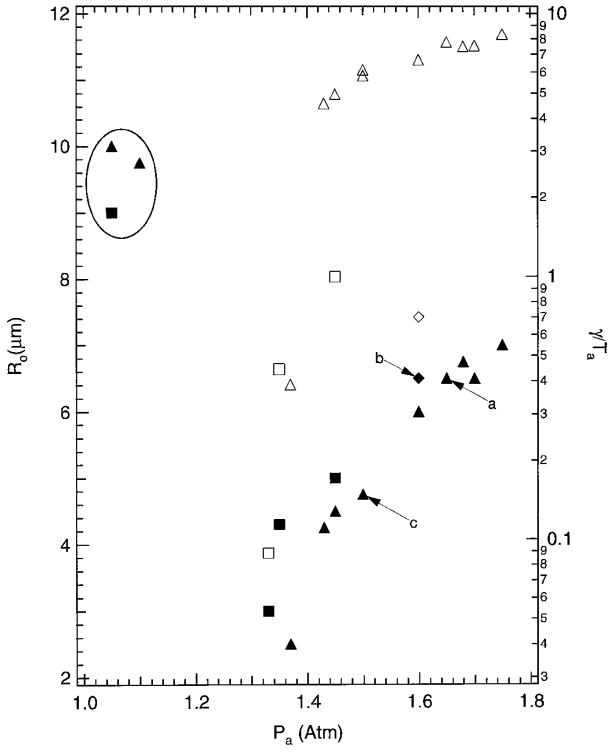

FIG. 3. R_0 (solid symbols) and γ/T_a (open symbols) as a function of drive level P_a for a 150 Torr 1% argon in nitrogen bubble at 20 °C (squares) and 0 °C (triangles). a, b, and c correspond to the curves shown in Fig. 4. The data contained in the oval correspond to non-light-emitting bubbles.

that nitrogen and oxygen are expelled from bubbles formed from mixtures with noble gases [24–26]. In this circumstance the expansion ratio is determined by the

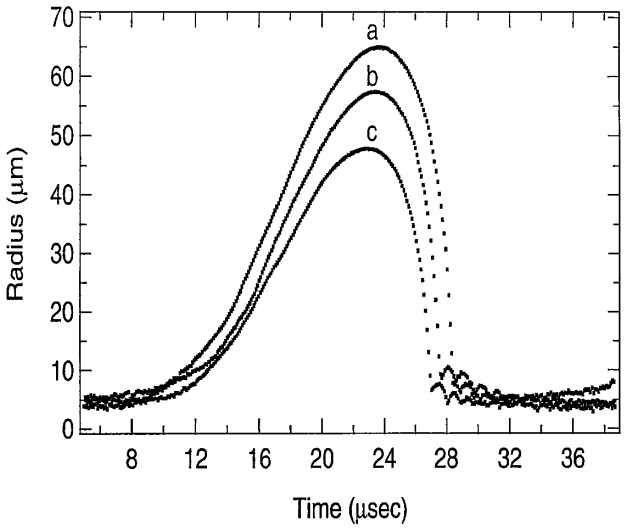

FIG. 4. Brightest bubble in 20% by wt glycerin/water at 20 °C (curve b, diamond Fig. 3) and pure water (curves a and c at 0 °C; see Fig. 3). Both systems are 150 Torr of 1% argon in nitrogen. The cycle-to-cycle motion of a bubble in the glycerin appears to be as stable for bubbles in pure water. Note also that the maximum drive at 0 °C is 17% higher than at 20 °C and one atmosphere.

equation of mass diffusion applied to the concentration of argon relative to saturation [27]:

$$\frac{c_\infty}{c_0} \approx 3\left(\frac{R_0}{R_m}\right)^3.$$

The sensitivity of SL to temperature is in part mirrored by a sensitivity to pressure as can be seen from Fig. 5, which displays the maximum intensity of SL for a 300 Torr 1% argon in a nitrogen bubble as a function of static pressure at 5 °C and 20 °C. Pressures in excess of 1 atm do not improve the light emission remarkably, but lower pressures reduce the emission. According to the argon rectification hypothesis [24], a 300 Torr 1% argon bubble should be very similar to a 3 Torr 100% argon bubble in the steady state. While this is true at 1 atm, there is a clear deviation at higher pressures. It remains to be seen whether the deviation from this hypothesis can be parametrized in terms of the amount of nitrogen retained in the incomplete rectified steady state bubble [28].

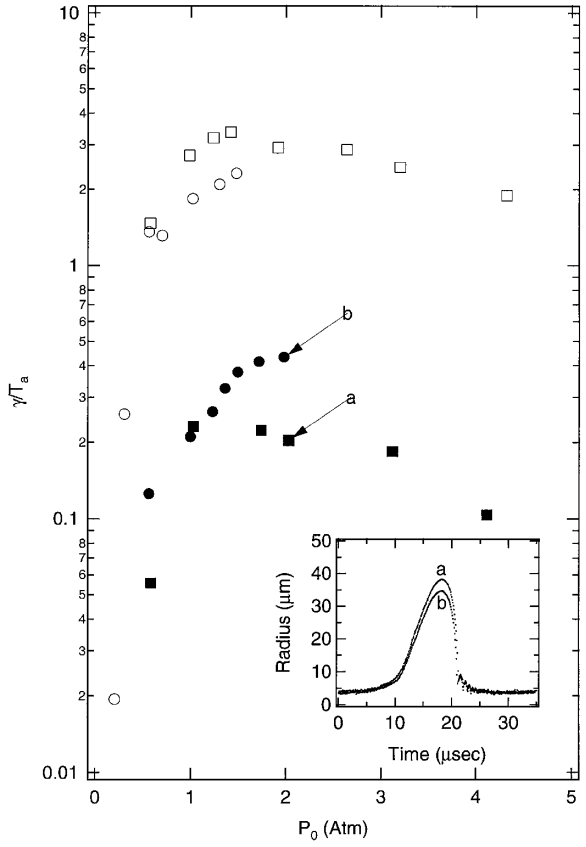

FIG. 5. Maximum intensity of SL as a function of static pressure. Data taken for pure argon bubbles dissolved at 3 Torr (circles) and 1% argon in nitrogen bubbles dissolved at 300 Torr (squares) 20 °C (solid symbols) and 5 °C (open symbols). The bubbles formed at or below 1/2 atm are very unstable and behave very similarly to bubbles formed in the SL liquid above 35 °C. The inset displays the two $R(t)$ curves for the respective intensities. Curve a corresponds to a drive level of 2.60 atm and curve b corresponds to a drive level of 2.45 atm.

Our observation that the maximum SL intensity decreases as the ambient pressure P_0 is lowered below 1 atm is consistent with the first observation of this effect [6], but disagrees with theory [16]. Our finding is also consistent with the claim [29] that the light emission from dim bubbles increases as P_0 is lowered at fixed drive amplitude P_a. Lowering P_0 at fixed P_a is equivalent to increasing P_a at fixed P_0, verifying the established result that SL increases with P_a [4–6].

We have reported a strong change in SL (as a function of temperature) that can be observed for slight changes in bubble dynamics. This suggests a sensitive dependence on an additional macroscopic variable. Although we have identified the hidden variable as the vapor content of the bubble [1,13–17,30], it should be noted that no specific marker for the water vapor has been detected. For instance, the excited OH line at 310 nm is absent from the spectrum measurements in the range of parameter space studied here. A qualitative argument can be made for the possible role of water vapor. The water vapor pressure at 20 °C is 17 Torr. This implies that only 2% of the equilibrium R_0 bubble is water vapor, but at R_m the number of water molecules exceeds argon by a factor of about 25. The off equilibrium dynamics of the Rayleigh collapse [31] could trap a portion of these molecules in the bubble's interior. If only 1% were trapped, the change in the minimum radius would be less than 50 nm, which is outside the range of experimental precision. The higher heat capacity of the water molecules would cool the interior [16] by almost a factor of 2; such changes in T can lead to large changes in light emission [32]. Experimental arrangements where the water vapor pressure is reduced would be of interest with regard to increasing SL. As discussed here, cooling is one such mechanism; perhaps other methods, such as adding salts, can further affect SL. Theories of the spectrum, flash width, and light-emitting mechanism must deal with the importance of water vapor in the bubble. An understanding of the means of action of the ambient temperature could yield a wide-ranging insight into the internal mechanism of SL.

We thank R. Löfstedt, P. H. Roberts, and Greg Swift for valuable discussions. We also thank K. R. Weninger for valuable advice regarding many aspects of this project, and the NSF (Division of Atomic Molecular and Optical Physics) and DARPA for their support.

[1] A. S. Walton and G. T. Reynolds, Adv. Phys. **33**, 595 (1984).
[2] S. J. Putterman and K. R. Weninger, Annu. Rev. Fluid Mech. **32**, 445 (2000).
[3] C. C. Wu and P. H. Roberts, Phys. Rev. Lett. **70**, 3424 (1993).
[4] B. P. Barber and S. J. Putterman, Nature (London) **352**, 318 (1991).
[5] D. F. Gaitan and L. A. Crum, J. Acoust. Soc. Am. **91**, 3166 (1992).
[6] B. P. Barber, R. A. Hiller, R. Löfstedt, S. J. Putterman, and K. R. Weninger, Phys. Rep. **281**, 67 (1997).
[7] R. Hiller *et al.,* Science **266**, 248 (1994).
[8] R. Hiller, S. J. Putterman, and K. R. Weninger, Phys. Rev. Lett. **80**, 1090 (1998).
[9] R. Pecha, B. Gompf, G. Nick, Z. Q. Wang, and W. Eisenmenger, Phys. Rev. Lett. **81**, 717 (1998).
[10] S. Hilgenfeldt, S. Grossman, and D. Lohse, Nature (London) **398**, 402 (1999).
[11] R. Hiller, S. J. Putterman, and B. P. Barber, Phys. Rev. Lett. **69**, 1182 (1992).
[12] S. Hilgenfeldt, D. Lohse, and W. C. Moss, Phys. Rev. Lett. **80**, 1332 (1998); **80**, 3164(E) (1998).
[13] K. Yasui, Phys. Rev. E **56**, 6750 (1997).
[14] R. D. Finch and E. A. Neppiras, J. Acoust. Soc. Am. **53**, 1402 (1973).
[15] W. C. Moss, D. B. Clarke, J. W. White, and D. A. Young, Phys. Lett. A **211**, 69 (1996).
[16] W. C. Moss, D. A. Young, J. A. Harte, J. L. Levatin, B. F. Rozsnyai, and G. B. Zimmerman, Phys. Rev. E **59**, 2986 (1999).
[17] A. J. Colussi and M. R. Hoffmann, J. Phys. Chem. A **103**, 11 336 (1999).
[18] B. P. Barber and S. J. Putterman, Phys. Rev. Lett. **69**, 3839 (1992).
[19] B. P. Barber *et al.,* Phys. Rev. Lett. **72**, 1380 (1994).
[20] C. C. Wu and P. H. Roberts, Phys. Lett. A **250**, 131 (1998).
[21] M. P. Brenner, D. Lohse, and T. F. Dupont, Phys. Rev. Lett. **75**, 954 (1995).
[22] S. J. Putterman and P. H. Roberts, Phys. Rev. Lett. **80**, 3666 (1998).
[23] P. R. Temple, M.S. thesis, The University of Vermont, 1970. Sonoluminescence from the gas of a single bubble. Also see S. J. Putterman, Phys. World **12**, 18 (1999).
[24] D. Lohse, M. P. Brenner, T. F. Dupont, S. Hilgenfeldt, and B. Johnston, Phys. Rev. Lett. **78**, 1359 (1997).
[25] D. Lohse and S. Hilgenfeldt, J. Chem. Phys. **107**, 6986 (1997).
[26] S. J. Putterman, Phys. World **11**, 38 (1998).
[27] R. Löfstedt, K. Weninger, S. J. Putterman, and B. P. Barber, Phys. Rev. E **51**, 4400 (1995).
[28] J. A. Ketterling and R. E. Apfel, Phys. Rev. Lett. **81**, 4991 (1998).
[29] M. Dan, J. D. N. Cheeke, and L. Kondic, Phys. Rev. Lett. **83**, 1870 (1999).
[30] Ya. B. Zeldovich and Yu. P. Raizer, *Physics of Shock Waves and High-Temperature Hydrodynamic Phenomena* (Academic Press, NY, 1967).
[31] Lord Rayleigh, Philos. Mag. **34**, 94 (1917).
[32] The role of temperature in improving cavitation luminescence has also been observed in systems where many bubbles are present [1], where the bubble is severely distorted from spherical shape [K. R. Weninger, H. Cho, R. A. Hiller, S. J. Putterman, and G. A. Williams, Phys. Rev. E **56**, 6745 (1997)], and where bubbles expand and collapse in a single pass through a Venturi tube [F. B. Peterson and T. P. Anderson, Phys. Fluids **10**, 874 (1967); K. R. Weninger, C. G. Camara, and S. J. Putterman, Phys. Rev. Lett. **83**, 2081 (1999)].