Sonoluminescence and high-pressure gas scintillators

W. Tornow
Department of Physics, Duke University, Durham, North Carolina 27708
and Triangle Universities Nuclear Laboratory, Duke Station, Durham, North Carolina 27708
(Received 17 November 1995)

We point out striking similarities between the light emitted in single-bubble sonoluminescence and high-pressure gas scintillators. This observation can account for important and largely unexplained phenomena surrounding sonoluminescence. Especially, it explains the observed dramatic effects of the noble gas content on the spectral density of the light emitted from a sonoluminescence bubble in water. Based on studies with high-pressure gas scintillators, we also propose an alternative explanation for the extreme temperature sensitivity of sonoluminescence.

PACS number(s): 47.55.−t, 78.60.Mq, 47.40.Nm, 42.65.Re

I. INTRODUCTION

Sonoluminescence (SL) is a nonequilibrium phenomenon which occurs when acoustic energy is focused on a bubble of air trapped in water such that the bubble emits light [1–4]. The light emission is visible to the unaided eye and appears blue. The acoustic energy (10^{-11} eV/atom) enters in a continuum at the macroscopic level and spontaneously focuses down to the molecular, atomic, and electron degrees of freedom, concentrating the ambient energy by more than twelve orders of magnitude [2,5].

The experimental arrangement for achieving stable single-bubble SL consists of piezoelectric transducers that excite breathing resonances in a water-filled spherical glass flask to which they are attached [6,7]. Above a certain threshold acoustic amplitude a bubble can be trapped. The transition to SL involves a sudden decrease in the bubble's size. The sound waves compress the air within the bubble (radius varies between a few 10^{-2} cm and a few 10^{-4} cm) to high pressures ($>10^4$ atmospheres), temperatures ($>10^4$ K), and densities [8]. The energy in the sound wave becomes highly concentrated so as to generate flashes of light. Light emission occurs only during an interval of less than 100 ps or 150 ps within each cycle in the sound field [2,5], which has a period of a few tens of microseconds (depending on the diameter of the glass flask). According to Ref. [9], the water acts like a piston that compresses and decompresses the air periodically and the collapse of a bubble formed by cavitation occurs such that the energy of collapse is delivered to a small number of molecules, which are thus excited or dissociated. Light is emitted in the deexcitation or recombination process. However, this simple model cannot account for the largely ultraviolet spectrum that has been observed [5]. Shock waves launched into the bubble's interior by the collapsing bubble may play an important role [9–11]. The temperature of 10^5 K required to explain the ultraviolet light emitted by the bubble suggests that an inward-moving shock wave remains intact to a radius of 2.5×10^{-4} cm from the center of the bubble. If the shock front survives down to 20 nm, the temperature would reach 10^6 K. However, the associated photons (soft x rays) do not propagate through water and therefore it is not clear what the highest temperature associated with the sonoluminescence really is [6]. According to Ref. [11] temperatures up to 1 million degrees appear to be feasible, thus opening the possibility of ''table-top fusion'' [4].

In the following, we will discuss important and largely unexplained observations which led us to believe that the light emission process associated with sonoluminescence is similar to the one that occurs in high-pressure gas scintillators which were studied extensively some 30 or 40 years ago. We will not try to explain the physics responsible for the transition from the non-SL regime to the SL regime and the energy transfer to molecules, atoms, and electrons in the bubble. Nevertheless, our observations may shed some light on the fascinating physics surrounding SL.

II. THE ROLE OF NOBLE GASES

According to Refs. [6,12], pure N_2 bubbles produced hardly any light. The same observation was made for O_2, an $80\%-20\%$ mixture of N_2 and O_2, and gas from a liquid-air container. It is the 1% presence of Ar in natural air that is responsible for the vast majority of the light emission. Subsequently, it was found that Ar can be replaced by other noble gases like He and Xe. The admixture of Xe (He) produces more (less) light than the admixture of Ar. The observed light emission depends strongly on the nature of the gas inside the bubble. For example, Xe yields a spectral peak at about 300 nm, whereas in the case of other noble gases the peak must be located further in the ultraviolet region, which is obscured by the cutoff of water [12].

The importance of noble gases for observing SL raised our suspicion of a possible relationship between the light emission processes in SL and high-pressure gas scintillators. In the following we will review some of the information necessary to support our conjecture.

The light output produced by charged particles in N_2-Ar gas mixtures at low pressure (<7 atm) was investigated by several authors. In the first studies reported by Grün and Schopper [13,14], which led to the development of gas scintillators, large scintillation pulse heights were obtained with N_2-Ar mixtures. The optimum N_2/Ar ratio depends on the total pressure and the type of photomultiplier tube (glass window or quartz window and spectral sensitivity of photocathode) used in the measurements. Measurements of optical

TABLE I. Energies (in eV) and associated wavelengths λ (in nm).

	para He	ortho He	Ne	Ar	Kr	Xe
1st metastable level	20.7	19.7	16.50	11.5	9.9	8.2
1st resonance level		21.1	16.55	11.59	10.0	8.4
2nd metastable level			16.60	11.61	10.5	9.41
1st ionization level	24.46		21.5	15.7	13.9	12.1
λ of resonance radiation	50.2	58.4	74.5	106.3	117.5	146.0

line and band emission from both atomic and molecular states in low-pressure N_2-Ar mixtures were performed by Grün [15], Bennett [16], Koch [17], and Birks [18]. N_2-Ar mixtures are well understood at low pressures. It is also well known that the admixture of O_2 quenches most of the light produced when charged particles traverse the gas volume.

High-pressure N_2-Ar and N_2-Xe mixtures were investigated by Engelke [19] and Tornow *et al.* [20]. It was found that the light output is independent of pressure at pressures above a few atmospheres. It was also observed that the measured light output can be increased significantly by using wavelength shifters like diphenyl stilbene or *p*-quaterphenyl, which were evaporated on the inner wall and the glass window of the gas scintillator housing. A wavelength shifter is capable of absorbing radiation of short wavelength and reemitting it in a region more appropriate for conventional phototubes. In Ref. [20], it was reported that a high-pressure N_2-Xe gas scintillator produces about a factor of 7 more light than an N_2-Ar gas scintillator at the same pressure. This observation is in qualitative agreement with the gas doping results of SL bubbles by Hiller *et al.* [12].

The relative scintillation efficiencies of noble gases and their mixtures were investigated by Northrop and Gursky [21,22]. Of all the noble gases, Xe produces the largest light output. The relative light output depends somewhat on the type of wavelength shifter and photomultiplier tube used in the measurements. It was found by Northrop and Gursky [21] and Henschel *et al.* [23] that a $10-15$ % admixture of He to Xe produces an even larger light output than observed for pure Xe. In striking similarity to the work of Refs. [21,22], Xe produced the largest light intensity in the SL studies of Ref. [12]. Therefore, we speculate that a 10% admixture of He to Xe will result also in a slightly larger light intensity than obtainable with Xe alone in single bubble SL.

Most of the light arising in a noble gas following the passage of a charged particle lies in the ultraviolet. Strickler and Arakawa [24] showed that pure Ar bombarded by α particles emits a continuum which extends from about 110 nm to 280 nm. The decay of the scintillation light has been reported to be as fast as 10^{-9} s with a decay period inversely proportional to pressure. The first excited state of nonionized and ionized noble gas atoms lies very high (i.e., at about two-thirds the energy required for ionization). Therefore, transitions from excited states to the ground state produce ultraviolet light. The energies of resonance levels of noble gas atoms from which the atoms are able to return directly to the ground state, without passing through intermediate excited states, are given in Table I [18]. According to this table, the statement made in Ref. [5] that the spectral peak of SL appears to be located at photon energies above 6 eV is not surprising. In fact, Table I explains why a spectral peak was observed in Ref. [12] for Xe and not for Ar and He. According to Table I, in the latter two cases the spectral peaks are expected to lie at a much shorter wavelength.

The speed of the scintillation light depends on the speed of their radiative transition and on the probability for reabsorption and subsequent reemittance (resonance radiation trapping). The lifetime of resonance levels of noble gas atoms or ions is $<10^{-9}$ s. It is well known that the effect of pressure broadening reduces the trapping time of resonance radiation at high pressures. In addition, considering the pressure and temperature associated with SL, the lifetime of the metastable levels given in Table I may be reduced by collisions with neutral or excited atoms or ions to the level of that expected for resonance radiation. Therefore, the subnanosecond time scale of SL is not too surprising. However, presently unknown mechanisms, most likely related to the high pressure in the bubble, must be responsible for the picosecond SL light pulses.

III. TEMPERATURE DEPENDENCE

In Ref. [5] it was found that by lowering the temperature from 20 °C to below 10 °C the total light emission from a single bubble can be enhanced by over a factor of 10. In a subsequent paper Barber *et al.* [10] reported that, as the water temperature decreased from 40 °C to 1 °C, the intensity of the light emission increased by a factor of over 200. In fact, at 1 °C the purple light emitted by the bubble was so bright that it could be seen by the unaided eye even in the presence of external lighting, but at 40 °C the SL was barely visible in a darkened room. According to Ref. [5] the increase in intensity of SL as the water is cooled is due to the fact that water dissolves about twice as much air at 0 °C than at 20 °C. In this case the collapse of a bubble is expected to be more violent, causing it to generate a greater energy focusing. However, in a more recent paper [25] it was argued that it now appears that the gas solubility in the driving fluid is not of key importance. The studies reported in Ref. [25] also suggest that SL is not sensitive to the vapor pressure of the liquid.

To establish the connection to gas scintillators we notice that they are very susceptible to poisoning of the light by contaminants, especially to organic substances like hydrocarbons contained in standard diffusion pump oil. Therefore, we speculate that the strong increase of light emission observed in SL with decreasing temperature is (similarly to gas scintillators) a direct consequence of the purification process accompanied with the temperature decrease. Not only is the partial vapor pressure of light absorbing contaminants in the

bubble and the surrounding driving fluid reduced, but the contaminants are also preferentially absorbed at the inner wall of the glass container where the temperature is lowest. Of course, the dramatic temperature dependence reported for SL has no parallel in present-day high-pressure gas scintillators. In the latter case one fills an evacuated ($\approx 10^{-6}$ Torr) container with highly purified gas and special care is taken with respect to O-rings, cold fingers, and vacuum pumps. Although an extremely clean gas filling system consisting of a turbo-molecular pump, viton and metal O-rings, cold fingers, etc., was used in Ref. [20], a small increase in light emission was still observed in pure Ar and N_2 and N_2-Ar gas mixtures when the temperature was decreased from 20 °C to −8 °C. Of course, the temperature dependence can easily be enhanced by adding tiny amounts of organic impurities. In fact, in our early studies of gas scintillators preceding the work of Ref. [23], we never managed to establish a stable light output over periods of days. The buildup of vapor pressure due to the presence of impurities caused a continuous poisoning of the gas and resulted in an approximately exponential decline of the light intensity over time due to quenching. Only after the impurity issue was under control were we able to obtain a stable light output over periods of months or even years.

IV. DEPENDENCE OF SONOLUMINESCENCE ON THE DRIVING FLUID

Until very recently, single bubble SL had not been observed in liquids other than water (Refs. [26,6]). Weninger *et al*. [25] reported the first observation of sonoluminescence from single bubbles in nonaqueous liquids. In an earlier paper Barber *et al.* [2] referred to a 25% solution (by volume) of glycerine in water. With low-viscosity silicon oil, Barber *et al.* [10] have trapped bubbles in the non-SL regime. However, a transition to the SL regime was not observed in Ref. [10]. In the work of Weninger *et al*. [25], a xenon bubble was trapped in driving fluids like *n*-dodecane, 1-pentanol, 1-propanol, ethanol, etc. As compared to air in water, these new systems behave very differently in many ways. However, in agreement with our expectation based on experience with gas scintillators, the importance of xenon and the strong temperature dependence of the SL intensity is a common feature. Xenon was found to be the most suitable gas for the nonaqueous driving fluids. Again, we speculate that the Xe-He mixture referred to above may work even a little bit better than pure Xe. The explanation for the temperature dependence is the same as stated above for the water-gas system.

V. CONCLUSIONS

In summary, the existing experimental information suggests that the light emission processes associated with SL are closely related, if not identical, to the ones known to occur in high-pressure gas scintillators. In fact, we think that the small SL-bubble trapped in water acts like a high-pressure micro-gas-scintillator.

ACKNOWLEDGMENTS

This work was supported in part by the U.S. Department of Energy, Office of High-Energy and Nuclear Physics under Contract No. DE-FG05-91-ER40619.

[1] D. F. Gaitan, and L. A Crum, in *Frontiers of Nonlinear Acoustics*, edited by M. Hamilton and D. T. Blackstok, Proceedings of the 12th International Symposium on Nonlinear Acoustics (Elsevier, New York, 1990), p. 459.
[2] B. P. Barber and S. J. Putterman, Nature **352**, 318 (1991), and references therein.
[3] L. A. Crum, Phys. Today **48** (9), 22 (1994), and references therein.
[4] L. A. Crum and R. A. Roy, Science **266**, 233 (1994), and references therein.
[5] R. Hiller, S. J. Putterman, and B. R. Barber, Phys. Rev. Lett. **69**, 1182 (1992).
[6] S. J. Putterman, Sci. Am. **Feb.**, 46 (1995).
[7] R. A. Hiller and B. P. Barber, Sci. Am. **Feb.**, 96 (1995).
[8] B. P. Barber and S. J. Putterman, Phys. Rev. Lett. **69**, 3839 (1992).
[9] C. C. Wu and P. H. Roberts, Phys. Rev. Lett. **70**, 3424 (1993).
[10] B. P. Barber, C. C. Wu, R. Löfstedt, P. H. Roberts, and S. J. Putterman, Phys. Rev. Lett. **72**, 1380 (1994).
[11] R. Pool, Science **266**, 1804 (1994).
[12] R. Hiller, K. Weninger, S. J. Putterman, and B. P. Barber, Science **266**, 248 (1994).
[13] A. E. Grün and E. Schopper, Z. Naturforsch. **6a**, 698 (1951).
[14] A. E. Grün and E. Schopper, Z. Naturforsch. **9a**, 134 (1954), and references therein.
[15] A. E. Grün, Z. Naturforsch. **9a**, 55 (1954).
[16] W. R. Bennett, Jr., Ann. Phys. **18**, 367 (1962).
[17] L. Koch, J. Phys. Rad. **21**, 169 (1960).
[18] J. B. Birks, *The Theory and Practice of Scintillation Counting* (Pergamon Press, Oxford, 1964), Chap. 14.
[19] C. E. Engelke, I. R. E. Trans. Nucl. Sci. **NS-7**, 32 (1960).
[20] W. Tornow, H. Huck, H. J. Köber, and G. Mertens, Nucl. Instrum. Methods. **133**, 435 (1976).
[21] J. A. Northrop and J. C. Gursky, Nucl. Instrum. Methods **3**, 207 (1958).
[22] J. A. Northrop, J. M. Gursky, and A. E. Johnsrud, I. R. E. Trans. Nucl. Sci. **NS-5**, 81 (1958), and references therein.
[23] G. Henschel, G. Mack, and W. Tornow, Nucl. Instrum. Methods **57**, 257 (1967).
[24] T. D. Strickler and E. T. Arakawa, J. Chem. Phys. **41**, 1783 (1964).
[25] K. Weninger, R. Hiller, B. P. Barber, D. Lacoste, and S. J. Putterman, J. Chem. Phys. **99**, 14 196 (1995).
[26] R. Löfstedt, K. Weninger, S. Putterman, and B. P. Barber, Phys. Rev. E **51**, 4400 (1995).