

Single-bubble sonoluminescence from noble gas mixtures

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Light emission intensities from single-bubble sonoluminescence at room temperature, water dissolved with admixtures of pairs of noble gases, He-Ar and He-Xe, were measured as a function of mixing ratio to search for effects of species segregation occurring during the violent bubble collapse. The observed relative effect on the light emission intensity is surprisingly greater in He-Ar than in a He-Xe mixture.

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Sonoluminescence is an interesting energy focusing phenomenon, in which extremely short pulses of light are emitted synchronously with a collapsing gas bubble trapped in a liquid-filled resonator cavity driven at high acoustic intensity. It has attracted numerous experimental and theoretical studies [1]. However, a clear physical understanding of the processes behind the light emission is still lacking [2]. A complicating feature of the effect is that the gas bubble contains a varying number of species of gas molecules during the very short collapse. Owing to high pressure and temperature present within the bubble, a variety of chemical reactions, ionization and other effects are thought to occur. It has been established that the presence of a noble gas in the bubble is crucial to the stable production of high light emission intensity [3]. A simulation study has suggested that the different molecular species in the bubble segregate spatially during the collapse [4]. Species segregation can affect the heat loss rate near the bubble wall, the internal gas temperature and the light emission, as the thermal conductivity and the ionization energies change. We present systematic experimental studies of the single-bubble sonoluminescence (SBSL) when two noble gases are dissolved into sample water at room temperature.

The diffusive mass fluxes of the species in the SBSL gas mixture is a sum containing terms proportional to the gradients in concentration, pressure, and temperature [4,5]. The proportionality constants that enter the sum are dependent on the masses of the species and hence segregation of species with different masses can occur [6]. A simulation study [7] of SBSL including molecular segregation effects indicates that even a “mild” segregation significantly affects the maximum temperature attained and the emitted light intensity. Another simulation study [8] shows that, while the noble gas rectification hypothesis is valid, a substantial fraction of the material at the time of collapse is made up of water vapor and its reaction products. It has become important then to carry out experimental studies and search for manifestations of material segregation on the sonoluminescence phenomenon. Little is known about the temperature and pressure gradients present in the sonoluminescing bubbles. Probing segregation effects might provide clues on the dynamics in the bubble interior.

Our SBSL apparatus is similar to a common design [1]. The acoustic resonator is a cylindrical quartz tube (inner diameter=75 mm, outer diameter=80 mm, and length=80 mm), whose ends are closed with flat plates made of

brass. A pair of piezoelectric transducers (PZT's) are epoxied onto the end plates. The resonator is connected to a small but flexible ballast volume to maintain the water pressure at the ambient atmospheric pressure. A bubble is made by applying a short reproducible pulse of current into a heater mounted on the bottom end plate. The working frequency is 17.3 kHz. The temperature of the water is monitored by observing the resonant frequency as well as a thermister attached to one of the end plates. The resonator temperature is regulated within $\pm 0.15^\circ\text{C}$ by a Peltier effect cooler attached to both the top and bottom plates. The SBSL light emission is detected by the induced current in a photodiode (UDT sensor, model UV-100) [9]. The photodiode current is proportional to the time and wavelength averaged light emission intensity. It is measured with an electrometer. Mie scattering measurements are taken with a fast integrated monolithic photodiode/amplifier chip (OPT210, Burr-Brown) by detecting the scattered light from a He-Ne laser beam incident on the bubble at an angle θ from the incident beam direction.

The sample water is prepared as follows in a nearly identical manner to dissolve various gas mixtures. A 1.6 liter volume of deionized water is sealed into a stainless steel tank and is degassed by pumping on its vapor phase for two hours while agitating with a magnetic stirrer. A gas mixture containing nitrogen (with partial pressure P_{N_2}), argon (P_{Ar}) or xenon (P_{Xe}), and ^4He (P_{He}) gases is prepared in a separate can and is allowed into the tank containing degassed water. The total pressure ($=P_{\text{N}_2}+P_{\text{Ar}}+P_{\text{He}}$) is fixed at 150 Torr for all mixtures in this paper. The total noble-gas fraction is kept constant at 1.0%, i.e., $P_{\text{Ar}}+P_{\text{He}}=P_{\text{Xe}}+P_{\text{He}}=1.5$ Torr. The mixture gas is dissolved into water while stirring for another two hours at room temperature [10].

Just prior to filling, the resonator is flushed with N_2 gas several times to remove remnant water from previous run and then it is evacuated to check for air leak. Subsequently, the mixed water is filled into the evacuated resonator in about 15 min and then the water pressure is brought up with N_2 in about 3 min to equalize the ballast volume with the ambient pressure. The short time required for the N_2 pressurization does not significantly change the N_2 content in the water. When measurements of one mixture is completed, the water is totally emptied from the resonator and refilled with another water sample.

After the sample water is filled, the temperature regulation system is turned on. It requires some two hours for the

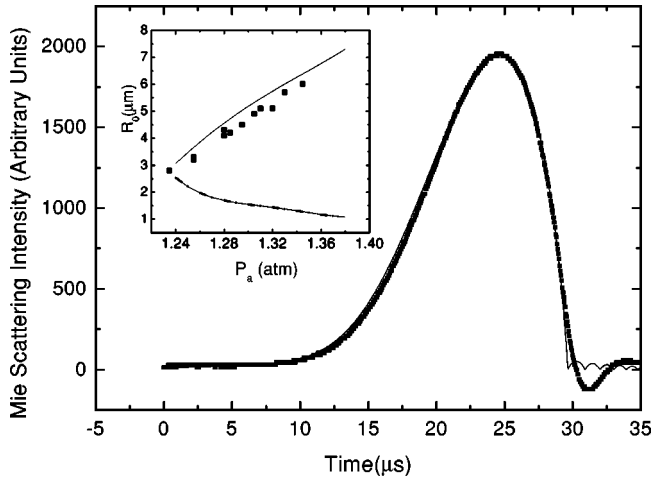


FIG. 1. An example of Mie scattering data (dots) and fit [line with P_a (acoustic drive pressure amplitude) = 1.26 atm and R_o (ambient radius) = 3.8 μm] for $x=0.15$ for the He-Xe mixture. The inset shows the extracted R_o - P_a relationship when the drive level is varied. The line in the inset is the R_o - P_a relation calculated from the diffusive equilibrium condition.

water temperature to settle down to the desired regulated temperature as verified by monitoring the resonance frequency. After the temperature regulation has kicked in, the acoustic coupling between the PZT's and the resonator is checked for reproducibility by measuring the frequency response. The working resonant frequency remains constant within ± 5 Hz during the experiment. The location of the levitated bubble is checked carefully. The position of the photodiode for measuring the SL intensity is adjusted (at most 1 mm) to account for slight changes in bubble location. The Mie scattering photodiode is placed on a rotating arm, whose position can be reproduced within $\pm 0.5^\circ$.

Adequacy of our degassing process is checked several times during the course of experiment by observing the sonoluminescence light emission intensity from water, which is dissolved with only N_2 . The bubble in this pure N_2 sample is unstable with a maximum light emission intensity which is 10% of that with $P_{\text{Ar}}=1.5$ Torr. This is comparable to the result ($\approx 3\%$) seen by Barber *et al.* [1] and demonstrates that our degassing is adequate.

Important parameters affecting the bubble dynamics are the amplitude of the driving acoustic pressure, P_a , and the ambient radius R_o . The Mie scattering measurement is commonly used to determine P_a . The sensor area spans an angular opening $\theta=80^\circ \pm 2^\circ$. At this value of θ , the Mie scattering intensity is closely proportional to the square of the time dependent bubble radius [11,12]. An example of our raw Mie scattering data (dots) is shown in Fig. 1, which displays a portion of the acoustic cycle, where slow expansion and the contraction takes place. The time interval of very rapid collapse and the subsequent after-bounce oscillation cannot be tracked owing to the detector time constant limitation. The interval of rapid change in $r(t)$ is not crucial for the purpose of determining P_a and R_o .

The time dependent radius $r(t)$ of a bubble (trapped in water under the ambient pressure P_o driven by a sinusoidal

acoustic pressure, $P=P_a \sin \omega t$) is governed by the Rayleigh-Plesset equation [1], $\rho[\ddot{r}r + (3/2)\dot{r}^2] = p_g - (P_o + P) + (r/c) \times (\dot{p}_g - \dot{P}) - 2\sigma/r - 4\eta\dot{r}/r$, where p_g is the gas pressure in the bubble, ρ the mass density, c the speed of sound, σ the surface tension ($=0.0725$ N/m), and η the viscosity ($=1.0 \times 10^{-3}$ N s/m²) of water. For the purpose of fitting data over time intervals excluding the collapse range, $r < R_o$, the temperature of the gas in the bubble can, to a good approximation, be taken constant: $p_g = P_o(R_o - b)^3 / (r - b)^3$ with $b = R_o/9$. A best fit to the data is obtained by adjusting R_o and P_a in the Rayleigh-Plesset equation and is shown by a line in Fig. 1. The fitted line is indistinguishable from the data, almost all of the time interval except around the collapse and the subsequent after-bounce oscillation interval. This portion of the data is ignored in the fitting. The fitting procedure is used to determine P_a and R_o as the drive level applied to the PZT is varied. The inset to Fig. 1 shows the relation between R_o and P_a , so determined for the bubbles which show sonoluminescence. The fitted R_o vs P_a is reproducible from one mixture to another within the scatter seen in the inset. The fitted R_o was checked with an absolute calibration method using the lobe pattern evolution [13] as a function of θ and was found to be consistent with each other within 5%. The P_a - R_o relation expected from the noble gas rectification hypothesis and establishment of diffusive equilibrium [14] is shown by the line in the inset in good agreement with our fits. Similar quality of fit is obtained in all dissolved gas mixtures presented in the paper. The observed relation between R_o and P_a is independent of the relative ratio between P_{He} with P_{Ar} and P_{Xe} . The relation is apparently set by the total noble gas content [15] which is fixed in our experiment. That the bubble dynamics is insensitive to mixtures is in accord with the simulation study [4].

For each gas mixture, a systematic measurement of the sonoluminescence light emission intensity, I_{SL} and Mie scattering data are taken as a function of the acoustic drive level. The measured I_{SL} is plotted as a function of P_a in Fig. 2 for two samples: (a) $P_{\text{He}}=0.23$ Torr and $P_{\text{Xe}}=1.27$ Torr and (b) $P_{\text{He}}=P_{\text{Ar}}=0.75$ Torr. A lower threshold in P_a must be exceeded before sonoluminescence turns on. Once the lower threshold is exceeded, a linear increase in I_{SL} is observed with a characteristic slope up to an upper threshold in P_a , where the bubble disintegrates and dissolves back into water. The linear relation between I_{SL} and P_a has also been seen by others [16]. All mixtures, we studied, showed a behavior similar to those in Fig. 2. The threshold drive levels depend slightly on the particular bubble observed, while the slope is more reproducible. We choose the slope of least squares linear fit to data in Fig. 2 to characterize the light emission from each mixture. The change in water temperature (which affects the solubility, surface tension, and viscosity) between Figs. 2(a) and 2(b) have negligible effects on the diffusion curve in Fig. 1.

Only the total energy emission can be estimated since we measure only the integrated light emission intensity. Roughly estimating average sensitivity of the photodiode (its maximum being 532 mA/W at $\lambda=850$ nm, falling to half at 500

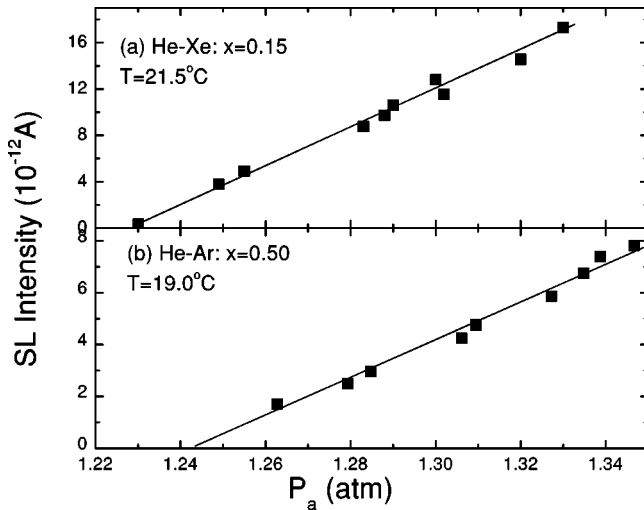


FIG. 2. Photodiode current (\propto SL emission intensity) as a function of acoustic drive amplitude: (a) He-Xe mixture with $x=0.15$ and water temperature = 21.5°C . (b) He-Ar mixture with $x=0.50$ and water temperature = 19.0°C . The lines are least squares fits.

and 1000 nm), the total energy output from the light emission is about 1×10^{-12} J/cycle. This is close to other estimates [1].

The slope in the I_{SL} vs P_a plots are shown in Fig. 3 for all the mixtures we have measured as a function of $x = P_{\text{He}}/(P_{\text{He}} + P_{\text{Xe}})$ or $P_{\text{He}}/(P_{\text{He}} + P_{\text{Ar}})$ [17,18]. If instead, the mixtures are characterized by the light emission intensity at a particular acoustic pressure, the observed dependence on x does not change. The measurements were carried out at 21.5°C and 19.0°C in all mixtures of He-Xe and He-Ar, respectively. The ratio of the slope of $x=0$ sample to that of $x=1$ sample for He-Ar (He-Xe) measurements is 1.5 (3.8). These values are consistent with the light emission ratios in

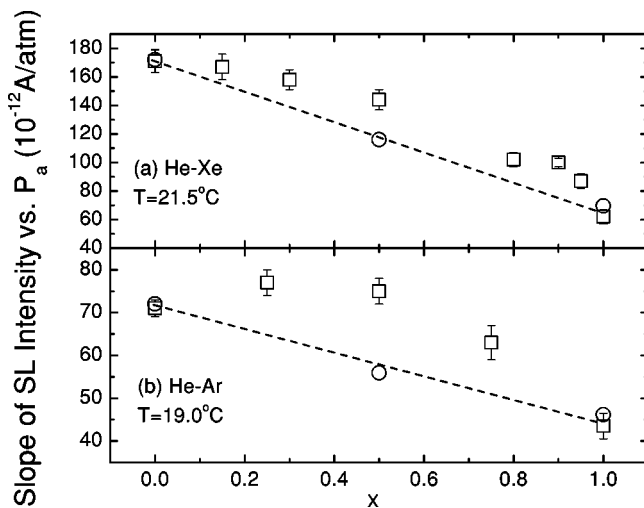


FIG. 3. The slopes of the least squares linear fits in Fig. 2 as a function of the He molar noble gas fraction, x : (a) He-Xe mixtures and (b) He-Ar mixtures. The dashed lines are the linear interpolation between end points and represent a simple extreme segregation of the noble gases in the SL bubble. Circles are the sum of intensities from Ref. [1]. See text for discussion.

the respective gases measured Barber *et al.* [1]. Near $x=0$, the emission intensity decreases slowly in He-Xe mixtures and it slightly increases in He-Ar mixtures as the He fraction is increased. On the other hand, when a small number of either Xe or Ar is added to He near $x=1$, the light emission increases rather rapidly. The net effect is the observed bulge in the slope of light emission intensity near $x=0.5$. It should be pointed out that the observed behavior as a function of x in Fig. 3 does not arise from the changes in the volume or the number of emitters. Since the total noble gas fraction is kept constant, the R_o - P_a relation is the same for all mixtures [17].

It is interesting to compare our data with what is expected from simply adding the SL emission intensities from the constituent noble gases in the mixtures. To do this, we take the blackbody spectral density measurements as reported by Vazquez *et al.* [19], for each noble gas (n) with its own blackbody temperature (T_{nBB}), multiply it by wavelength dependent transmission coefficient of water and the quartz tube and by the responsivity of our detector [9], and finally sum over wavelength between 200 nm and 700 nm to arrive at the integrated SL intensity, \bar{I}_n . The magnitudes of \bar{I}_n are scaled such that they match our measurements of the slopes for samples with “pure” noble gas. The expected intensity from mixtures is $\bar{I} = (1-x)\bar{I}_n(T_{nBB}) + x\bar{I}_{\text{He}}(T_{\text{HeBB}})$, where x is the fraction of He gas and n is either Ar or Xe. The evaluated intensities are shown by the dashed lines in Fig. 3. The linear interpolations are expected if each noble gas acted independently without interaction; the two gases in the mixture would have two different blackbody temperatures. Our experiment shows that the mixtures produce light emissions similar to but greater than the expectation from the noble gases having independent blackbody temperatures.

Another simple comparison can be made with the existing data on the maximum light emission intensity from mixtures of 150 Torr N_2 and a single noble gas (see Fig. 22 of Ref. [1]). Their maximum intensities with 1% noble gas are normalized to our pure Ar and Xe slope data. The circles at $x=0.5$ are the sums of the intensities (normalized by the same factors) of 0.5% each of the constituent noble gases. The sum of the separately measured intensities of single noble gases turns out to be very similar to the extreme segregation dependence and our data again are different from the sums.

The above naive picture ignores the effects of the water vapor and its reaction products present in possibly large quantities in the bubble at the time of light emission [8]. The amount of water vapor displaced by He would depend on x and this can affect the light emission intensity. The effect of admixing He is surprisingly greater in He-Ar than in a He-Xe mixture. Though not clear, this might be an indication that water vapor plays a more important role in He-Ar owing to the closeness of the molecular weight of water to that of Ar. Addition of He would displace the water molecules nearer towards the bubble wall. In the He-Xe mixture, Xe being the most massive would still remain nearer to the bubble wall. Experimental evidence for the importance of water vapors has also been reported [20,21]. It is not yet known how the integrated light intensity is related to the temperature in the

bubble interior. The maximum temperature reached depends on a host of possible effects including species segregation, chemical reactions, and shock wave formation. Our experimental studies of light emission from noble gas mixtures may be combined with further theoretical studies to shed light on sonochemistry effects [22,23], where the dispersion of water vapor owing to the segregation of noble gas mixtures alters the chemical yield of OH and H₂O₂. Indeed, the observed and simulated dependence of chemical yields on mixture fraction is reminiscent of our light emission data. Our studies of SBSL provide another tool for probing the sonochemical effects.

An interesting extension to our studies is the measurement of the spectral radiance. The blackbody spectrum is observed from a submicrometer region of bubble with only one dissolved noble gas by Vazquez *et al.* [19], who found the ratio of the ionization energy to the fitted blackbody temperature to be a constant, independent of any particular noble gas dissolved in water. Whether the mixture bubbles emit with

blackbody radiation spectrum is an interesting question. In the case of an extreme segregation model, in principle, a bimodal spectrum is expected. If the gases distribute homogeneously, the spectrum would be a blackbody radiation with single characteristic temperature. Another extension is to measure the segregation effects near 0°C to eliminate the complications related to the presence of water vapor. If segregation indeed takes place, the spatial gradient so generated in speed of sound can have a large influence on the formation of shock [24]. Experiments with noble gas mixtures would then allow exploration of more extreme conditions of temperature and pressure.

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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] S.J. Putterman, P.G. Evans, G. Vazquez, and K. Weninger, *Nature (London)* **409**, 782 (2001).
- [3] R. Hiller, K. Weninger, S.J. Putterman, and B.P. Barber, *Science* **265**, 248 (1994).
- [4] B.D. Storey and A.J. Szeri, *J. Fluid Mech.* **396**, 203 (1999).
- [5] J. O. Hirschfelder, C. F. Curtiss, and R. B. Bird, *Molecular Theory of Gases and Liquids* (Wiley, New York, 1954).
- [6] S. Chapman and T.G. Cowling, *The Mathematical Theory of Non-Uniform Gases* (Cambridge University Press, London 1970).
- [7] K. Yasui, *Phys. Rev. E* **63**, 035301(R) (2001).
- [8] B.D. Storey and A.J. Szeri, *Phys. Rev. Lett.* **88**, 074301 (2002); B.D. Storey and A.J. Szeri, *Proc. R. Soc. London, Ser. A* **456**, 1685 (2000).
- [9] UDT Sensors, Inc., Hawthorn, CA.
- [10] Variations in room temperature during mixing leads to at most +0.015/-0.15 Torr uncertainty in the total dissolved noble-gas pressure. This uncertainty does not affect the qualitative variation of light intensity as a function of mixture ratio shown in Fig. 3.
- [11] D.F. Gaitan, Ph. D. thesis, University of Mississippi, 1990 (unpublished).
- [12] M. Honevac, software package *LightLab: Far Field Mie Scattering* (Valley Scientific, Valley City, OH, 1998).
- [13] W.J. Lentz, A.A. Atchley, and D.F. Gaitan, *Appl. Opt.* **34**, 2648 (1995).
- [14] S. Hilgenfeldt, M.P. Brenner, S. Grossman, and D. Lohse, *J. Fluid Mech.* **365**, 171 (1998).
- [15] J.A. Ketterling and R.E. Apfel, *Phys. Rev. E* **61**, 3832 (2000).
- [16] D.F. Gaitan and R.G. Holt, *Phys. Rev. E* **59**, 5495 (1999).
- [17] A dynamic diffusive equilibrium is independently established between each constituent noble gas in the bubble and that dissolved in water. The segregation is likely to take place over a short time compared to the acoustic period and it does not affect this diffusive equilibrium [4]. It is then reasonable that the mixture fraction as applied during the mixing procedure is the mixture fraction in the bubble itself. In evaluating x , the differences in the diffusivity and solubility of different noble-gas species do not enter. Since the total noble-gas content is kept constant at 1% of the total dissolved gas, the R_o - P_a relation is the same for all mixtures in agreement with our observation.
- [18] The entire acoustic apparatus was reassembled between the two sets of measurements (a) and (b) in Fig. 3. The reassembly is the likely cause for the systematic difference in the data of “pure” He in the two datasets.
- [19] G. Vazques, C. Camara, S. Putterman, and K. Weninger, *Opt. Lett.* **26**, 575 (2001). They find $T_{HeBB}=20,400$ K, $T_{ArBB}=13,100$ K, and $T_{XeBB}=8000$ K.
- [20] G.E. Vazquez and S.J. Putterman, *Phys. Rev. Lett.* **85**, 3037 (2000).
- [21] R. Toegel, B. Gompf, R. Pecha, and D. Lohse, *Phys. Rev. Lett.* **85**, 3165 (2000).
- [22] G. Mark *et al.*, *Ultrason. Sonochem.* **5**, 41 (1998).
- [23] B.D. Storey and A.J. Szeri, *Proc. R. Soc. London, Ser. A* **457**, 1685 (2001).
- [24] V.Q. Vuong, A.J. Szeri and D.A. Young, *Phys. Fluids* **11**, 10 (1999).