

Single Bubble SL at Ambient Conditions

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Abstract: Single bubble sonoluminescence (SL) explained by a high pressure and temperature plasma produced in an adiabatic bubble collapse is shown to be unphysical. In the alternative, SL may be simply explained at ambient conditions by the quantum electrodynamics (QED) frequency up-conversion of electromagnetic (EM) radiation. The bubble is treated as a QED cavity having high EM resonance while water molecules in the bubble wall are Planck oscillators emitting low frequency thermal radiation. During collapse as the oscillators move to the QED cavity surface, their thermal radiation is suppressed only to be conserved by producing EM radiation at the resonant frequency of the QED cavity. In support of SL at ambient conditions, a microwave radiometer experiment is presented.

1 INTRODUCTION

Recently, claims [1] were made of the *measurement* of high pressure, density, and temperature in sonoluminescence (SL) bubbles. Two techniques were reported: (1) plasma diagnostics applied to Ar emission lines, and (2) light scattering measurements of bubble 'radius v time'. For dim and bright bubbles, pressures of 1500 and 3700 bar including temperatures of 15,000 K were claimed. In fact, the claims in [1] are shown to arise from unphysical assumptions made in the *calculation* – not *measurement* - of pressure and density. In the alternative, an SL theory and supporting experiment is presented showing SL is produced under ambient conditions.

2 DISCUSSION

Contrary to the claims in [1] there are no *measurements* of high pressure and temperature in a collapsing SL bubble. Since the 1930's, the microscopic size of the SL bubble has precluded such *measurements*. Then as now, there are only *calculations* of pressure and temperature based on assumptions of how the bubble gases respond to the bubble collapse.

Indeed, the adiabatic compression of SL bubble gases is usually assumed in *calculations* of the high SL pressure and temperature plasma. Even higher pressure and temperature plasmas were claimed by assuming the shock in the collision of the liquid bubble walls occurs in the bubble gas. In fact, the claim of SL temperatures of 100 million degrees [2] is based on *calculations* that show a bubble filled with condensable water vapor increases to unphysical high pressure and temperature instead of condensing to liquid at ambient conditions during collapse.

Calculations of SL bubble gas pressure and temperature including condensation [3] are very sensitive to the accommodation coefficient [4] that quantifies how efficiently water vapor condenses. Simulations [5] that claim to include water vapor condensation assume accommodation coefficients of 0.4 to obtain temperatures from 5,000 to 20,000 K. However, if the accommodation coefficient is taken to be unity, as it should, simulations [6] show SL temperatures and pressures of the water vapor to remain near ambient conditions during collapse. Hence, the unphysical pressure and density [1] *calculated* from the volume changes based on 'radius v time' data are artifacts of unphysical accommodation coefficients.

Moreover, the SL bubble gas pressure and density *calculated* in [1] from broadening of spectral lines assume the SL emission occurs in the gas phase. Spectral broadening [7] in the liquid phase is well-known, but should SL occur in the bubble wall the unphysical gas phase is not necessary to explain spectral broadening.

Paradoxically, the unphysical claims [1] actually support SL occurring in the liquid bubble wall under ambient conditions, but how is this possible?

One explanation is QED induced EM radiation [3]. The bubble is treated as a QED cavity having an extent beyond the wall surfaces that includes the penetration depth of standing resonant EM radiation. During collapse, the water molecules that eventually fill the bubble are treated as Planck oscillators having low frequency thermal kT radiation at wavelengths greater than 100 microns, but the QED cavity always has higher EM resonance. During bubble collapse, the low frequency oscillators from the bulk that enter the penetration depth are then physically located inside the high EM resonant frequency QED cavity, and therefore their thermal radiation is suppressed. Under QED constraints, the suppressed thermal radiation may only be conserved by frequency up-conversion, the lowest available frequency inside the QED cavity being its instant EM resonant frequency. In this way, bubble collapse produces a continuous broadband SL spectrum from the infrared (IR) through the visible (VIS) to the vacuum ultraviolet (VUV).

By this theory, the SL bubble is a broadband continuous frequency IR to VUV laser that excites *all* chemical species that enter the penetration depth of the QED cavity wall during bubble collapse. Any broadening of SL spectral lines occurs because the emission occurs in the liquid state of the bubble wall consistent with [8, 9].

3 EXPERIMENT

3.1. Theory

Radiometer theory [10] is based on the flux of EM radiation escaping through a small hole of a enclosure housing a blackbody source and is quantified by the brightness B_{bb} expressed in terms of power per unit area per unit bandwidth per unit solid angle,

$$B_{bb} = \frac{2h\nu^3}{c^2 \left[\exp\left(\frac{h\nu}{kT}\right) - 1 \right]} \text{ W / m}^2 - \text{Hz} - \text{ster} \quad (1)$$

where, h is Planck's constant, ν is frequency, c is the speed of light, k is Boltzmann's constant, and T is absolute temperature.

$$\text{For } h\nu \ll kT, \quad \exp(h\nu/kT) - 1 \approx h\nu/kT$$

$$\text{And } B_{bb} = \frac{2k\eta T_p}{\lambda^2} \quad (2)$$

where, T_p is the physical temperature, and η is the emissivity of the surface, typically $\eta \sim 0.3$.

3.2. Analysis

In the microwave radiometer [11] experiment, the antenna power P_{antenna} is,

$$P_{\text{antenna}} = A_{\text{em}} S \quad (3)$$

where, A_{em} is the antenna aperture area, and S is the Poynting vector. At distance R from the SL bubble,

$$A_{\text{em}} = \frac{4\lambda^2}{\pi^3} \quad \text{and} \quad S = e^{-\alpha R} \frac{P_{\text{bubble}}}{4\pi R^2} \quad (4)$$

Combining,

$$P_{\text{antenna}} = e^{-\alpha R} \frac{4\lambda^2}{\pi^3} \frac{P_{\text{bubble}}}{4\pi R^2} \quad (5)$$

In water [11] at ambient temperature for the center frequency $\nu = 2$ GHz of a 1 GHz bandwidth, the attenuation coefficient $\alpha = 0.8 \text{ cm}^{-1}$. For $R = 2$ cm between the antenna and bubble, $e^{-\alpha R} = 0.2$. The central frequency wavelength is $\lambda = c/\nu = 0.15$, but the refractive index n in water is, $n = 9$. So, $\lambda = 0.017$ m.

$$P_{\text{antenna}} / P_{\text{bubble}} = 1.5 \times 10^{-3} \quad (6)$$

Brightness at the antenna is based on the *measurement* of antenna power. The minimum detectable power is 2 pW, i.e., the actual antenna power $P_{\text{antenna}} < 2$ pW. The corresponding brightness B_{bb} is,

$$B_{bb} = P_{\text{antenna}} \left(\frac{1}{1 \times 10^9 \text{ Hz}} \right) \left(\frac{1}{A_{\text{em}} \text{ m}^2} \right) \left(\frac{1}{4\pi \text{ ster}} \right) \quad (7)$$

where, $A_{\text{em}} = 3.7 \times 10^{-5} \text{ m}^2$. Thus, $B_{bb} < 4.3 \times 10^{-18} \text{ W / m}^2 - \text{Hz} - \text{ster}$.

The antenna temperature T_{antenna} is,

$$T_{\text{antenna}} = \frac{B_{bb} \lambda^2}{2k\eta} \quad (8)$$

Thus, $T_{\text{antenna}} < 150$ K.

Similarly, the bubble temperature T_{bubble} depends on the power P_{bubble} . From Eqn. 6, $P_{\text{bubble}} < 1.3$ nW. Substituting P_{bubble} for P_{antenna} in Eqn. 7 gives the bubble brightness $B_{bb} < 2.8 \times 10^{-15} \text{ W / m}^2 - \text{Hz} - \text{ster}$. By Eqn. 8, the bubble temperature T_{bubble} is,

$$T_{\text{bubble}} < 97,500 \text{ K}$$

Clearly, this is consistent with the claim [1] of temperatures from 5,000 to 20,000 K.

However, the bubble temperature is lower because of differences in: (1) the data for absorption coefficient of water in the GHz region, and (2) the expression for the antenna aperture area A_{em} .

Absorption and Refractive Index Data

With regard to the absorption coefficient α of water in the microwave region at 2 GHz, the *calculated* value (Fig. 1 of [11]) shows $\alpha = 0.8 \text{ cm}^{-1}$. However, the *measured* value [12] is, $\alpha = 0.36 \text{ cm}^{-1}$. Thus, the fraction of microwave radiation reaching the antenna 2 cm away is $e^{-\alpha R} = 0.48$.

In [11] the 15 cm wavelength at the 2 GHz center frequency in water was reduced by the refractive index $n = 9$ giving $\lambda = 1.7$ cm. A review of the literature shows the index $n = 9$ is a reasonable.

Gain of Effective Aperture Area

The conical horn [11] has an effective aperture area A_{em} given [13] in terms of its directivity D_o by,

$$A_{\text{em}} = \frac{\lambda^2}{4\pi} D_o \quad (9)$$

The gain of a conical horn is optimum when its diameter d_m is,

$$d_m = \sqrt{3l\lambda} \quad (10)$$

where, l is the length along the side of the conical horn. The length L of the horn along its axis makes an angle ψ with the side of the horn. Numerically, the horn in [11] has $L = 2$ cm and $\psi = 20^\circ$, and therefore $l = L/\cos\psi = 2.12$ cm and $d_m = 3.29$ cm.

The directivity D_o of a conical horn in units of decibels (dB) having an aperture efficiency e_{ap} is,

$$D_o \text{ (dB)} = 10 \log_{10} \left[e_{\text{ap}} \frac{4\pi}{\lambda^2} \left(\frac{\pi d_m^2}{4} \right) \right] \quad (11)$$

For $e_{\text{ap}} = 0.51$, $D_o = 12.8$ dB, or $D_o = 10^{\text{dB}/10} = 19.1$. Thus,

$$A_{\text{em}} = \frac{\lambda^2}{4\pi} D_o = 4.3 \times 10^{-4} \text{ m}^2$$

Eqn. 5 is modified,

$$P_{\text{antenna}} = e^{-\alpha R} \frac{\lambda^2}{4\pi} D_o \frac{P_{\text{bubble}}}{4\pi R^2} \quad (12)$$

Combining,

$$P_{\text{antenna}} / P_{\text{bubble}} = 0.041 \quad (13)$$

Again, brightness at the antenna is based on the minimum detectable power $P_{\text{antenna}} < 2$ pW. For $A_{\text{em}} = 4.3 \times 10^{-4} \text{ m}^2$, Eqn. 7 gives the antenna brightness $B_{\text{bb}} < 3.7 \times 10^{-19} \text{ W / m}^2 \cdot \text{Hz} \cdot \text{ster}$. Eqn. 8 gives temperature $T_{\text{antenna}} < 12.9 \text{ K}$. From Eqn. 13, the bubble power is $P_{\text{bubble}} < 48$ pW. In Eqn. 7 substituting P_{bubble} for P_{antenna} with $A_{\text{em}} = 4.3 \times 10^{-4} \text{ m}^2$ gives the bubble brightness $B_{\text{bb}} < 8.9 \times 10^{-18} \text{ W / m}^2 \cdot \text{Hz} \cdot \text{ster}$. Thus, Eqn. 8 gives the bubble temperature near ambient,

$$T_{\text{bubble}} < 310\text{K} \quad (14)$$

4 RELATED ISSUES

4.1. LTE in Plasma

In [1] the claims of a high pressure and temperature plasma in SL are supported by *calculations* that rely on local thermodynamic equilibrium (LTE). In plasma, there are collisional and radiative distributions of energy [14] that may not be in LTE with each other.

The radiative energy governed by the Planck expression for the brightness is given by Eqn. 1. Since radiative equilibrium requires the plasma to be optically thick at all frequencies, radiative equilibrium is most likely out of balance with collisional equilibrium. The criterion for LTE is that collisional processes must be much more important than radiative, so that overall the lack of radiative equilibrium does not matter. This is equivalent to requiring that the excited state must have much larger probability of de-excitation by an inelastic collision than by spontaneous radiation. For this to occur, the plasma is required to have a high electron number density N_e . The criterion [14] for LTE is,

$$N_e \gg 1.5 \times 10^{12} T^{1/2} (\Delta E)^3 \text{ cm}^{-3} \quad (15)$$

where, T is the electron temperature in K, ΔE is the Planck energy in eV between a state and any other state to which it can make transitions.

In [1] the SL spectra showed the Ar+ line emission that is $\Delta E = 37 \text{ eV}$ above the Ar $3p^6$ ground state. At 10,000 K, the number density $N_e = 3.8 \times 10^{17} \text{ cm}^{-3}$. However, Eqn. 15 requires $N_e \gg 7.6 \times 10^{18} \text{ cm}^{-3}$, and therefore claims of SL bubble temperatures from 5,000 to 20,000 K assuming LTE are unjustified.

Moreover, the SL bubble emission is well-known [11] to occur in 100 ps or less. In such cases, it is necessary [14] to estimate whether there is sufficient time for collisional equilibrium to be established. Typically, full LTE in plasma at 10,000 K would take

about 1 microsecond for $N_e \sim 10^{16} \text{ cm}^{-3}$. Since time to establish equilibrium varies inversely with N_e , about 10 ns is required for equilibrium in the SL plasma. Even so, electron temperatures are not in LTE on the time scale of 100 ps.

SL bubble gas temperature or the gas kinetic temperature *calculated* by assuming an adiabatic bubble collapse is not likely equal to the electron temperature to justify claims of 5,000 to 20,000 K. Typically, gas kinetic equilibrium for electrons is established [14] in about 10 ns. In [1] it is stated that the LTE exists between the Ar atoms, but not between the electrons. Indeed, it is questionable that LTE even exists between the Ar atoms as the 10 ns required for LTE is not possible in the SL pulse of 100 ps.

4.2. LTE at Ambient Conditions

SL at ambient conditions is unequivocally in LTE. But radiative processes in producing the broadband IR to VUV spectrum are only optically thin as otherwise the SL spectrum could not be measured, and therefore LTE does not exist. But overall LTE is dominated by the thermal kT energy of a large number of Planck oscillators that fill the bubble during collapse, of which only a very small fraction produces the SL spectrum from the IR to the VUV. In a 30 micron radius bubble, the total thermal kT energy of oscillators at ambient temperature is 16 μJ while the SL pulse producing 30 mW in 100 ps utilizes only 3 pJ. Since the radiative loss is a fraction of the total kT energy, LTE is justified.

At ambient conditions, the probability of de-excitation by an inelastic collision of the excited Ar+ state in the bubble wall is extremely unlikely compared to the spontaneous QED induced EM radiation emitted as the oscillators enter the penetration depth of the collapsing QED cavity

4.3. Line Broadening

It is stated [1] that when SL spectrum broadenings no longer allows line emission analysis, the 'R v t' curve can still be used for the quantitative determination of the pressure. Nothing in pressure induced broadening of spectral lines justifies this statement. In principle, there should not be any limit to the amount of line broadening unless the broadening is caused by another mechanism. If so, pressure determination by line emission analysis is simply not applicable.

The SL broadband spectrum is a natural consequence of QED induced EM radiation produced as the bubble collapses and has nothing to do with pressure broadening. In fact, the QED cavity effectively is a continuous broadband IR to VUV laser that excites any chemical species in the water surroundings that moves into the penetration depth during bubble collapse. Broadening of Ar spectral lines occur because the Ar atom is excited by QED induced EM radiation in the liquid state of the bubble wall. Thus, *calculations* of pressure based on the assumption that SL occurs in the gaseous phase of the bubble are unphysical.

5 CONCLUSIONS

In SL the importance of microwave radiometry is that the *measurement* of antenna power extended by *calculation* to the bubble shows the temperature to be near ambient, thereby avoiding the need to explain SL with claims of unphysical pressure, density, and temperature. Although confirmation of the effective aperture area of the conical horn antenna remains, there is no doubt that further microwave radiometry tests of single bubble SL will be performed in the future.

Claims of SL plasma based on *calculations* that assume LTE at pressures from 1500 to 3700 bar and temperatures from 5,000 to 20,000 K over the 100 ps time scale whether by (1) plasma diagnostics of SL line broadening, or (2) volume changes by 'R v t' data are simply unphysical. In contrast, LTE for SL at ambient conditions is unequivocal as de-activation of excited Ar⁺ states by collision at ambient temperature is unlikely compared to QED induced spontaneous emission.

But there is yet an even simpler argument against in SL by a high pressure and temperature plasma. The high electron number density necessary to justify LTE means that a large number of electrons are moving within the plasma of the bubble, and therefore the fact that microwaves were not detected in SL means there simply are not only no electrons in the plasma, but there is no high pressure and temperature plasma.

Credible SL theories require the light to be produced under ambient conditions. QED induced EM radiation [3] is only one such SL theory and others should be explored.

6 REFERENCES

- [1] D. J. Flannigan, S. D. Hopkins, C. G. Camara, S. J. Putterman, and K. S. Suslick, "Measurement of Pressure and Density inside a Single Sonoluminescing Bubble," *Phys. Rev. Lett.*, 96, 20431(2006).
- [2] S. J. Putterman, B. P. Barber, R. A. Hiller, and R. Lofstedt, US Patent 5,659,173 (1997).
- [3] T. Prevenslik, "Sonoluminescence at Ambient Temperature?" See www.geocities.com/sonoluminescence2007/SLrev5.pdf
- [4] The accommodation coefficient is defined as the probability that a water vapor molecule sticks to a liquid surface, and is required to be unity for vapor condensing inside a bubble.
- [5] M. P. Brenner, S. Hilgenfeldt, and D. Lohse, "Single bubble sonoluminescence," *Rev. Mod. Phys.*, 74, 425-452 (2002).
- [6] T. Prevenslik, *Ultrasonics*, "The cavitation induced Becquerel effect and the hot spot theory of sonoluminescence," 41, pp. 312 (2003).
- [7] B. Myers, "Molecular Electronic Spectral Broadening in Liquids and Glasses," *Annu. Rev. Phys. Chem.*, 49, pp. 267-95 (1998).
- [8] M. Ashokkumar and F. Grieser, "Sono photoluminescence from aqueous and non-aqueous solutions," *Ultrasonics-Sonochemistry*, 6, 1-5 (1999).
- [9] A. Troia, D. M. Ripa, S. Lago, and R. Spagnolo, "Evidence for liquid phase reactions during single bubble acoustic cavitation," *Ultrasonics-Sonochemistry*, 11, 317-21 (2004).
- [10] E. Nyfors and P. Vainikainen, *Industrial Microwave Sensors*, Artech House, 1989.
- [11] J. N. Kordomenos, M. Bernard, and B. Denardo, "Experimental microwave radiometry of a sonoluminescing bubble," *Phys. Rev. E*, 29, 1781-4 (1998).
- [12] D. J. Segelstein, "The complex refractive index of water," MS Thesis, University of Missouri, Kansas City, 1981.
- [13] C. A. Balanis, *Antenna Theory – Analysis and Design*, J. Wiley & Sons, New York, 1982.
- [14] A. P. Thorne, *Spectrophysics*, Chapman and Hall & Science Publishers, London, 1974.