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# Towards higher energy density processes in sonoluminescing bubbles

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A bubble in a standing sound wave in water pulsates with such power that, as its minimum radius is approached, the interior undergoes a first order phase transition to a dense plasma. A pulse of blackbody radiation is emitted with temperatures ranging from 6,000K to 20,000K depending on the gas inside the bubble. To date experiments on sonoluminescence inside water, sulfuric acid and phosphoric acid have yielded similar blackbody temperatures. These liquids are hydrogen bonded and so the question arises as to whether their compressibility limits the energy concentration achieved by Sonoluminescence. Liquids where repulsion between nearest neighbor electron shells \such as small ion molten slats\ should be more incompressible. Scaling law estimates of the energy loss due to: acoustic radiation; shear viscosity, and compressibility of the surrounding fluid will be discussed with the goal of predicting a fundamentally new regime of sonoluminescence.



# **1. INTRODUCTION**

Nature likes to concentrate acoustic energy. A bubble much smaller than the wavelength of a pressure wave in a fluid can concentrate that energy so as to emit an ultraviolet flash of light. The parameter space for sonoluminescence [SL] is amazing. It has been observed in one-off implosions of an isolated mm sized bubble [1,2,3] and from nanometer sized regions in water that is forced to randomly cavitate at 10.Mhz [4]. From 40Hz[5] to 40kHz[6,7] a single bubble can be locked synchronously to a standing wave acoustic drive so that there is one precisely timed flash of light for each cycle of sound. In all of the synchronous cases the emission is blackbody and depending on the gas content of the bubble [Hydrogen[8], noble gases [9] water vapor [1]] and the fluid [Water, Phosphoric Acid [10], Sulfuric acid [11]] the spectral temperature ranges from 5,000K to 20,000K. A blackbody is opaque which requires the gas inside the bubble to be a plasma with an extremely high charge density over  $10^{20}$  free charges/cc. This has been measured by Suslick [12,13] and demonstrated by using an SL bubble to block a laser[14]. The inferred charge densities are orders of magnitude higher than follow from Saha's equation [15] and are due to new plasma equations of state that apply to the regions of parameter space that were made observable via SL[5]. When these densities and temperatures are achieved in laser breakdown[16] and electric discharges [17] similar lowering of the ionization energy is observed. The sphericity of picosecond pulses of synchronous SL have also been used for calibrating the transit time spread of arrays of photo detectors used for high energy physics experiments[18].

The energy of a photon relative to the volume from which it is emitted [ $\sim eV/A^3$ ] is 12 orders of magnitude larger than the vibrational energy of a molecule of water in which a standing wave is excited with a dynamic amplitude of 1.3atm. Such a standing wave is strong enough to drive synchronous SL. So the nonlinear processes which generate SL concentrate energy density by 12 orders of magnitude. An abiding question is: what are the limits of energy density concentration that can be achieved by the processes of continuum mechanics that give rise to SL? Can the energy density be boosted by an additional factor of 100-1,000 so that the energy density for a hydrogenic bubble is close to that required for nuclear fusion? Even if SL leads to a highly inefficient source of neutrons it would nevertheless be very interesting because a desktop thermonuclear neutron generator does not exist.

# 2. ENERGY CONCENTRATION and BUBBLE DYNAMICS

The energy available for concentration is the work  $4\pi R_m^3 p_{\infty}/3$  supplied to expand a bubble to its maximum radius  $R_m$  against an external pressure  $p_{\infty}$ . Figure 1 shows a synchronously driven bubble passing through it maximum radius at time 34.µs. The rarefaction phase of the sound field has caused this Argon bubble to expand to  $R_m$  from its ambient radius  $R_0$  [=4.5um], which would be its size in the absence of sound. The expansion by a factor of 10 of the micron sized bubble is driven by the 5.6cm wavelength sound wave shown as a dashed line in the figure. The work done on this Argon bubble amounts to an energy  $(R_m^3/R_0^3)kT_0$  =25eV per Argon atom that is available for delivery to the bubble's interior when it collapses to its minimum radius [at ~37.5µs] to form a plasma and emit a flash of light. The highest energy photons observed have an energy of about 6eV and ionization and transport processes will also soak up the available energy.



Figure 1: The radius of an Argon bubble during a cycle of sound with a frequency of 26.5kHz. The ambient radius is 4.5.µm and it expands to 10 times that value. The dynamic pressure of the sound wave is shown by the dashed line and measurements of bubble radius by the dots. The argon bubble forms from a bubble that is initially air [19.20].

#### **3-ENHANCING SONOLUMINESCENCE WITH IMPLODING SHOCKS**

One route to higher energy density is to figure out how to achieve larger values of  $R_m / R_0$ .

Another route is based upon a key insight of Wu and Roberts [21]. They note that the above analysis assumes a bubble interior that has uniform density. However, prior to the emission of a flash of light the wall of the bubble is collapsing supersonically. Under certain conditions an imploding shock wave could form in the gas. Such a shock wave could run through the already compressed gas and in the processes of focusing through r=0 further concentrate the energy density.

At the maximum radius the external pressure acting on the bubble is  $p_{\infty}$ . In Figure 1,  $p_{\infty}=1.35$  atm. Initially the gas in the bubble is greatly expanded and exerts little back pressure so that the bubble at  $R_m$  starts to collapse as though it is an empty cavity. An empty cavity in an incompressible fluid of density  $\rho_0$  was shown by Rayleigh to collapse to zero radius in a

time  $t_0 = .915\sqrt{\rho_0 R_m^2 / p_\infty}$ . During the final stages of collapse the velocity of the bubble wall is  $\dot{R} = -\sqrt{\frac{2p_\infty}{3\rho_0}} \{\frac{R_m}{R}\}^{3/2}$ . This value becomes faster than the speed of sound in ambient

argon when R=R<sub>m</sub>/11 and faster than the speed of sound in water when R=R<sub>m</sub>/30. The ultimate limiting factor to the collapse is the backpressure in the bubble which has a contribution from the van der Waals hard core at R= a=R<sub>m</sub>/90. Wu and Roberts found that an imploding shock is created inside the bubble when  $\dot{R}/c_0 \sim 1$ , and that the focusing of the shock to the origin led to a

temperature of about  $10^7$  K. In a remarkable thesis [2] spanning theory and experiment Ramsey points out a limitation of this calculation. The pressure at points 'r>R' in the fluid close to the

bubble wall before back pressure builds up is  $p = \frac{1}{2}\rho \dot{R}^2 \{\frac{R}{r} - \frac{R^4}{r^4}\}$ . The maximum pressure

in the fluid is  $p_{mf} = .16 p_{\infty} \frac{R_m^3}{R^3}$  which occurs at r=1.6R. So when the collapse velocity

reaches  $c_0$ ;  $p_{mf} = 5,000.atm$  which is greater than the yielding stress B in the Tait equation

of state for water:  $\frac{p+B}{B} = (\frac{\rho}{\rho_0})^{\Gamma}$  where B= 3000atm and  $\Gamma$ =7. Hunter [22] evaluated the

correction to the motion of a bubble wall surrounding an empty cavity that incorporates the compressibility of water into the nonlinear Euler equations. Whereas the incompressible limit gives  $R = A_0 (t - t_0)^{2/5}$  Hunter finds a slower collapse:  $R = A_c (t - t_c)^{5/9}$ . Ramsey's calculations reveal that the incompressible collapse can launch a shock wave under conditions where the Hunter collapse does not. A simple visualization of this issue is attempted in Figure 2 and Figure 3, which compares characteristics launched into the interior of a collapsing Argon bubble for the Rayleigh and Hunter cases. The characteristics here move at the velocity of sound in the gas plus the speed of the wall  $r(R,t) = R - \{t - t(R)\} \{c_g(R) + \dot{R}(R)\}$  where  $c_g(R) = dp_g / d\rho$  and to leading order we model the gas in which the characteristics move as being uniform and obeying the van der Waals adiabatic equation of state.  $p_g(R) = p_0 R_o^{3\gamma} [R^3 - a^3]^{\gamma}$ .



Figure 2: Characteristics launched by the wall of a collapsing argon bubble in an incompressible fluid cross consistent with the calculations in [21].



Figure 3: characteristics launched into a collapsing Argon bubble do not cross when the compressibility of the water is taken into account.

As an equation of motion for the bubble wall we have written down a generalization of the Rayleigh-Plesset equation [ $\beta=0$ ] that includes Hunter's compressibility effect [ $\beta=7/4$ ] when  $|\dot{R}| > c_0 / 4$ :  $(\frac{\dot{R}}{.25c_0})^{\beta} \{(2+\beta)R\ddot{R}+3\dot{R}^2\} = -2\frac{p_{\infty}}{\rho_0} + 2\frac{p_g(R)}{\rho_0} - \frac{2\dot{R}}{\rho_0c_0}\frac{dp_g}{dR}$ 

As can be seen in Figures 2,3 the characteristics cross for incompressible collapse whereas they do not cross for motion when the water is compressible. This suggests that an imploding shock wave is not generated by a collapsing gas bubble in water when  $R_m/R_0 = 10$  [1,2]. But the parameter space of SL is large and when water is cooled from room temperature to near 0C the light emission increase by over a factor of 10 [23]. These bubbles are characterized by  $R_m/R_0 = 15$ . Figure 4 shows the characteristics for this realization of bubble motion in compressible water. The characteristics barely cross in this case suggesting that a borderline situation for shock wave launching can be realized even in this compressible system.

## 4. ENHANCING SONOLUMINESCENCE WITH LESS COMPRESSIBLE FLUIDS

All of the fluids on which detailed experiments on sonoluminesce from single bubbles have been carried out are hydrogen bonded and so these fluids are only weakly incompressible. We raise here the possibility of carrying out sonoluminescence with fluids where the incompressibility would originate in the repulsion of nearest neighbor electron shells. This would be the case with small molecule molten salts such as molten LiF. We speculate that at high pressure these fluids will be more incompressible than water. But the data for the nonlinear acoustic properties of these systems is scant. Here we notice that Hunter's analysis suggests a 2 parameter characterization of fluids in terms of  $\beta$  and  $c_0$ . Figure 5 shows the collapse of a bubble with  $R_m/R_0 = 15$  and a postulated  $\beta=1$  and  $c=2c_0$ . The greater incompressibility of this putative fluid leads to a clear



Figure 4: Characteristics launched from the wall of an Argon bubble collapsing in water near 0C when the compressibility of the water is included. These bubble have a larger expansion ratio.



Figure 5: Characteristics launched from the wall of a collapsing Argon bubble with  $R_m/R_0 = 15$  for a fluid where the speed of sound is twice that of water and the collapse proceeds according to  $R \sim (t-t_c)^{1/2}$ .

tendency to shock formation inside the bubble. In his forefront experiment Ramsey interprets his data as indicating the formation of an imploding shock in a 1.8mm  $[=R_m]$  vapor bubble that is driven to collapse with  $p_{\infty} = 22atm$ .

In order to get to nuclear fusion will require an imploding shock in a hydrogenic gas but such gases have a high speed of sound and resist shock formation even in the incompressible limit. Here one will need to use fluids which are less compressible than water and also seek the extreme mass segregation that can be observed when hydrogen is mixed with xenon [24].

At the moment of collapse the bubble interior exhibits highly complex transport and plasma equations of state. It is amazing that controlled reproducible- especially synchronoussonoluminescence exists. If we wish to go to even higher energy densities via nonlinear acoustics a different class of less compressible fluids, and fluids with low vapor pressure should be employed.

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