Water temperature dependence of single bubble sonoluminescence

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(Phys. Rev. Lett., February 16, 1998)

The strong dependence of the intensity of single bubble sonoluminescence (SBSL) on water temperature observed in experiment can be accounted for by the temperature dependence of the material constants of water, most essentially of the viscosity, of the argon solubility in water, and of the vapor pressure. The strong increase of light emission at low water temperatures is due to the possibility of applying higher driving pressures, caused by increased bubble stability. The presented calculations combine the Rayleigh-Plesset equation based hydrodynamical/chemical approach to SBSL and full gas dynamical calculations of the bubble’s interior.

One of the remarkable features of single bubble sonoluminescence (SBSL) is the sensitivity of the light emission to the water temperature experimentally found by the UCLA group [2, 3], cf. figure 1. To obtain these results, Barber et al. proceeded as follows (refs. [2, 3] and B. Barber, private communication, August 1991): Water was cooled to a temperature of 2.5°C and completely degassed. An air pressure overhead of 150 Torr, corresponding to about 20% of gas saturation, was adjusted and SL experiments were performed, still at 2.5°C. Then the water was heated to 20°C without readjusting the gas concentration, and the SL experiment was repeated. Finally, the same measurement was performed after heating the water to 33°C. At all three temperatures, the forcing pressure amplitude $P_a$ of the driving sound field was adjusted in order to give maximum light intensity, while maintaining bubble stability against fragmentation (stable SL). According to the “waterfall plots” shown e.g. in ref. [2], the highest light intensity always corresponds to the largest achievable driving pressure ($P_a^{\text{max}}$). The experimental values for $P_a^{\text{max}}$ are shown in figure 2.

The bubble radius dynamics $R(t)$ was detected in these experiments (using Mie scattering techniques) and fitted by the Rayleigh-Plesset (RP) equation [2]. In this fit, several parameters were allowed to vary: the driving pressure amplitude $P_a$, the ambient radius $R_0$ (radius of the bubble under normal conditions) and also the surface tension $\sigma$ of the air/water interface and the water viscosity $\nu_l$. We consider the treatment of the latter two quantities as free parameters arbitrary, as $\sigma$ and $\nu_l$ are well defined material constants, but are varied in [2, 3] by as much as a factor of four and beyond. Fitting $\sigma$ and $\nu_l$ may be avoided if a more realistic model for the thermal behavior of the gas inside the bubble is applied: in the RP equation, the internal gas pressure is taken to vary polytropically with volume, with an effective exponent $\gamma$. In [2, 3] $\gamma$ changes abruptly from its isothermal to its adiabatic value at $R(t) = R_0$. Taking instead a more realistic model $\gamma = 1$ throughout the whole oscillation with the exception of the immediate vicinity of the strong bubble collapse results in a satisfactory fit with the physical values for $\sigma$ and $\nu_l$.

Because of the complications in the fits of refs. [2, 3], the resulting data should be read with some care. This is also reflected in figure 3, where we display the data from these two references for the expansion ratio (maximum radius $R_a^{\text{max}}$ divided by $R_0$): they show large deviations at otherwise unchanged parameters. The expansion ratio is a quantity closely related to the violence of collapse and therefore, presumably, to the intensity of energy concentration and light emission [4]. It is therefore puzzling that the same light intensity has been observed in refs. [2, 3] in spite of the different expansion ratios reported in figure 3.

The central claim of this paper is that the observed dependence on water temperature $T$ in figures 1 – 3 can be accounted for by the $T$ dependence of the material constants of water which are listed in table 1. In our analysis the most relevant effects are the temperature dependence of the gas solubilities (as already conjectured in ref. [3]) and of the viscosity; also, the temperature dependence of water vapor contributes. The variations of these material constants with temperature conspire to
allow for larger shape stable bubbles and larger forcing at lower temperatures, resulting in more light emission.

When dissolving air with its various constituents in water, only the argon concentration $c^a_A$ (far away from the bubble) is relevant in the SL regime, as SL air bubbles rectify argon $\epsilon$. The degree of saturation $c^s_A/c^a_A$ is one of the central parameters to determine the diffusive equilibrium (ambient) radius of SL bubbles. This equilibrium radius can be calculated within the hydrodynamical/chemical approach to SL $\epsilon$, which takes into account the mass exchange between the bubble interior and the liquid due to (i) diffusion and (ii) dissociation of molecular gases, which leads to argon rectification in the bubble. Here we follow that approach and calculate the phase diagrams in $R_0 - P_a$ space, resulting from the diffusional/chemical stability of the bubble for varying temperature, i.e., varying material constants and also varying $c^s_A/c^a_A(T)$. The calculations (based on Rayleigh-Plesset dynamics) are described in detail in refs.

### TABLE I

| water temperature | 25°C | 20°C | 33°C |
|-------------------|------|------|------|
| $\sigma$ [kg/s²]  | 0.075 | 0.0728 | 0.0707 |
| $\rho$ [g/cm³]    | 1.000 | 0.998 | 0.995 |
| $\nu_1$ [m²/s]    | 1.66  | 1.01  | 0.75  |
| $p_{vap}$ [kPa]   | 0.734 | 2.339 | 5.034 |
| $c_1$ [m/s]       | 1414  | 1483  | 1516  |
| $c_{0A}$ [kg/m³]  | 0.0892 | 0.0611 | 0.0495 |
| $c_{0N2}$ [kg/m³] | 0.0399 | 0.0283 | 0.0238 |

We choose the material constants given in table I, the frequency $26.5kHz$ as in experiment $\epsilon$, and an ambient pressure of 1 atm. We modeled air as a mixture of 99% nitrogen and 1% argon (in the gas phase above the liquid). At a given total gas concentration of $c_\infty/c_0 = 20\%$ for $T = 25°C$, the corresponding argon concentrations for the three temperatures are readily calculated from table I: $c^a_A/c^a_A(25°C) = 0.20\%$, $c^a_A/c^a_A(20°C) = 0.29\%$, and $c^a_A/c^a_A(33°C) = 0.36\%$, respectively.

The resulting equilibrium curves in phase space are shown in figure $\epsilon$. Diffusively stable bubbles are possible on the branches B and C $\epsilon$, however, according to the (qualitative) energy focusing condition $|\dot{R}|/c_a > 1$ $\epsilon$, (where $c_a$ is the speed of sound in the gas bubble) most bubbles on branch B will not be able to emit SL light, in agreement with recent experiments $\epsilon, \epsilon$.

A key issue is that the upper limit in both $P_a$ and $R_0$ on branch C is given by the parametric shape instability of the bubble wall $\epsilon, \epsilon, \epsilon, \epsilon$. The shape instability depends on the viscosity of the water $\epsilon$, which strongly increases with decreasing water temperature, thus stabilizing the bubble. For the values given in table I we find (within the approximations of $\epsilon$) that shape instability sets in at an ambient radius of roughly $R_0 = 3.5µm$, $4µm$, and $5µm$ for $33°C$, $20°C$, and $2.5°C$, respectively, see the dashed lines in figure $\epsilon$.

From figure $\epsilon$ one immediately realizes that the bubble can be driven harder (larger $P_a^{\text{max}}$) for lower temperatures. The values $P_a^{\text{max}}$ from figure $\epsilon$ are displayed in figure $\epsilon$ showing good agreement with experiment.

On decreasing the water temperature, the material constants of water are changed according to table I; this has...
the following effect on the curves in figure 4: (i) The water viscosity $\nu_l$ is increased; as stated above, $\nu_l$ determines the damping of bubble shape oscillations; therefore, bubbles are more stable; the stability line shifts towards larger $R_0$. (ii) The relative argon concentration $c_{Ar}^t/c_{Ar}^o(T)$ becomes smaller; in order to counteract dissolution, the bubbles must oscillate more violently to rectify gases. Thus, diffusive equilibria are reached only at higher $P_a$, and curves B and C are shifted to the right, allowing for larger $P_a^{max}$. (iii) The water vapor pressure $p_{vap}$ decreases; this, too, slightly shifts curves B and C to the right. The reason is that the reduced total pressure inside the bubble leads to smaller $P_a$. (iv) The small temperature dependences of $\sigma$, $p$, and $c_t$ have hardly any effect.

In figure 4 we include the theoretical expansion ratio $R_{max}/R_0$ calculated with the Rayleigh-Plesset equation for $P_{a}^{max}$ and $R_{a}^{max}$ determined from figure 3. Here, the agreement with the data from the fits of 2, 3 is not as good. We speculate that part of the discrepancy originates from the fit procedure (to obtain $R_0$) of refs. 2, 3 (see our criticism above) and suggest direct measurements of $R_{max}/R_0$, either by Mie scattering at two different frequencies or by employing the method developed by Holt and Gaitan [11].

One may wonder why a relatively modest change in $P_a$ and $R_{a}^{max}$ (cf. figs. 3 and 2) leads to considerably enhanced light emission, cf. fig. 1. The reason is twofold: (i) With increasing $P_a$ the bubble collapse becomes much more violent and therefore the gas inside the bubble undergoes stronger heating. The violence of the bubble collapse can be quantified by maximal pressure $p_{max}$ reached inside the bubble (within the hydrodynamical approach), cf. fig. 3, lower part. The observed increase with decreasing water temperature is a direct consequence of the observed increase in $R_{max}/R_0$ (upper part of fig. 3): For bubbles with a larger expansion ratio, more potential energy can be converted into kinetic energy during the collapse, compressing the bubble more strongly. (ii) Larger $R_{max}^{max}$ means that a larger number of gas molecules $\propto (R_{a}^{max})^3$ can be heated, which also leads to more light emission.

To complete our analysis, we would like to calculate the light intensity and compare it with the experimental data in figure 1. The light intensity can of course not be obtained from the RP approach to SBSL. It sensitively depends on the exact temperature achieved in the bubble at the collapse which can only be estimated within the Rayleigh-Plesset bubble approach, using one of a number of simple models, e.g., adiabatic heating at the collapse.

However, numerical codes that calculate the gas dynamics of the bubble’s interior and the resulting optical emission have been developed [4, 15]. We use the model recently proposed by Moss et al. [11] to calculate the gas dynamics inside the bubble at the collapse and the resulting optical emission. The model solves the gas dynamic equations for the conservation of mass, momentum, and energy in the gas bubble and the surrounding liquid, using a finite differencing scheme for the assumed spherical growth and collapse of the bubble. Accurate high pressure/temperature equations of state are used to describe the water and the argon. Energy loss by thermal conduction in the partially ionized gas created during the collapse is included in the model. Although light emission is the primary diagnostic of SBSL, the energy loss by the radiation of light is negligible compared to that by thermal conduction. This simplifies the calculation of the emitted optical power and spectra, which are calculated using the opacity and the calculated densities and electron temperatures of the partially ionized gas. Full details are given in [4]. The model was able to account for many experimentally found features of SBSL, including spectra and the sensitivity of the spectral intensity to the applied acoustic pressure (or, equivalently, $R_{max}$). This last result establishes the relevance of the model to the current analysis. We use the parameter pairs $(P_{a}^{max}/R_{a}^{max}$, $R_{a}^{max}$) or equivalently $(R_{max}/R_{a}^{max}$, $R_{a}^{max}$) resulting from above hydrodynamical/chemical approach for the three different analyzed water temperatures as input parameters for the gas dynamical code. The number of photons obtained in the wavelength window $[200nm, 750nm]$ is compared to the experimental data in figure 1, showing

![Figure 4](image-url)
reasonable agreement.

To summarize, the experimentally found water temperature dependence of SBSL can consistently be accounted for by the water temperature dependence of the material constants of water: At lower water temperature bubbles are more stable and can therefore be forced more strongly, resulting in more light. Combined with a gas dynamical simulation of the bubble’s interior, this approach can reproduce the three orders of magnitude increase in the observed photon number when changing the temperature from $\sim 30^\circ C$ to $\sim 0^\circ C$.

We suggest several control experiments to study the temperature dependence of SBSL. In all cases the experiments should be done with stable SBSL, whose regime in phase space can always (at least approximately) be calculated a priori, following the hydrodynamical/chemical approach.

(i) The bubble dynamics $R(t)$ should be measured as a function of the water temperature – this should reveal that the water temperature dependence of SBSL is mainly a bubble dynamical effect. In particular, as already mentioned above, the experimental data of figure 3 should be remeasured.

(ii) In addition, as suggested in a recent preprint by Vuong, Fyrillas, and Szeri, one should study how the bubble dynamics and the light intensity change (for fixed forcing pressure) with water temperature for xenon doped and helium doped nitrogen bubbles. The two noble gases should show different behavior, because $c^X_0$ strongly depends on the water temperature $T$, whereas $c^He_0$ only shows a weak dependence, see figure 3 of ref. [14]. This kind of experiment will be a useful diagnostic of the identity of the gas in the bubble. It will also help to distinguish between the gas solubility effects on the light intensity and other bubble dynamical effects, e.g., those caused by the temperature dependence of the water viscosity and of the vapor pressure.

(iii) Finally, instead of preparing the gas concentration at one temperature (resulting in a water temperature dependent relative concentration $c^Ar_0/c^Ar(T)$), one could keep $c^Ar_0/c^Ar_0$ constant at various temperatures, e.g. by using noble gas doped oxygen bubbles and controlling the actual gas content with oxyometry, cf. Gompf et al. [17]. If in addition the forcing pressure amplitude is kept constant, the location of the shape instability and its dependence on water viscosity will also become irrelevant. Altogether, we therefore predict that the water temperature dependence of SBSL light emission at otherwise fixed parameters $P_0$, $c^Ar_0/c^Ar_0$ will be only weak.

Acknowledgments: We thank all participants of the NATO-ASI workshop on Sonoluminescence and Sonoochemy for various discussions and in particular Larry Crum for the organization of that workshop. We are very grateful to M. Brenner and S. Grossmann for invaluable discussions and stimulating comments. Support for this work by the DFG under grant SBF185-D8 and by a joint NSF/DAAD grant is acknowledged. WCM’s work was performed under the auspices of the U.S. Department of Energy by LLNL under contract W-7405-Eng-48. S. H. and D. L. thank L. Kadanoff and the University of Chicago (where part of this work has been done) for hospitality and acknowledge partial support by the Office of Naval Research.

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[1] D. F. Gaitan, Ph.D. thesis, The University of Mississippi, 1990; D. F. Gaitan, L. A. Crum, R. A. Roy, and C. C. Church, J. Acoust. Soc. Am. 91, 3166 (1992); L. A. Crum, Physics Today 47, 22 (1994).
[2] B. P. Barber et al., Phys. Rep. 281, 65 (1997).
[3] B. P. Barber et al., Phys. Rev. Lett. 72, 1380 (1994).
[4] W. C. Moss, D. B. Clarke, and D. A. Young, Science 276, 1398 (1997).
[5] R. Hiller, S. J. Putterman, and B. P. Barber, Phys. Rev. Lett. 69, 1182 (1992).
[6] D. R. Lide, Handbook of Chemistry and Physics (CRC Press, Boca Raton, 1991).
[7] D. Lohse et al., Phys. Rev. Lett. 78, 1359 (1997); D. Lohse and S. Hilgenfeldt, J. Chem. Phys. 107, 6986 (1997). Further experimental hints for argon rectification in sonoluminescing air bubbles is given in recent work by T. Matula and L. Crum, preprint (1997); also the numerical simulation points at argon rectification.
[8] S. Hilgenfeldt, D. Lohse, and M. P. Brenner, Phys. Fluids 8, 2808 (1996).
[9] M. P. Brenner, S. Hilgenfeldt, and D. Lohse, in NATO-ASI on Sonoluminescence and Sonoochemistry, edited by L. Crum (Kluwer Academic Publishers, Dordrecht, 1997).
[10] M. Brenner, D. Lohse, and T. Dupont, Phys. Rev. Lett. 75, 954 (1995).
[11] G. Holt and F. Gaitan, Phys. Rev. Lett. 77, 3791 (1996).
[12] M. Plesset, J. Appl. Phys. 25, 96 (1954); A. Prosperetti, Quart. Appl. Math. 34, 339 (1977); Atti Accad. Naz. Lincei Rend. Cl. Sci. Fis. Mat. Nat. 62, 196 (1977).
[13] S. Hilgenfeldt, M. P. Brenner, S. Grossmann, and D. Lohse, J. Fluid Mech., in press (1998).
[14] The theoretical results for the onset of shape instabilities seem to (slightly) underestimate the experimental ones, possibly because heat conduction was neglected. For a detailed comparison between the theoretical and experimentally found onset of shape instabilities we refer the reader to reference [11].
[15] C. C. Wu and P. H. Roberts, Phys. Rev. Lett. 70, 3424 (1993); W. C. Moss, D. B. Clarke, J. W. White, and D. A. Young, Phys. Fluids 6, 2979 (1994); L. Kondic, J. I. Gersten, and C. Yuan, Phys. Rev. E 52, 4976 (1995); V. Q. Vuong and A. J. Szeri, Phys. Fluids 8, 2354 (1996).
[16] V. Q. Vuong, M. M. Fyrillas, and A. J. Szeri, preprint, submitted to J. Acoust. Soc. Am. (1997).
[17] B. Gompf et al., Phys. Rev. Lett. 79, 1405 (1997).