How hot do sonoluminescing bubbles become?

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Recent theoretical work in single bubble sonoluminescence has suggested that water vapor in the collapsing bubble leads to energy-consuming chemical reactions, restricting the peak temperatures to values for which hardly any light emission would occur. We demonstrate in an ab initio statistical approach that the excluded volume of the non-ideal gas results in a pronounced increase of the equilibrium constant and a respective suppresion of the (endothermic) particle producing reactions. Thus, sufficiently high temperatures for considerable bremsstrahlung emission can be achieved

INTRODUCTION

Stable clock-like light emission from an isolated gas bubble known as single bubble sonoluminescence (SBSL) was first observed in 1990 [1] and has been studied extensively since. The probable origin of the light has been identified as thermal bremsstrahlung and recombination radiation from the optically thin bubble heated to a few 10^4 K peak temperature [2–4].

Recently, it has been pointed out that water vapor may significantly reduce the heating at collapse of the bubble [5–8]. As the final phase of collapse is so fast that vapor cannot readily diffuse to the bubble wall to maintain the equilibrium composition, a considerable portion of vapor is trapped [5, 6, 8], which acts to reduce heating in two ways: (i) As the rest of the bubble consists largely of noble gas, the presence of water reduces the effective adiabatic exponent of the mixture, restricting the maximum temperature of the bubble to about 15000–20000K [2, 4, 5, 8]. These temperatures are still sufficiently high for thermal bremsstrahlung emission. (ii) However, taking chemical reactions of the water vapor into account [7, 9], drastically decreases the temperature since most of the reactions are endothermic and hence consume a major part of the thermal energy of the bubble. At the residual temperature of 6000–8000K [5] hardly any thermal bremsstrahlung or recombination radiation would occur.

The present work addresses the paradox in terms of an ab initio statistical approach [10] which provides evidence that *extremely* high densities as they occur in SBSL lead to a considerable suppression of the particle producing reactions in the bubble and hence to about 60% higher temperatures again. The mechanism at work qualitatively is as follows: Dissociated water molecules in the bubble take up more space than undissociated ones. When the bubble volume becomes comparable to the excluded volume of the gas molecules, little free space remains available, favoring the undissociated state.

MATHEMATICAL FORMULATION

The starting point is the partition function of a mixture of non-ideal gases from which the necessary thermodynamical quantities like energy, chemical potentials, and in particular the chemical equilibrium composition, i.e., the law of mass action, can be extracted. It is suitably expressed in terms of the number of particles of the various species N_X, the temperature T, and the volume V.

A comparison with the equilibrium constant for water dissociation obtained from the empirical forward and backward reaction rates shows good agreement for low and moderate particle densities, but significantly deviates for high density, see Figure 1. The effect is related to the exponential increase of this constant in a non-ideal gas as the excluded volume fraction $(\sum_X N_X)B/V$ approaches unity.

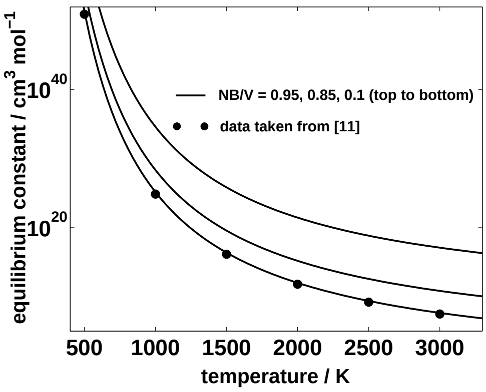

FIGURE 1. The equilibrium constant for water dissociation as a function of temperature for different excluded volume fractions. As the excluded volume fraction is raised, the equilibrium constant shifts to higher values equivalent to less dissociation. For comparison the circles mark the data from [11] for the low density limit.

In a second step this formalism for reaction thermodynamics is coupled to the heat and mass flux model of [8] to evaluate the effects of excluded volume on the temperature and the composition of a typical SBSL bubble.

Due to the simplifications in the model, the results should be understood as approximative, but they show a robust trend. Figure 2 depicts the dynamics of the number of OH and H_2O molecules and the temperature T for a time span of 6ns around collapse for typical SBSL conditions.

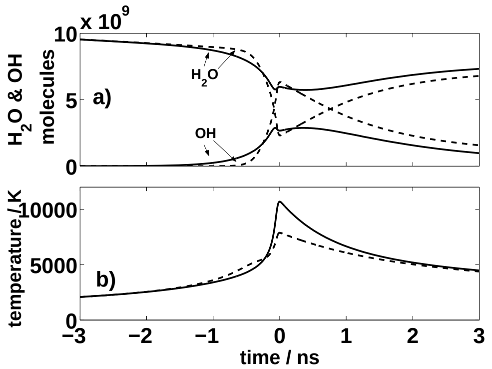

FIGURE 2. (a) Number of H_2O and OH molecules in an oscillating SBSL bubble as a function of time around collapse. (b) Bubble temperature for the same time interval. Dashed lines show calculations using semi-empirical reaction rates [7, 9], solid lines the present formalism. The excluded volume correction raises the maximum temperature to $\approx$ 11000K instead of $\approx$ 8000K. The equilibrium radius is $R_0^{\mathrm{Ar}} = 5\mu m$, the driving pressure $P_a = 1.4$ bar, the liquid temperature $T_0 = 293.15$K, and the driving frequency $f = 20$kHz.

Applying the semi-empirical reaction rates from [7, 9], more than 70% of the trapped water vapor is dissociated at collapse. However, the present excluded-volume calculation leads to only half as much dissociation (Fig. 2a). Accordingly, as the water dissociation is energy-consuming, the peak temperature rises from $\approx$ 8000K to $\approx$ 11000K (Fig. 2b).

CONCLUSION

We have offered a mechanism to resolve the paradox from recent SBSL models [5, 6] which predict temperatures too low for light emission when the endothermic reaction chemistry of water vapor is taken into account. Due to the vanishing free volume as the bubble approaches the excluded volume of the hard-sphere gas, the particle-producing dissociation reactions are found to be suppressed to a degree far beyond what could be expected in an ideal gas.

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