Density threshold for acoustic cavitation in water

F. Caupin, K. Davitt, A. Arvengas
Laboratoire de Physique Statistique, École Normale Supérieure,
UPMC Paris 06, Université Paris Diderot, CNRS, 24 rue Lhomond 75005 Paris, France

Introduction

When liquid water is subjected to mechanical tension, it is metastable with respect to the vapour phase. This metastable state exists only until thermal fluctuations trigger the nucleation of a vapour bubble, an event termed cavitation. Liquids can be subjected to tension in a variety of ways; see Ref. [1] for a recent review. However, many of these methods involve large volumes of liquid or are under tension for considerable lengths of time; conditions that favor heterogeneous nucleation of bubbles on impurities or on the vessel surface. In this report, we stretch liquid water with a focused acoustic wave produced by a hemispherical ultrasonic transducer. This method allows us to reach, with a very good reproducibility, among the largest tensions reported in water [2].

To characterize the cavitation limit, we measure the lowest density attained before nucleation of the vapour. The density of water during the acoustic burst is measured using a fiber optic probe hydrophone (FOPH) modeled on one first described by Staudenraus and Eisenmenger [3]. This type of hydrophone has been used in a report measuring negative pressures in water [4], but to our knowledge, has never been used in a systematic study of the cavitation threshold. The robustness and ease with which the fiber optic probe can be repaired makes this an extremely useful tool when studying water near the density threshold where energetic cavitation events can damage many other probes.

Apparatus

An acoustic wave in water alternatively submits the liquid to positive pressure and to mechanical tension, during the acoustic compression and depression phases respectively. To generate a focused acoustic wave, we use a hemispherical PZT transducer with inner and outer diameters of 16 and 20 mm (see Figure 1). The transducer is driven at resonance by a burst of 6 cycles at 1.03 MHz, where its impedance is real and equal to 23.5 Ω. Given the drive frequency, in water the ultrasonic wave is thus focused to a region of size a fraction of the sound wavelength $\lambda = 1.5\,\mathrm{mm}$ and during a fraction of the sound period $T = 1\,\mu\mathrm{s}$. These small values reduce the influence of nuclei for heterogeneous cavitation. In previous experiments with this same configuration we estimated that a tension of $-26\,\mathrm{MPa}$ was attained [2].

When cavitation occurs, bubbles of the vapour phase are nucleated. These bubbles can be detected by the echo produced by the reflection of the ultrasonic wave off of their surface. Counting the number of cavitation events for a given number of bursts applied to the transducer yields the cavitation probability, Σ. A plot of Σ as a function of the voltage, V, applied to the transducer produces a characteristic S-shaped curve, as shown in Figure 2. The cavitation statistics for this system have been previously studied in detail, see Ref. [2] for a full description. Their reproducibility is interpreted as an indication that the cavitation in these experiments is homogeneous. Owing to the shape of the rise of the Σ from 0 to 1 over a narrow range of applied voltage, a cavitation threshold can be easily defined. The latter is the voltage at which the probability of nucleating a bubble is 0.5, which we refer to as V_{cav} in the following. More specifically, to obtain V_{cav}, the probability measurements are fitted with the function expected for a thermally activated nucleation process [2]:

$$\Sigma(V) = 1 - \exp\left\{ -\ln 2 \exp\left[\xi \left(\frac{V}{V_{\mathrm{cav}}} - 1 \right) \right] \right\} \quad (1)$$

where ξ and V_{cav} are treated as adjustable parameters.

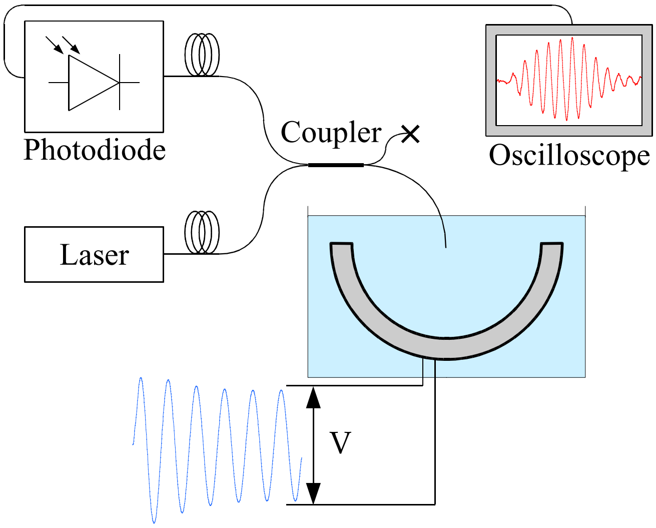

Figure 1: Experimental apparatus for measuring the density modulation of water subjected to an acoustic wave.

To measure the modulation of the density of the water at the acoustic focus, we built a fiber optic probe hydrophone (FOPH) modeled on that described in Ref. [3]. The basic principle of measurement is that from the modulation of the light intensity reflected from the endface of an optical fiber, the modulation in the refractive index of the liquid at the end of the fiber tip can be deduced. In Ref. [3], the refractive index is interpreted as a pressure via the Gladstone-Dale model

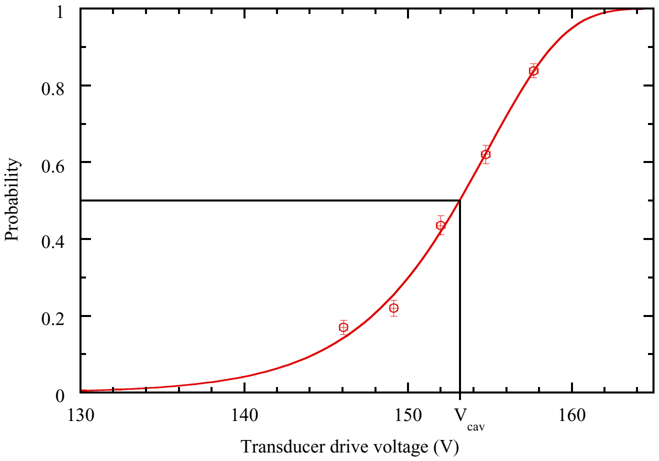

Figure 2: Cavitation probability versus transducer voltage for 6 cycle bursts at $T = 12°\mathrm{C}$. Each of the 5 data points was measured over 400 repeated bursts. V_{cav} is the cavitation threshold for which the probability of nucleation is 0.5.

(relating the index and density) and the Tait equation of state (relating the pressure and density). As our fundamental motivation is to investigate the equation of state (EOS) of water, we prefer to directly measure a thermodynamic variable. Thus, after careful calibration of the system, described below, we convert the reflected light intensity to an absolute density.

Our FOPH uses an 808 nm, 400 mW, fiber-pigtailed laser which is located at the input of a 3dB fiber-optic coupler, fabricated from a pure silica step index fiber with core and cladding diameters of 50 and 125 μm respectively. The laser light reflected from the interface between the fiber tip and the water is measured by a silicon photodiode located at another input of the coupler (see Figure 1). To ensure reproducibility of the reflection measurement, great care is taken to prepare and maintain the quality of the optical facet at the fiber extremity. Immediately after cleaving the fiber, and before each series of experiments we check the ratio

$$\beta = \frac{I_{\mathrm{H_2O}} - I_{\mathrm{d}}}{I_{\mathrm{air}} - I_{\mathrm{d}}} \tag{2}$$

where $I_{\mathrm{H_2O}}$, I_{air} and I_{d} are the reflected light intensities measured by the photodiode in absence of the acoustic wave, when the fiber is in water, air and when the laser is switched off, respectively. This provides an indication of the quality of the optical surface, specifically, if the reflectivity has changed due to cavitation events occurring at the endface of the fiber. If this is the case, the fiber is re-cleaved.

Measurements and Results

To verify operation of the FOPH, we acquired maps of the acoustic field along the axis of transducer symmetry and in the equatorial plane by scanning the clamped fiber using a 3D micrometer stage. As the fiber is relatively flexible, its length beyond the clamp is kept short (roughly 20 mm) to reduce the possibility of displacement caused by the ultrasonic wave or otherwise.

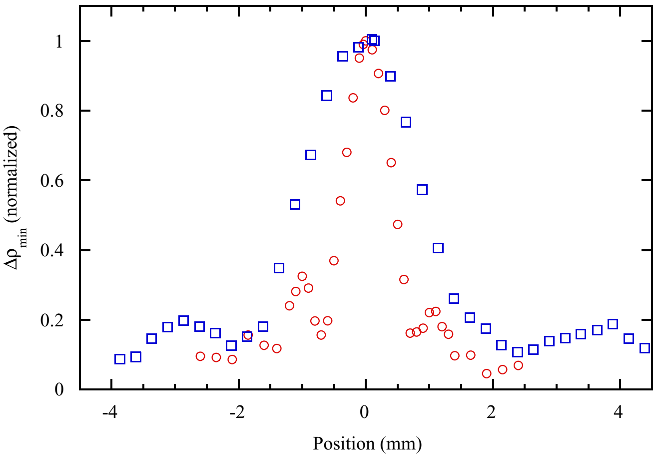

Figure 3: Maps of the acoustic field around the focus of the hemispherical transducer performed with the FOPH, at $T = 20°\mathrm{C}$ and $V = 0.5\,V_{\mathrm{cav}}$. Open circles/squares correspond to the field in the equatorial plane (x-y) and vertical (z) direction respectively.

As shown on Figure 3, the volume of the acoustic focus is found to be a prolate ellipsoid whose semi-major axes are 1.5 mm (x-y plane) and semi-minor axis is 3 mm (z direction). This corresponds to dimensions of $\lambda \times \lambda \times 2\lambda$, in the x, y and z directions. To ensure that the probe is placed at the acoustic focus, where the tension reaches its maximal value, a local acoustic map is performed before each experiment. The precision of this procedure is such that the fiber tip is placed within roughly 100 μm of the focus.

At the start of each measurement, the fiber is retracted from the water and a series of 400 bursts at 5 different transducer drive voltages in the vicinity of the cavitation threshold are used to determine V_{cav}, as in Figure 2. The fiber is removed in order to protect it from cavitation events, which can destroy the optical face, and it is repositioned using the micrometric translation stage. For the same reason, the density at the cavitation threshold cannot be measured directly. Instead, the transducer drive voltage is ramped up to $0.6\,V_{\mathrm{cav}}$ in roughly 15 steps. At each step, the optical intensity received by the photodiode, $I_{\mathrm{AC}} + I_{\mathrm{DC}}$, is averaged over 100 bursts in order to improve the signal-to-noise ratio. This is necessary as the modulation of the reflected light intensity is at most 20 μW. The data obtained are filtered with a cutoff at 10 MHz before being converted to a modulated reflected intensity, ΔR, via

$$\frac{I_{\mathrm{AC}}}{I_{\mathrm{DC}}} = \frac{\Delta R}{R_0 + S} \tag{3}$$

where R_0 is the reflection in static water and ΔR is the modulated reflection. S is a residual term included to account for stray reflection within the fiber coupler and by the unused output. Using a set of 20 calibrated refractive index liquids, it has been evaluated to be

$(3.1 \pm 0.8) \times 10^{-5}$ and the index of refraction of the fiber, n_f, is confirmed to be 1.453. A complete description of the calibration will be given elsewhere [5].

The modulated reflected intensity is converted to the modulation of the refractive index, n_{H_2O}, via the equation for reflection at normal incidence,

$$R_0 + \Delta R = \left[\frac{n_{H_2O} - n_f}{n_{H_2O} + n_f}\right]^2 \qquad (4)$$

Finally, the refractive index is converted to a density using a semi-empirical formula from the International Association for the Properties of Water and Steam [6]. We extrapolate this relation beyond the range in which it has been experimentally verified, which amounts to an assumption that the polarizability of water does not change character under tension. In this manner, we obtain $\Delta\rho(t)$ at each step of the ramp towards V_{cav} (see Figure 4). The minimum density, $\Delta\rho_{min}$, attained at each drive voltage is tabulated. To determine the change in density of liquid water at the cavitation threshold, $\Delta\rho_{cav}$, we extrapolate $\Delta\rho_{min}(V)$ to V_{cav} using a second order polynomial fit (see Figure 5). In order to ascertain the validity of this extrapolation, several test ramps were performed at higher fractions of V_{cav}, during which the fiber tip was ultimately destroyed by cavitation. We reached up to 0.9 V_{cav} and the fit was shown to hold.

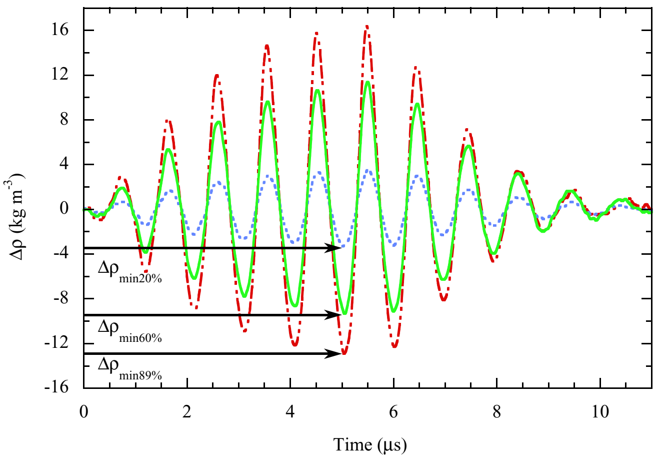

Figure 4: Density modulation of water subjected to an acoustic burst, shown for 3 different transducer drive voltages, 0.20 (dashed line), 0.60 (solid line) and 0.89V_{cav} (dotted-dashed line), all at $T = 12\,°C$. The largest decrease in density attained at each voltage is denoted as $\Delta\rho_{min}$.

Discussion

The hydrophone described above allows us to directly measure the density of water stretched by the application of an acoustic burst. The maximum tensions sustained by liquid water correspond to a reduction of the density of roughly 1.5% with respect to the saturation value. In repeated measurements, a statistical error bar of ±5% on $\Delta\rho_{cav}$ is found.

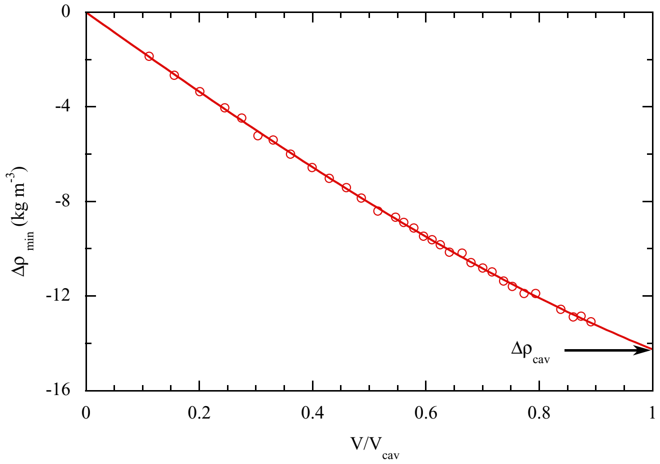

Figure 5: Largest decrease in density in the wave, $\Delta\rho_{min}$, as a function of the fraction of the cavitation voltage V/V_{cav} at $T = 12\,°C$. The measurements for V/V_{cav} below 0.6 are extrapolated to 1 using a second order polynomial fit going through zero.

On their own, these results can be interpreted as a measure of the limiting tensile strength of water. The tensile strength of water has been the subject of numerous investigations, and is usually given in terms of a maximum negative pressure sustained, or P_{cav}. The vast majority of experimental results (see Ref. [1] for a review) find $P_{cav} > -30\,\text{MPa}$. Only experiments performed on quartz inclusions have yielded $P_{cav} < -140\,\text{MPa}$ [7], which is close to theoretical predictions. Previously, it was alleged that all studies but the latter were plagued by heterogeneous cavitation. However, the direct comparison is complicated by the fact that different experiments measured different thermodynamic quantities that could only be compared with the use of an extrapolated equation of state. It happens that in [7] the measured variable is ρ_{cav}; we are therefore able to make a direct comparison with our results. We find that there is indeed a large discrepancy; Ref. [7] reaches a lower ρ_{cav} and hence a larger degree of metastability. Ref. [7] reports that in one quartz inclusion $\rho_{cav} = 903.6\,\text{kg}\,\text{m}^{-3}$ at 39.9°C, whereas we find $\rho_{cav} = 982.1\,\text{kg}\,\text{m}^{-3}$ at 37.8°C. Owing to the very reproducible nature of the cavitation statistics in our system, it is unlikely that this discrepancy can be ascribed to heterogeneous nucleation. A possible explanation for the difference relies on the fact that methods of subjecting water to tension are completely different in these two experiments. In [7], an isochoric path is followed at high temperatures, whereas we follow a quasi-isothermal path at considerably lower temperatures. We have proposed a scenario of complex nucleation in water that depends on the thermodynamic path followed [5], which could account for both experimental results. A detailed EOS of water under tension would allow us to check the plausibility of this scenario. Up to now, the EOS was only measured down to $-3.4\,\text{MPa}$ at room temperature in an early work [8]. Our method is able to reach a larger degree of metastability. The results for ρ_{cav} presented here constitute half of the EOS. In addition, we have assembled a Brillouin scattering experiment to measure the speed of sound in water during the acoustic burst. This will provide us with a second independent variable, measured under the same conditions (*ie* the same acoustic transducer at V_{cav} and T), and allow us to establish an EOS of water under mechanical tension.

References

[1] F. Caupin and E. Herbert, C. R. Phys. **7** (2006), 1000-1017

[2] E. Herbert, S. Balibar and F. Caupin, Phys. Rev. E **74** (2006), 041603 (1-22)

[3] J. Staudenraus and W. Eisenmenger, Ultrasonics **31** (1993), 267-273

[4] C. Wurster et al., Proceedings of the 1st World Congress on Ultrasonics, Berlin, 1995

[5] K. Davitt, A. Arvengas, and F. Caupin, in preparation.

[6] The International Association for the Properties of Water and Steam, *Release on the Refractive Index of Ordinary Water Substance as a Function of Wavelength, Temperature and Pressure* (1997)

[7] Q. Zheng, D.J. Durben, G.H. Wolf and C.A. Angell, Science **254** (1991), 829-832

[8] J. Meyer, Abhandl. d. Deutsch. Bunsen-Gessellschaft **6** (1911), 1-53