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Cavitation in Heavy Water and Other Liquids

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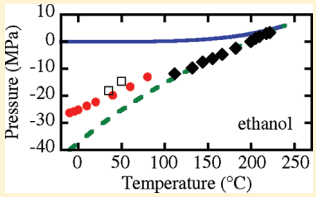
ABSTRACT: We report on measurements of the cavitation pressure in several liquids subjected to tension in an acoustic wave and compare the results to classical nucleation theory (CNT). This study is motivated by the sizable discrepancy between the acoustic cavitation threshold measured in water and the value predicted by CNT. We find that the same discrepancy is present for heavy water, whereas the agreement is better for ethanol and heptane and intermediate in the case of dimethyl sulfoxide. It is well-known that water is an anomalous liquid, a consequence of its hydrogen-bonded network. The other liquids studied represent very different molecular interactions. Our results indicate that the cavitation threshold approaches the prediction of CNT as the surface tension gets smaller. Conversely, this raises the question of the validity of a simple theory such as CNT to account for high surface tension liquids and suggests that an appropriate microscopic model of such liquids may be necessary to correctly predict the cavitation threshold.



INTRODUCTION

The attractive interactions between molecules in a liquid give rise to a cohesion that enables the liquid to sustain mechanical tension. A liquid that is stretched below the saturated vapor pressure is metastable with respect to the vapor phase; it exists only for a finite amount of time before "breaking" by nucleation of a vapor bubble in a process called cavitation. When nucleation is due to the thermal fluctuations in a pristine liquid, it is called homogeneous nucleation. However, the presence of impurities or nearby surfaces can lower the barrier to nucleation and cause heterogeneous nucleation.[1] Thus, in nucleation experiments where the objective is to study the fundamental properties of the liquid itself, one must take great care to reach the homogeneous limit.

Cavitation in water has been studied by numerous techniques.[2] We have used an acoustic method and have reached larger negative pressures than all but one other method.[3,4] The high reproducibility of the results[5] and their dependence on the sample size[6] are strong indications that it is the homogeneous nucleation limit that we observe. However, the cavitation pressure, around −30 MPa at room temperature,[7,8] is much less negative than expected from theoretical models.[9]

Water is an unusual liquid in many respects,[10,11] and explaining the origin of its anomalous behavior is an active topic of research.[12,13] Three main explanations have been proposed: the stability limit conjecture,[14] the metastable liquid−liquid critical point hypothesis,[15] and the singularity free scenario.[16] Knowledge of the cavitation limit of water may provide insight into this ongoing debate.[9] In particular, the form of the cavitation line in the $P-T$ plane (whether it is monotonic or whether it exhibits a minimum) may aid in discriminating between these proposed scenarios. Therefore, we decided to further investigate the discrepancy between acoustic cavitation experiments and theoretical predictions and in particular to determine to what extent it is unique to water.

First, we studied cavitation in heavy water (D_2O), which exhibits anomalous properties very similar to ordinary water (H_2O).[17] For instance at low temperature, both liquids exhibit a minimum in the static structure factor $S(q)$ at small q, the anomaly being even more pronounced in D_2O than in H_2O.[18] For cavitation, we find results in D_2O that are consistent with those in H_2O; again the magnitude is far from theoretical predictions. We then examined a series of other liquids. The cavitation pressure is expected to be strongly dependent on the surface tension; therefore, we studied heptane and ethanol, which have much lower surface tensions than water. As an intermediate case, we examined dimethyl sulfoxide (DMSO), which is interesting as it is a polar molecule, like water, and has a surface tension intermediate between typical solvents and water. For all liquids, we obtain very precise cavitation statistics. We argue that this is evidence that the acoustic experiment reaches the homogeneous cavitation limit in all of the liquids examined here. In addition, although these liquids have very different molecular interactions, we observe the general trend that the experimental cavitation threshold approaches predictions from classical nucleation theory (CNT) as the surface tension gets smaller.

In this paper, we begin by recalling the formalism of CNT. Then, we briefly describe the method used to produce cavitation by stretching a liquid in an acoustic wave and the procedure by which we determine the cavitation pressure. Finally, the results obtained for the four liquids investigated are compared to CNT and, when available, to other experimental data.

CLASSICAL NUCLEATION THEORY

CNT is the simplest way to address the nucleation of a more stable phase in a metastable one. It considers a spherical nucleus

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and assumes that its free energy can be split into two terms: a volume energy that accounts for the new phase being more stable and a surface energy arising from the bulk interfacial tension σ between the two phases. In the case of cavitation in a liquid stretched to pressure P below its saturated vapor pressure P_{sat}, the minimum work required to create a spherical bubble of radius R filled with vapor writes

$$\Delta W_{min} = \frac{4}{3}\pi R^3 (P - P_{sat})\delta + 4\pi R^2 \sigma \quad (1)$$

The factor $\delta = 1 - \rho_{vap}(P_{sat})/\rho_{liq}(P_{sat})$ appears because the vapor in the bubble is in chemical equilibrium with the metastable liquid;[1] this Poynting correction can be ignored far away from the critical point. ΔW_{min} reaches a maximum value E_b at a critical radius R_c, with

$$E_b = \frac{16\pi\sigma^3}{3(P_{sat} - P)^2\delta^2} \text{ and } R_c = \frac{2\sigma}{\delta(P_{sat} - P)} \quad (2)$$

A bubble whose radius is larger than R_c will grow spontaneously. Cavitation is a thermally activated stochastic process with a nucleation rate Γ per unit volume and time given by the Boltzmann factor

$$\Gamma = \Gamma_0 \exp\left(-\frac{E_b}{k_B T}\right) \quad (3)$$

When a volume V of liquid is stretched during a time τ, the probability Σ to observe at least one cavitation event is

$$\Sigma = 1 - \exp(-\Gamma V \tau) \quad (4)$$

We define the cavitation pressure P_{cav} as the pressure at which Σ reaches one-half. CNT thus predicts

$$P_{cav}^{CNT} = P_{sat} - \frac{1}{\delta}\left[\frac{3k_B T}{16\pi\sigma^3}\ln\left(\frac{\Gamma_0 V \tau}{\ln 2}\right)\right]^{-1/2} \quad (5)$$

The exact value of Γ_0 is debated, and V and τ vary between experiments. However, as they appear only through a logarithm, these parameters are not critical, and the value of P_{cav}^{CNT} is essentially given by the surface tension. Therefore, for simplicity, we have chosen a constant value $\Gamma_0 V\tau = 10^{19}$, typical for acoustic cavitation in water.[5] From eq 5, one can see that a change by 1 order of magnitude in $\Gamma_0 V\tau$ around this value results in only a 2.6% change in $P_{cav}^{CNT} - P_{sat}$.

MATERIAL AND METHODS

Acoustic Cavitation. There are many ways to generate tension in a liquid.[2] We use a focused ultrasonic burst and study the liquid during the period of rarefaction. Only a small volume of bulk liquid is stretched during a short period of time, thus minimizing any effect of heterogeneous cavitation, which takes place on surfaces or impurities. It is straightforward to repeat the acoustic bursts many times under the same conditions and thereby obtain cavitation statistics. In our work on water,[5] we have demonstrated that the cavitation probability can be precisely measured and that the results are extremely reproducible. These advantages are confirmed in the present study of other liquids. Below, we provide a brief summary of the experimental details, which have already been reported elsewhere.[5]

Acoustic waves are generated by a hemispherical piezoelectric transducer (16 and 20 mm inner and outer diameter) driven at its thickness resonance frequency of 1 MHz, by six-cycle bursts repeated at 1.75 Hz. We detect a cavitation bubble via the echo of the acoustic wave as it is reflected off the liquid−vapor interface and back to the transducer. In this manner, we can accurately count the number of bubbles in a series of acoustic bursts and thereby determine the cavitation probability Σ for any given drive amplitude U_{rms} of the transducer. The resulting curve is well described by a double exponential function

$$\Sigma = 1 - \exp\left\{-\ln 2 \exp\left[\xi\left(\frac{U_{rms}}{U_{cav}} - 1\right)\right]\right\} \quad (6)$$

which we call an S-curve. This expression is obtained by developing eq 4 around $\Sigma = 1/2$. It involves two fit parameters: the steepness ξ and the voltage U_{cav} at which P_{cav} is reached.

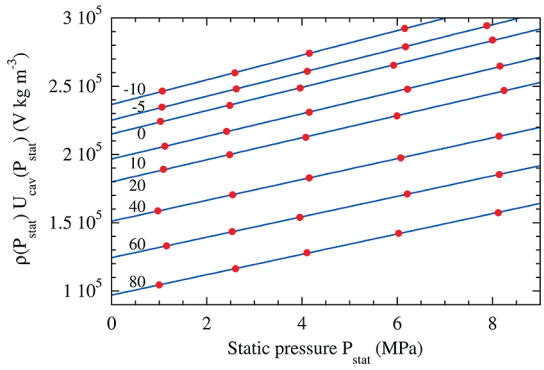

Figure 1. Static pressure method applied to ethanol. The red discs show the experimental data, with error bars smaller than the symbols. The solid blue lines are fits to eq 7 and are labeled with the temperature in °C.

Static Pressure Method. A calibration is needed to convert the experimental cavitation voltage U_{cav} into a pressure, to make a comparison with the value of P_{cav} predicted by CNT (eq 5). We use a static pressure method (SPM) where the transducer and liquid are placed in a closed cell in which the static pressure P_{stat} can be set between 0 and 10 MPa using a bellows system. P_{stat} is the pressure of the liquid without any sound wave. The higher P_{stat}, the larger the acoustic tension that must be applied to cavitate the liquid. Using a linear approximation for the sound wave focusing yields[19]

$$P_{stat} = P_{cav}(T) + K(T)\rho_{liq}(T, P_{stat})U_{cav} \quad (7)$$

at a temperature T. For all liquids studied, we found such a linear relation between P_{stat} and $\rho_{liq}(T,P_{stat})U_{cav}$. An example is given for ethanol in Figure 1. The factor $\rho_{liq}(T,P_{stat})$ exhibits only a small variation with pressure but was included in the analysis using the references listed in Table 1. In the case of DMSO, for which data are only available in the low-temperature range,[20] we have neglected this correction. In line with our previous work,[5] we usually fitted five points between 1 and 10 MPa at each temperature to estimate P_{cav} from eq 7. Within this linear approximation, the error bar on P_{cav} (which mainly arises from the small noise on the drive amplitude U_{rms}) never exceeds a few percent. Simulations of the focusing of a spherical wave[19] show that nonlinearities appear only at large amplitudes (when the minimum pressure gets closer to the liquid spinodal) and that their effect is to make P_{cav} more negative than the value determined by the SPM. We have confirmed this experimentally in water; we have performed direct measurements of the density[6] and of the sound velocity[8] $c = [(\partial P/\partial\rho)_S]^{1/2}$ of stretched water. Integrating

$c^2(\rho)$ gives the equation of state of the liquid in the usual $P-\rho$ plane, which yields a reliable value for the experimental P_{cav}. We did indeed find a more negative value than with the SPM and observed that the difference decreases with increasing temperature:[7] from 35% at 0 °C to 20% at 50 °C. The difference, although not insignificant, is much smaller than the discrepancy with CNT. We expect a similar behavior for heavy water. For the other liquids, we have not performed such an extensive study. For simplicity, we use the estimate of P_{cav} with the SPM. This is by far the largest source of uncertainty and outweighs any error due to the use of a constant $\Gamma_0 V\tau$ in comparison with CNT.

Liquids. Ethanol (>99.8% purity) was purchased from Fluka. Heptane (>99% purity) and DMSO (>99.5% purity) were purchased from Sigma-Aldrich. Heavy water (>99.7% purity) was obtained from the Commissariat à l'Energie Atomique, Saclay, France. All liquids were degassed with a combination of dry pumping and ultrasonic bath and transferred to the experimental cell under vacuum, following the procedure described in ref 5. The cell was immersed in a thermostatted bath operated between −10 and +80 °C, except for DMSO, which freezes around 19 °C. Although we simply report the bath temperature in the following, strictly speaking, an acoustic wave stretches a liquid adiabatically rather than isothermally. For H_2O, we have shown that the resulting temperature change increases with increasing temperature but is at most −2 °C at 80 °C.[6] This is because of the vicinity of the line of density maxima (4 °C at ambient pressure), on which the slope of an isentropic line in the $P-T$ plane becomes vertical. Thus the effect is also small in D_2O, which exhibits a density maximum at 11.2 °C at ambient pressure. The other liquids do not have a line of density maxima, and they might experience a larger adiabatic cooling. However, the effect remains limited. For instance, using the extrapolation at negative pressure of the equation of state of ethanol,[21] we calculate a maximum temperature decrease of less than 6 °C at −10 °C.

For CNT calculations with eq 5, values of P_{sat} and the bulk surface tension σ as a function of temperature were taken from the references listed in Table 1. For the surface tension of DMSO, we chose the only work[26] to our knowledge that spans the same temperature range as our experiment, between 20 and 80 °C (we note that other data, reported over a narrower range, are available and give slightly smaller values for the surface tension). For heavy water and DMSO in the temperature range studied, the Poynting correction δ (see eq 2) can be safely omitted (i.e., by putting $\delta = 1$). On the other hand, for ethanol and heptane, since we have included experimental data near the critical point in our discussion, we have taken δ into account using data for $\rho_{liq}(P_{sat})$ and $\rho_{vap}(P_{sat})$ from ref 21 for ethanol and from ref 22 for heptane.

Table 1. References of Data Used in CNT Calculations[a]

liquid	D_2O	ethanol	heptane	DMSO
reference for P_{sat}	22	21	22	23
P_{sat} at 20 °C (kPa)	2.0	5.9	4.7	0.06
reference for σ	24	25	22	26
σ at 20 °C (mN m^{-1})	72.6	22.2	20.6	43.7

[a] The saturated vapor pressure and the surface tension at 20°C are given for illustrative purposes.

RESULTS AND DISCUSSION

The features of the acoustic experiment with ordinary water are confirmed for all the liquids studied: high reproducibility, high quality fits of the S-curves, and linear dependence between P_{stat} and $\rho_{liq}(P_{stat})U_{cav}$. Therefore, for each liquid, we display only two plots: a typical S-curve and P_{cav} as a function of temperature. When available, we include cavitation data obtained

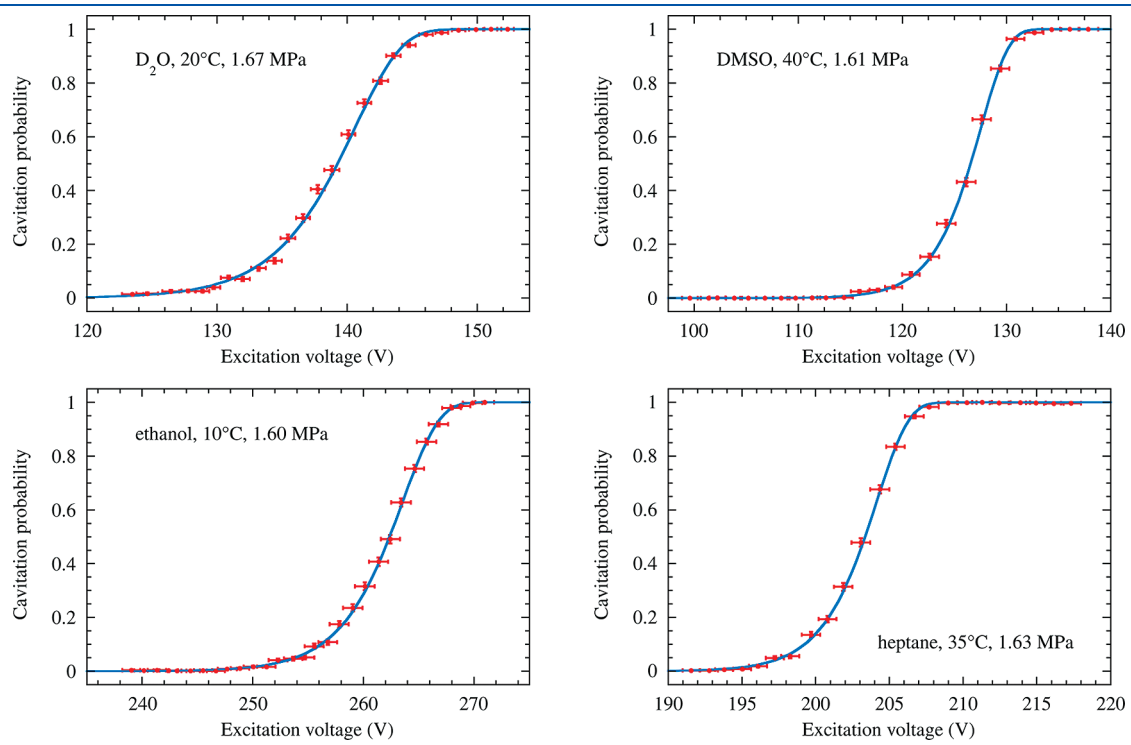

Figure 2. Cavitation probability as a function of excitation voltage. A typical curve is shown for each of the four liquids studied. The filled red circles (with error bars) are the experimental data, with each probability measured over 1000 repeated bursts. The solid blue lines are fits to eq 6.

in other experiments. For each liquid, we compare the experimental P_{cav} to the prediction of CNT.

Let us first discuss the cavitation statistics. A typical S-curve for each liquid is shown in Figure 2. Each cavitation probability is measured by counting the number of cavitation events detected over 1000 bursts repeated under the same experimental conditions. It can be seen that the data are perfectly fitted by eq 6. The corresponding double exponential function has a characteristic shape typical of a thermally activated process. As we discussed in the case of water,[5] this excludes the possibility that cavitation takes place by a mechanical instability occurring at a fixed threshold on impurities, such as pre-existing bubbles trapped at the surface of floating objects. Thermally activated heterogeneous nucleation remains a possibility, but it requires that a specific impurity be present with a sufficient concentration to trigger nucleation around the observed threshold. In our opinion, this is an unlikely scenario, and the precision of the cavitation statistics is indicative of homogeneous cavitation.

We now turn to the results for the shape of the cavitation line $P_{cav}(T)$. The results for D_2O are shown in Figure 3. Two independent runs were performed with two different D_2O samples and show good reproducibility. We are unaware of any existing data for cavitation in D_2O. However, for comparison, we mention a related experiment by Henderson and Speedy.[27] When liquid water is cooled at nearly constant volume in a so-called Berthelot tube, a negative pressure develops until the line of density maxima (LDM) is reached; further cooling then releases the tension in the liquid. Henderson and Speedy built a Berthelot−Bourdon tube that allowed them to measure the negative pressure in the liquid during cooling. They studied H_2O, D_2O, and a mixture and located the LDM for each case. In Figure 3 we include the largest tension that they were able to reach in D_2O; our data exceed this value. Also shown in Figure 3 are the data collected for 9 runs in H_2O using the same acoustic technique.[5] Interestingly, we see that the results for both water isotopes are identical within the error bars. This is expected from the very similar values of the surface tension: in the temperature range studied, the surface tensions taken at the same temperature differ by less than 0.25%. According to CNT, this would lead to a change in P_{cav} of less than 0.37%. This predicted proximity of P_{cav} in H_2O and D_2O is thus borne out by the acoustic experiment. However, the experimental and CNT values for P_{cav} in D_2O are in strong disagreement, by a factor of approximately 8. The discrepancy between the acoustic experiment and CNT is thus as large in D_2O as it is in H_2O.

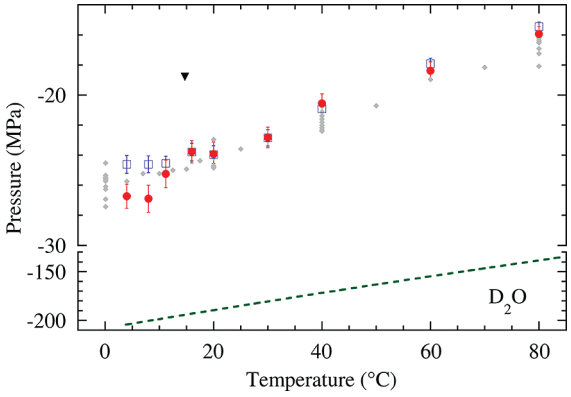

Figure 3. Cavitation pressure as a function of temperature for heavy water. The filled red circles and empty blue squares show the data from two independent runs of the present acoustic experiment. The black triangle is the largest negative pressure reached in a Berthelot tube experiment with D_2O.[27] For comparison, the combination of nine H_2O runs from our previous work[5] is displayed (gray diamonds). The error bars of these data are omitted for clarity; they are consistent with the data scatter. The green dashed curve shows the prediction of CNT, eq 5, with $\Gamma_0 V \tau = 10^{19}$; note the break in the vertical axis.

We now turn to ethanol and heptane (Figure 4). Our results can be compared to previous work. For ethanol, we display the largest negative P_{cav} reached at two temperatures in repeated runs in a metallic Berthelot tube:[28] they are less negative than the one obtained with the SPM in the acoustic experiment. Vinogradov and Pavlov measured P_{cav} in ethanol and heptane using a heat pulse on a thin platinum wire to superheat the liquid and simultaneously stretched it using a plane acoustic wave;[29] their study was limited to high temperature. The comparison of our data with Vinogradov and Pavlov is indirect because the temperature ranges do not overlap. However, if one compares the data of one group with the extrapolation of the other, our data seem slightly less negative for ethanol and slightly more negative for heptane but qualitatively fall on the same line. As for the prediction of CNT, it is in very good agreement with the data of Vinogradov and Pavlov at high temperature, near the critical point, whereas it starts to deviate at lower temperature. This behavior persists at lower temperature, where our data fall between 30 and 36% above the prediction of CNT for ethanol

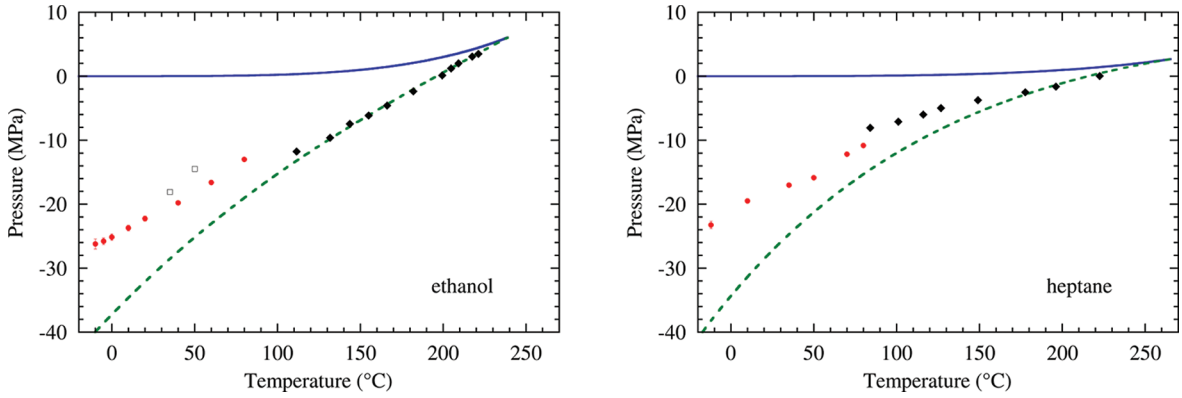

Figure 4. Cavitation pressure as a function of temperature for ethanol (left) and heptane (right). Each panel displays the saturated vapor pressure (solid blue curve), P_{cav} deduced from the SPM (filled red circles), and the prediction of CNT, eq 5, with $\Gamma_0 V \tau = 10^{19}$ (dashed green curve). Experimental data from previous work are included: ref 28 (empty black squares) and ref 29 (filled black diamonds).

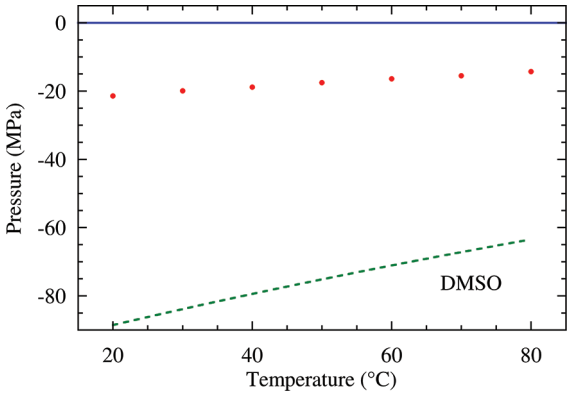

Figure 5. Cavitation pressure as a function of temperature for DMSO. The SPM gives values of P_{cav} shown by the filled red circles; the error bars are the size of the symbols. The solid blue line is the saturated vapor pressure, and the dashed green curve is the prediction of CNT, eq 5, with $\Gamma_0 V \tau = 10^{19}$.

and between 25 and 39% above CNT for heptane. Remembering that the SPM gives an upper bound, the actual P_{cav} will be closer to CNT; this calls for further studies. In any case, the agreement is significantly better than for water isotopes.

Figure 5 displays our results for DMSO. We are not aware of any cavitation experiments reported in this liquid. We thus compare only to CNT, which predicts values of P_{cav} more than four times more negative than the experimental ones. DMSO thus appears as an intermediate case, the disagreement with theory being less pronounced than for water. It would be interesting to go beyond CNT to understand the discrepancy. For instance, one could use modern simulation techniques to determine the energy barrier, as has been done in the case of the Lennard-Jones liquid,[30,31] but including the more complex (e.g., dipole−dipole) interactions present in DMSO.

CONCLUSION

For all liquids investigated here, the acoustic experiment yields very reproducible cavitation statistics and pressures. In addition, when other experiments are available for comparison, the present values of P_{cav} are in good agreement or more negative. These findings suggest that we are observing homogeneous nucleation. This conclusion is supported in part by the comparison to CNT. For ethanol and heptane, our experimental P_{cav} values are around 30% less negative than CNT predictions, which can be explained by remembering that the SPM gives an upper bound on P_{cav}. Moreover, CNT makes some crude assumptions. It considers a spherical cavitation nucleus filled with vapor whose boundary with the liquid is infinitely sharp. Accounting for smoother density profiles, e.g., by density functional theory, yields P_{cav} less negative than within CNT.[1,9,32] Also, the cavitation nucleus has a microscopic size, and it is not clear that one can use the bulk, macroscopic surface tension, as is done in CNT. Furthermore, the cavitation nucleus gets smaller with decreasing temperature, exacerbating this assumption. This might explain why CNT appears to be in better agreement with experiments at high temperature in ethanol and heptane.

On the other hand, the values of P_{cav} in the other liquids are much less negative than the predictions of CNT, by a factor of around 8 in heavy water and around 4 in DMSO. Such a large discrepancy cannot be ascribed to the error expected in the pressure calibration. Interestingly, the experiments in heavy and ordinary water yield very similar values for P_{cav}, with the same ratio between experiment and theory.

Overall, these observations suggest that acoustic cavitation is well suited for the study of homogeneous nucleation. The results for simple liquids can, to a large extent, be understood with a theory as simple as CNT. On the other hand, the study of DMSO and of water isotopes reveals a surprising behavior that is likely to be related to the more complex interactions in these liquids. This also adds to the puzzle raised by the experiments on water inclusions in quartz,[3,4] which find P_{cav} values close to CNT predictions; see the discussion in ref 7.

Further studies are needed. In particular, as we have done for ordinary water,[7] we plan to use a fiber optic hydrophone to measure the density of other liquids at the cavitation threshold. This will provide an accurate calibration of the cavitation pressure and allow a better quantitative comparison to theory.

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