

Supplemental material: Brillouin spectroscopy of metastable superfluid helium-4

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1 Averaging issues on the spatial density profile measurements of metastable superfluid helium-4.

The spatial resolution of 20 μm of the density measurement stated in ref.[1] should not be directly compared to the spatial resolution of the Brillouin frequency measurement presented in the current article. In ref.[1], the spatial resolution is defined as the spatial period for which the contrast of a transmission sinusoidal pattern observed through the imaging system is half its maximum. In the present article, it is defined as the full-width at half maximum of the gaussian distribution by which physical properties are convoluted during the measurement due to the gaussian beam profile of the lasers. The spatial resolution mentioned in ref.[1] corresponds to an effective convolution by a gaussian function whose standard deviation is $\sigma = 3.75 \mu\text{m}$, which is more than 5 times lower than the spatial resolution on the measurement of the Brillouin frequency f_B . Thus, there is a very low effect of spatial averaging on the measurement of the density ($\sim 1\%$) and the corresponding shift on the value of ρ_{cav} is within the error bar given for this quantity in ref.[1].

2 Estimation of the destabilization pressure of metastable superfluid helium-4

Here we provide a detailed calculation for the estimation of destabilization pressure P_c of metastable superfluid helium-4. As stated in the main text of the article, we assume a linear dependence of the Brillouin frequency f_B of the (metastable) liquid on its density ρ :

$$f_B = a\rho + b \quad (1)$$

with a and b two constant parameters. In order to compute these parameters, we use two known values of the couple (f_B, ρ) .

The first one corresponds to the thermal equilibrium state ($T = 0.96 \text{ K}$, saturated vapor pressure, no acoustic wave $V = 0$). We have measured $f_B^0 = 317.8 \text{ MHz}$ and the corresponding density is well tabulated $\rho_0 = 145.1 \text{ kg/m}^3$ [2].

The second one corresponds to the space time minimum of the density and of the Brillouin frequency at the destabilization threshold where $\Sigma = 1/2$, Σ being the cavitation bubble creation probability. In the present work, the space time minimum Brillouin frequency (swing (C) of Fig.3) at the destabilization threshold is measured to be $f_B^*(\Sigma = 1/2) = 283 \text{ MHz}$. In ref.[1], the space time minimum density (also in swing (C)) has been measured to be $\rho^*(\Sigma = 1/2) = 133.8 \text{ kg/m}^3$.

We thus compute the a and b parameters to find:

$$a = 3.1 \text{ MHz m}^3/\text{kg} \quad (2)$$

$$b = -129 \text{ MHz} \quad (3)$$

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Note that, because of averaging issues on the Brillouin frequency explained in the main text, 283 MHz is an upper bound for $f_B^*(\Sigma = 1/2)$. A lower bound for $f_B^*(\Sigma = 1/2)$ (prior to averaging) corresponds to a 3 % reduction of the quantity leading to 274.5 MHz.

This will change the (a, b) parameters to:

$$a' = 3.8 \text{ MHz m}^3/\text{kg} \quad (4)$$

$$b' = -238 \text{ MHz} \quad (5)$$

Those changes have a modest impact on the determination of the destabilization pressure as it will be shown hereafter.

To estimate P_c , we square and re-write Eq.(1) of the main article and insert Eq.(1) of the supplemental material to get:

$$f_B^2 = \frac{1}{K} n^2 v^2 \quad (6)$$

$$\left. \frac{\partial P}{\partial \rho} \right|_S = K \frac{(a\rho + b)^2}{n^2} \quad (7)$$

where $K = \frac{\lambda_0^2}{4 \sin^2(\theta/2)}$ is a constant. Its value is determined by measuring the Brillouin frequency $f_B^0 = 317.8(3)$ MHz at thermal equilibrium. Knowing the values of the refractive index $n_0 = 1.028$ and of the first sound velocity $v_0 = 237.9$ m/s[2] in the thermal equilibrium state, one finds $K = 0.592(1) \mu\text{m}^2$. Note that the determination of K gives in fact the scattering angle as $\lambda_0 = 1064, 46$ nm[3] leading to $\theta = 87.5(1)^\circ$.

The refractive index slightly depends on the density via the Lorentz-Lorenz relation which is well approximated for liquid helium by:

$$n^2(\rho) = 1 + \beta\rho \quad (8)$$

with $\beta = 3.87 \cdot 10^{-4} \text{ m}^3/\text{kg}$ [3]. Eq(7) can be analytically integrated at constant entropy from an initial state (P_i, ρ_i) to a final state (P_f, ρ_f) :

$$\int_{P_i}^{P_f} dP = \int_{\rho_i}^{\rho_f} K \frac{(a\rho + b)^2}{1 + \beta\rho} d\rho \quad (9)$$

$$P_f - P_i = \left[\frac{1}{2\beta^3} \{ a\beta\rho(4b\beta + a(\beta\rho - 2)) + 2(a - b\beta)^2 \ln(1 + \beta\rho) \} \right]_{\rho_i}^{\rho_f} \quad (10)$$

We define the destabilization pressure to be the space time minimum pressure P^* at the destabilization threshold $P_c = P^*(\Sigma = 1/2)$. This corresponds to the minimum pressure in swing (C) of the acoustic wave. To evaluate it, we take the initial state to be the 0.96 K vapor pressure thermal equilibrium state ($P_i = P_0 = 10$ Pa, $\rho_i = \rho_0 = 145.1 \text{ kg/m}^3$) and the final state to be ($P_f = P_c$, $\rho_f = \rho^*(\Sigma = 1/2) = 133.8 \text{ kg/m}^3$).

Doing so, using the values given by Eq.(2) and Eq.(3) for the a and b parameters, we find :

$$P_c = -5.7 \text{ bar} \quad (11)$$

Using the values a' and b' (Eq.(4) and Eq.(5)) corresponding to a lower value of the minimum Brillouin frequency, we find :

$$P'_c = -5.6 \text{ bar} \quad (12)$$

a modest change which has been included in the error bar of the result given for P_c in the main text of the article.

Finally we shall stress that the fact that $P_c < P'_c$ is not surprising. The Brillouin frequency is a measure of the compressibility of the liquid. If at fixed density density variations $(\rho^*(\Sigma = 1/2) - \rho_0)$ is a constant parameter in Eq.(10) f_B^* decreases, the liquid becomes more compressible and it is harder to build up a pressure difference for a given density variation.

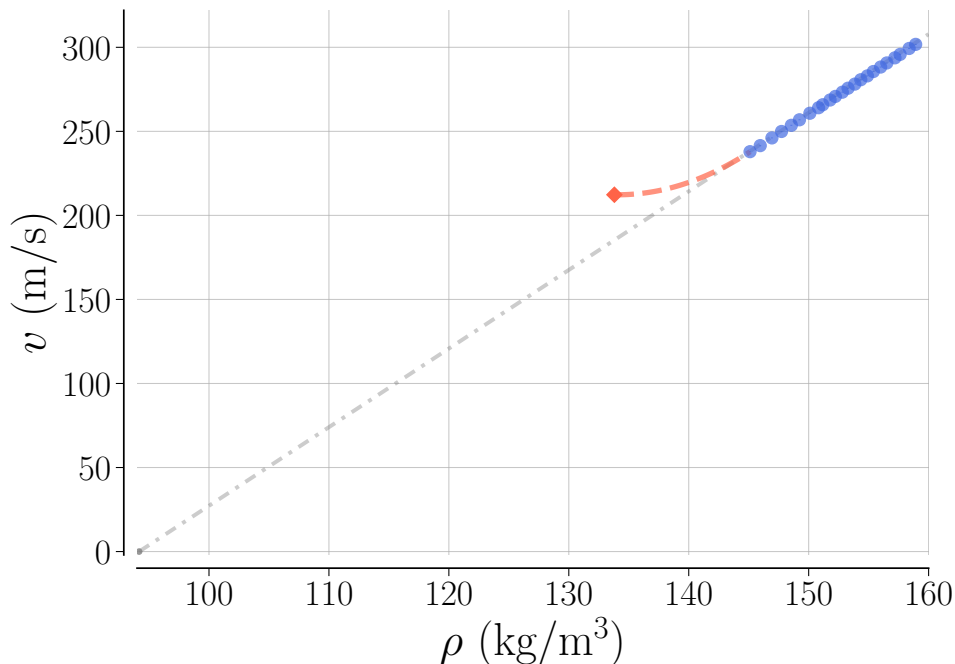


Figure 1: Speed of first sound in liquid helium versus density at $T \sim 1$ K. Blue points : stable states ($P > 0$, ref.[3]) ; red diamond : this work ; dotted line : quadratic fit function for $v(\rho)$ at $P < 0$ (see text) ; dash-dotted line : theory (ref.[4]).

3 Link to positive pressure measurements

It is interesting to analyze those results in terms of the values of the sound velocity instead of the Brillouin frequency ones because sound velocity values are independent of the scattering angle θ . Hence, one can plot on a same graph the sound velocity density dependence ($T \sim 1$ K) obtained with our setup at positive pressure[3] and the value obtained in present work at negative pressure.

The corresponding $v(\rho)$ curve is shown on Fig.1 and calls for several comments. On the positive pressure side ($\rho \geq \rho_0 = 145.1$ kg/m³), the sound velocity is a linear function of the the density of slope $\frac{\partial v}{\partial \rho}|_{P>0} \simeq 4.6$ m⁴/s/kg. If one considers a linear relation between the data points $(\rho_0, v(\rho_0))$ and $(\rho^*(\Sigma = 1/2), v(\rho^*(\Sigma = 1/2)))$, one finds that the slope must change to $\frac{\partial v}{\partial \rho}|_{P<0} \simeq 2.8$ m⁴/s/kg in the negative pressure domain. The destabilization pressure can also be computed from the above curve as $P_c = P_0 + \int_{\rho_0}^{\rho^*(\Sigma=1/2)} v^2(\rho') d\rho'$. The linear dependence of v on ρ in the metastable region leads to the value $P_c = -5.7$ bar, equivalent to the one obtained by fitting the Brillouin frequencies with a linear relationship. One may object that the derivative $\frac{\partial v}{\partial \rho}$ must be continuous at ρ_0 . To avoid this problem, one can estimate the destabilization pressure using an hypothetical quadratic $v(\rho)$ function in the metastable domain passing through the points $(\rho_0, v(\rho_0))$ and $(\rho^*(\Sigma = 1/2), v(\rho^*(\Sigma = 1/2)))$ and with the slope $\frac{\partial v}{\partial \rho}|_{P>0}$ at point $(\rho_0, v(\rho_0))$, see the red dashed curve in Fig.1. Doing so, one finds $P_c^{quad} = -5.5$ bar, not far from P_c and still in clear contradiction with previous experimental reported values.

Finally, we want to stress that the disagreement between our measurement and available EoS of metastable superfluid helium-4 already mentioned in the main text of the article can be clearly seen on Fig.1. The gray dotted curve corresponds to the EoS at 0 K from ref.[4]. Within the pressure range explored in the present experiment, the theoretical EoS at 1 K is essentially the same than the one at 0 K (see article). One sees that the sound velocity value we obtain in the negative pressure domain deviates from the expectation of the theory. It should be noted that our measurement suggests that the actual $v(\rho)$ curve in the metastable domain must show an inflection point to bend back towards zero at the spinodal limit where the sound velocity vanishes. This anomalous sound velocity density dependence and its possible link to dispersion effects in superfluid helium at negative pressure is certainly of interest to be further investigated.

References

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