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Brillouin spectroscopy of metastable superfluid helium-4

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Abstract

We report measurements of the Brillouin frequencies of acoustically driven metastable states of superfluid ⁴He at $T \sim 1$ K. This is done using a stimulated Brillouin gain spectrometer operating on a confined spatio-temporal domain ($\sim 40 \mu\text{m}$, 200 ns). Our work questions previous experimental results regarding the metastable states of liquid ⁴He and suggests that thermal effects or vortices can play a significant role in the cavitation process. Thanks to our time-resolved measurements, we also find that it is possible to probe metastable superfluid ⁴He beyond the commonly defined cavitation threshold.

1 Introduction

Because liquid helium-4 at low temperature is the purest possible liquid, it is a model system to probe deep condensed matter metastable states and homogeneous nucleation phenomenons. Especially the stability limits of superfluid ⁴He with respect to its gaseous phase at negative pressure have been studied experimentally and theoretically, see for instance the review articles[1, 2, 3] and references therein. The absence of impurities and the little thermal energy available in the fluid make superfluid ⁴He an appealing candidate to reach experimentally the close vicinity of the spinodal limit where the compressibility of the liquid diverges making it mechanically totally unstable even in the limit $T \rightarrow 0$. For instance, Caupin *et al.* have developed an indirect method to measure the cavitation pressure of acoustically driven metastable liquid helium-4 and found it to be between -10 bar and -8 bar at 1 K[4], which is compatible with theoretical estimates of the spinodal limit $P_{\text{spin}}(1 \text{ K}) \sim -9$ bar[5].

In this Letter, we report local and time-resolved measurements of the Brillouin frequencies in acoustically driven metastable states of superfluid ⁴He at $T \sim 1$ K. Associated with previous density measurements[6], these Brillouin frequency measurements enable us to estimate a cavitation pressure of superfluid ⁴He at $T \sim 1$ K in disagreement with previous results. Our time-resolved technique also allowed us to show that, at the destabilization threshold, cavitation does not occur at the minimum amplitude of the sound wave creating the metastable states. This suggests that either thermal effect or vortices can play a significant role in the cavitation process. Our estimation of the cavitation pressure at the destabilization threshold is compatible with a vortex assisted cavitation model.

Experiment: Deep metastable states of superfluid ⁴He are experimentally produced using a high amplitude sound wave focused in the bulk liquid[7, 8, 9, 4, 6, 10], see Fig.1. At acoustic focus and during the negative swing of the wave, the liquid is locally stretched by the acoustic wave and one can thus explore its metastable (negative pressure) states. The experimental cell containing liquid helium is cooled in a cryostat with four optical ports. In the experiment described in this paper, the temperature is fixed at $T = 0.96$ K. Liquid helium is at saturated vapor pressure. A hemispherical piezoelectric transducer excites and focuses ultrasound waves in helium at the resonant frequency $f_{ac} = 1.14$ MHz of its first thickness vibration mode. The transducer inner diameter is 12 mm and the thickness is 2 mm. The radius of the acoustic focus is $\sim 100 \mu\text{m}$ [6]. Metastable states of superfluid ⁴He are produced and must be probed on a time scale of less than ~ 400 ns (half period of the sound wave) and on a spatial scale of less than $\sim 100 \mu\text{m}$.

For that purpose, we have developed a Stimulated Brillouin (SB) gain spectrometer capable of measuring the Brillouin frequencies of superfluid ⁴He on such a narrow spatio-temporal domain[11, 12, 13]. Brillouin scattering refers to the scattering of light by a transparent medium due to the coupling of incoming photons with phonons of the material[14]. The energy-momentum conservation in the photon/phonon collision imposes that the Brillouin scattered light is frequency shifted by the amount:

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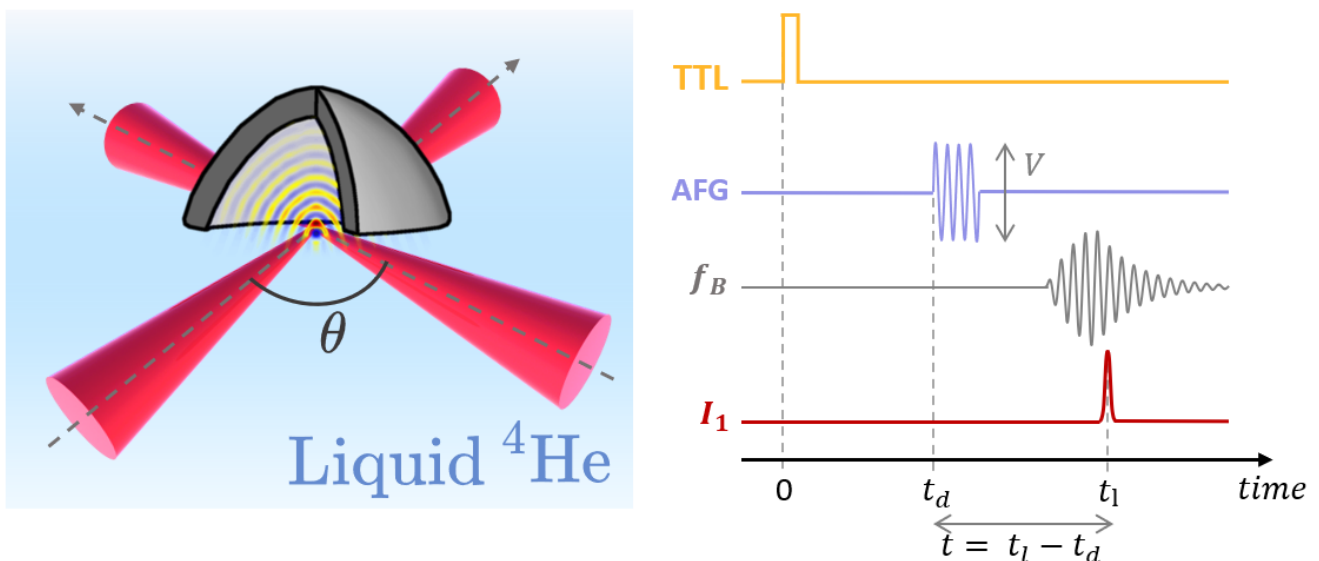


Figure 1: Left: scheme of the experiment. A piezoelectric transducer focuses 1.14 MHz acoustic waves into bulk liquid helium ($T \sim 1$ K) producing metastable states during the negative swings of the wave. A SB gain spectrometer consisting of two crossed laser beams, a pulsed pump and a frequency tuneable CW probe, is used to measure time-resolved Brillouin frequencies of the liquid at acoustic focus. θ is the crossing angle between the lasers. Right: timeline of the experiment. Orange: 10 Hz TTL clock signal delivered by the SB pulsed pump laser, Blue: driving piezo transducer voltage (amplitude V , frequency f_{ac}), Gray: Brillouin frequency at acoustic focus, Red: Pump SB laser intensity I_1 .

$$f_B = 2n \frac{v}{\lambda_0} \sin(\theta/2) \quad (1)$$

where n is the refractive index, v is the (adiabatic) speed of sound in the material, λ_0 is the (vacuum) wavelength of the incoming light and θ is the angle between the incoming and the scattered light. f_B is called the Brillouin frequency. SB gain spectroscopy is a pump/probe laser spectroscopy technique. When the frequency difference $f = f_1 - f_2$ between the crossing probe (f_2) and pump (f_1) laser beams is approaching $\pm f_B$, energy is transferred from the high frequency laser to the low frequency one. Monitoring the probe intensity I_2 as a function of f gives a resonance curve (the Brillouin gain spectrum $g(f)$) of central frequency $\pm f_B$ and width Γ . Note that spontaneous Brillouin scattering was previously used for studying acoustically driven metastable states of water at room temperature[15], but cannot be used here since the acquisition time is expected to be prohibitive for liquid ^4He at 1 K due to the scarcity of thermal phonons.

The pump laser of our SB spectrometer of central wavelength $\lambda_0 = 1064$ nm is pulsed with a frequency repetition rate of 10 Hz and a pulse duration $\tau = 190$ ns (FWHM). τ gives the time scale on which Brillouin frequencies are measured. The probe laser is a single frequency CW laser diode also centered at λ_0 tunable over couple of GHz by modulating its feeding current. The spectral resolution of the experiment is about 3.5 MHz given by the linewidth of the beat note between pump and probe fields. The waists of the lasers are about $w_1 \sim w_2 \sim 20$ μm [12]. To minimize the interaction volume, they are crossed at an angle $\theta \sim 90^\circ$ and then superimposed on the acoustic focus, see Fig.1. Brillouin frequencies are measured on a space scale of $(2w)^3 = 40^3$ μm^3 . Slightly higher temperature and higher frequency measurements suggest the absence of dispersion at our temperature (~ 1 K) and frequency (~ 300 MHz)[16, 17, 18, 19]. We used this SB gain spectrometer to measure the EoS of stable liquid helium-4 ($P > 0$) at $T \sim 1$ K[13] and the experimental EoS obtained is in very good agreement with the theoretical one at $T = 0$ K[20, 21, 22].

The experiment runs as follows. The thickness vibration mode of the transducer is driven by a radio frequency amplifier fed with an arbitrary function generator (AFG). The excitation signal consists in a train of four periods of a harmonic wave voltage of peak-to-peak amplitude V at the transducer resonant frequency f_{ac} . The AFG is triggered by a TTL signal originating from the pulsed pump SB laser at a 10 Hz repetition frequency so that sound pulses are sent into the liquid at a 10 Hz rate. In a given cycle, the delay t_d between the trigger pulse and the beginning of the AFG excitation signal can be varied, see the timeline in Fig.1. The pump laser fires its light pulse of duration τ at a

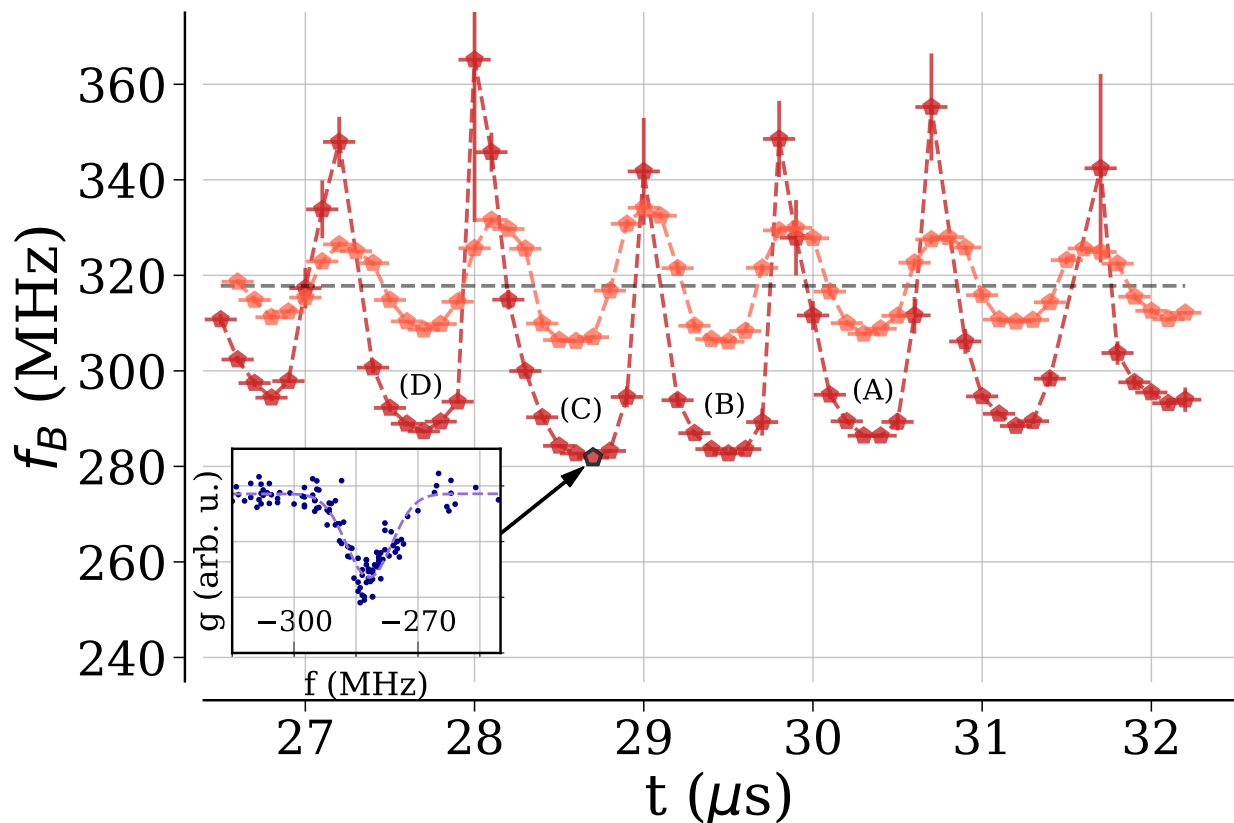


Figure 2: Time evolution of the Brillouin frequencies (f_B) at acoustic focus. Orange circles: driving voltage $V = 150$ mV, Red circles: $V = 540$ mV. The point linking lines are guides to the eye. Horizontal dashed line: static value of the Brillouin frequency $f_B^0 = 317.8(3)$ MHz. (A), (B), (C), (D) mark the different local negative swings of the Brillouin frequency. Inset: Brillouin gain spectrum obtained for $V = 540$ mV at $t = 28.7$ μ s, crosses: data, solid line: gaussian fit[12].

time $t_l \sim 994$ μ s after the trigger pulse with a jitter of about $\Delta t_l \sim 0.5$ μ s. The actual value of t_l in a given cycle is determined by recording and fitting the temporal intensity profile I_1 of the pump pulse. During the time interval τ , the pump and the probe lasers are interacting with the liquid and the temporal intensity profile of the probe laser I_2 is also recorded. The acoustic wave time of flight (ToF) between the inner part of the transducer and the acoustic focus is given by $t = t_l - t_d$ so that by varying t_d , we build 2-D arrays of data $(t, I_2(t))$. We sample these data in time by interval of $\delta t = 100$ ns. One then gets the Brillouin gain spectrum $g(f, t)$ from which we can extract the corresponding value of the Brillouin frequency $f_B(t)$ as detailed in refs[12, 13]. Finally by varying t we are able to record a "movie" of the local variations of the Brillouin frequencies as the wave propagates through the acoustic focus.

Results: Fig.2 displays the time evolution of the Brillouin frequencies at acoustic focus for two different values of the AFG driving voltage. Each measurement of $f_B(t)$ comes from the fit of the corresponding Brillouin spectrum at ToF t . Such a spectrum is shown in the inset of Fig.2. The dashed line corresponds to the value of the Brillouin frequency $f_B^0 = 317.8(3)$ MHz at thermal equilibrium (saturated vapor pressure, $V = 0$ mV). When $f_B(t) > f_B^0$, the liquid at acoustic focus is in a stable state of higher pressure and density. On the contrary, when $f_B(t) < f_B^0$, the liquid is in a metastable state. The different negative swings are labeled (A), (B), (C), (D). At low driving voltage ($V = 150$ mV), the Brillouin frequencies time evolution is sinusoidal as expected when the sound field propagates linearly through the liquid. As V and hence the sound amplitude increase, the sound propagation becomes nonlinear. The effect of the nonlinearity is to flatten the bottom of the wave and to sharpen its crest. This is well known for liquid helium ^4He [23]. However the positive sharp peaks which must exist in the Brillouin frequencies are averaged both in space and time so that they are barely seen for $V = 540$ mV. This is not a major concern for our study as we are interested in the metastable negative pressure states of the liquid which are located in time at the trough of the

acoustic wave. The measured Brillouin frequencies are averaged on a space scale of $2w = 40 \mu\text{m}$ and on a time scale of $\sqrt{\tau^2 + \delta t^2} \sim 220 \text{ ns}$. We can estimate the effect of this averaging by assuming that the Brillouin frequency has the same spatial profile than the density which has been measured in ref.[6]. Doing so, we find that when the local minima are flatter than in the sinusoidal case, the averaged minimum measured Brillouin frequency value is overestimating the local instantaneous one by less than 3 %. Such averaging effects are negligible for the density measurements of ref.[6] (see [24]).

Having these curves, we can easily find the minimum in time of the Brillouin frequency at acoustic focus $f_B^*(V)$ for a given driving voltage V . For the measurement at $V = 540 \text{ mV}$ shown on Fig.2, it is the point at time $t = 28.7 \mu\text{s}$ whose spectrum is presented in the inset.

By increasing the driving voltage V , one can explore deeper and deeper the metastable states of the liquid until a gas bubble nucleates at acoustic focus because of cavitation[4, 6]. This bubble first expands to reach a maximal radius of hundred of μm and then collapses in a typical lifetime of about 1 ms[10] (much longer than the total duration of the acoustic wave) so that it can easily be detected. The bubble signal consists in a dramatic reduction of the pump and probe detected intensities during the bubble lifetime. This signal is actually used for the fine alignment of the Brillouin lasers on the acoustic focus[25]. The creation of the bubble is a stochastic process activated by the thermal fluctuations of the fluid. Its probability is described by the ‘‘asymmetric S-curve formula’’[4]:

$$\Sigma(V) = 1 - \exp[-\ln 2 \exp(\xi(V/V_c - 1))] \quad (2)$$

where V is the excitation voltage, V_c the threshold voltage where the bubble creation probability is 1/2, and ξ a dimensionless parameter. We have determined V_c by measuring the probability of bubble creation as a function of V and fitting the data with eq.(2) as shown in the inset of Fig.3. We found $V_c = 546.6(1) \text{ mV}$.

We have performed several measurements of $f_B(t)$ at this destabilization threshold. The minimum value of the Brillouin frequency f_B^* at the cavitation threshold is $f_B^*(V_c) = 283 \pm 2 \text{ MHz}$.

Both f_B^* and ρ^* (spatio-temporal minimum of the density) are approximately linear with V from $V = 0$ to $V = V_c$ (see Fig.3 and ref.[6]). f_B^* and ρ^* occur in the same swing of the acoustic wave (swing (C))[26]. Setting $v^2 = \left. \frac{\partial P}{\partial \rho} \right|_S$ in eq.(1) and assuming a linear dependence of f_B^* with ρ^* , we can thus estimate the minimum pressure in swing (C) at the $\Sigma = 1/2$ destabilization threshold P_c by integrating eq.(1) at constant entropy S from the static density ρ_0 to the destabilization density $\rho^*(V_c)$. The detail of this calculation is given in the supplemental material [24]

We find $P_c = -5.7 \pm 0.1 \text{ bar}$. This value is in clear contradiction with the one reported in ref.[4] ($-10 < P_c < -8 \text{ bar}$ for $T_{\text{cell}} \sim 1 \text{ K}$). It is also in contradiction with theoretical estimates of bulk cavitation pressure at 1 K $P_c \sim -7 \text{ bar}$ [8, 27, 5].

An EoS of metastable superfluid ^4He at finite temperature has been proposed in ref.[28]. In this study, it is shown that the line of constant entropy in the $P - T$ plane passing through the point $(P_{\text{svp}}(T), T = 1 \text{ K})$ is almost vertical for $-6 \text{ bar} < P < P_{\text{svp}}(1 \text{ K})$. Hence, according to this theory, the temperature during the adiabatic expansion of the liquid down to the cavitation threshold we measure remains almost constant. Consequently, in this pressure range and neglecting any thermal effect related to sound dissipation, the adiabatic and isothermal velocities are approximately equal. Within this approximation, the prediction of the theory is that the EoS at 1 K is essentially the same than the one at 0 K. Then we can use available theoretical equation of states[20, 21, 22] at 0 K in order to convert the measurement of the density at the cavitation threshold of ref.[6] to a pressure value. Doing so, one finds $P_c \simeq -5.1 \text{ bar}$ in disagreement with our estimation. This calls for an accurate experimental determination of the EoS of the metastable states of superfluid ^4He . It must be done by measuring *simultaneously* the density and the Brillouin frequency in the negative pressure domain of the phase diagram and look for a potential deviation of the before assumed linear relation between them.

While measuring the Brillouin frequency near the bubble creation threshold, we have discovered an interesting feature. We are able to determine the swing in which cavitation occurs thanks to a time resolved measurement of the light intensity scattered by the bubble and have noticed that at $V = V_c$, bubbles nucleate in swing (A) of the acoustic wave (see Fig.2). This is quite surprising regarding Fig.2 since, close to V_c , the absolute minimum f_B^* is reached in swing (C). For $V > V_c$, it is still possible to measure Brillouin frequencies in the swings preceding the one in which the bubble has nucleated. Fig.3 displays the local time minima of the Brillouin frequencies $f_{B_{\text{min}}}$ as a function of the driving voltage V . Blue, yellow, red and grey data points respectively correspond to the local minimum of the negative swing labeled (A), (B), (C), (D) in Fig.2. The different symbols correspond to independent runs. As V increases, the time at which the bubble nucleates jumps from one negative swing of the wave to the earlier one. This is represented in Fig.3 by the vertical dashed lines. From the blue to the yellow dashed lines, the bubble nucleates in swing (A) ; from the yellow to the red one it nucleates in swing (B) and over the red line in swing (C). The experiment was not performed at $V > 950 \text{ mV}$ to avoid irreversible damage of the piezo transducer.

In the shaded area of Fig.3 where the probability for bubble detection is 1, the measured Brillouin frequencies are

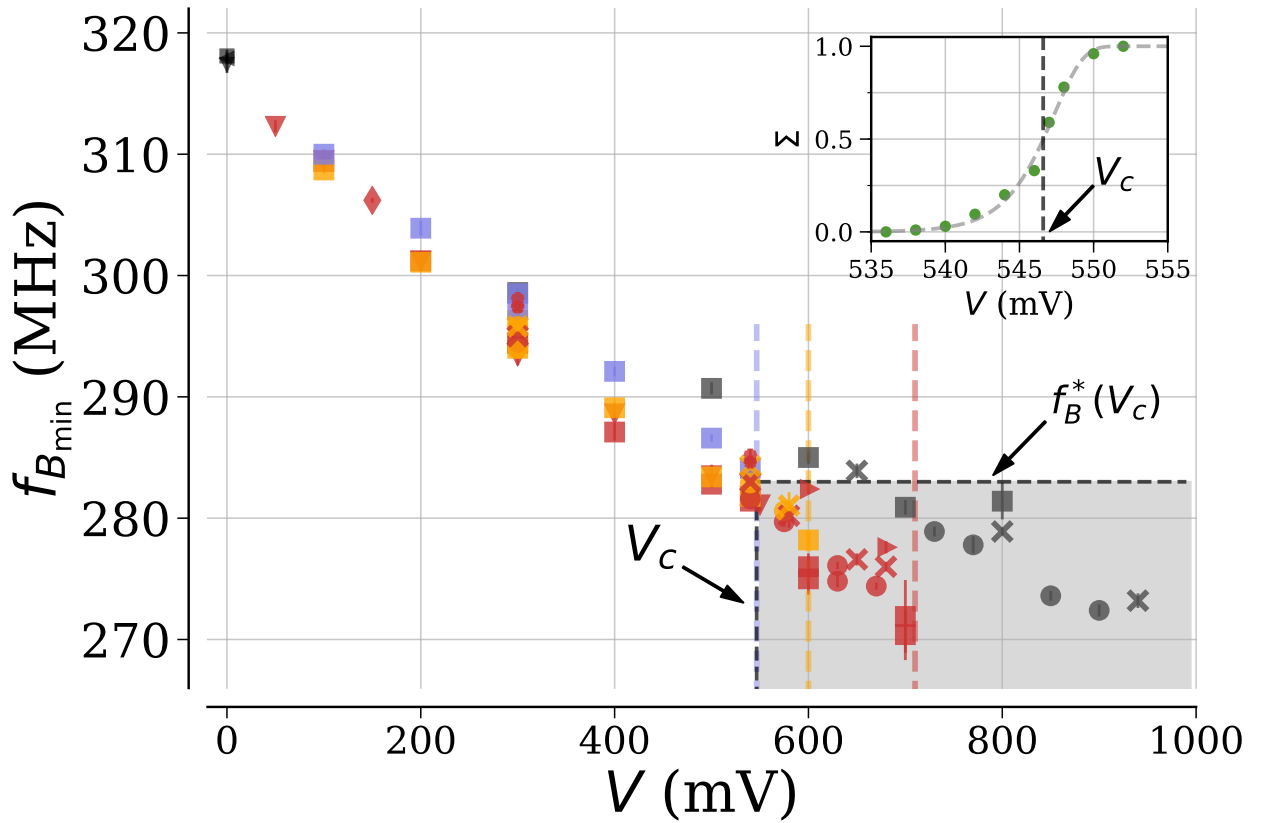


Figure 3: Local time minima of the Brillouin frequencies $f_{B,min}$ as a function of the AFG driving Voltage V . Blue, Yellow, Red, Gray symbols: minimum Brillouin frequency of swing (A), (B), (C), (D) respectively (see Fig.2). The different symbols (crosses, squares, diamonds, circles, triangles) correspond to different runs. Vertical dashed lines: see text. Inset: Bubbles creation probability Σ as a function of V defining V_c : $\Sigma(V_c) = 1/2$.

lower than $f_B^*(V_c)$. This questions the way the cavitation threshold is defined in acoustic driven experiment[4, 6, 9, 3]. In a simple picture of a static cavitation threshold, one could think that the lower possible value of the Brillouin frequency (or pressure or density) of the liquid in the acoustic wave is reached when the bubble detection probability is 1. This is not the case.

One possible explanation for this unexpected finding is that the temperature of liquid helium-4 at focus increases as a function of time during exposure to the sound wave. Indeed, there is attenuation of sound at 1 MHz and at 1 K[29], which can modify the temperature. Nucleation would occur in swing (A) rather than (C), because temperature during swing (A) is higher than during swing (C). For higher voltage, thermal effects are more pronounced, heating rate is more important, and cavitation is triggered earlier. The minimum Brillouin frequency in swing (A) at V_c is 286 MHz. Using our linear extrapolation $f_B(\rho)$ [24] we can estimate the corresponding density and thus find the corresponding pressure by integration of eq.(1). We find -5.3 bar. This value corresponds to a theoretical cavitation pressure at about 1.5 K[8, 27, 5] so that we must invoke a temperature rise of ~ 0.5 K to make the thermal effects responsible for our observation within the frame of available theories. Another scenario is that the acoustic wave is creating vortices while propagating through the liquid and that heterogeneous nucleation occurs on those as proposed in ref.[30]. As for thermal effects, for higher voltage, vortices are created at higher rate and cavitation is triggered at lower time. It is interesting to notice that if we now forget temperature effects, the estimation of the minimum pressure in swing (A) (-5.3 bar) is compatible with a model of cavitation assisted by quantized vortices at 1 K which gives, depending on the vortex density, $-5.8 < P_c^{\text{vortices}} < -5.1$ bar [30]. In this scenario, the extrapolation method of ref.[4] is questionable since vortex creation is not considered in the nucleation mechanism. To further investigate these possibilities, a systematic study of the Brillouin frequencies of the metastable states as function of the number of acoustic pulses and of the acoustic excitation repetition rate would be of great interest.

Finally we shall remark that the absolute minimum value we have reached for the Brillouin frequency is 271 MHz corresponding by integration of eq.(1) to a pressure of -7.4 bar. It is interesting to note that this value is compatible with theoretical estimates of bulk homogeneous cavitation pressure at 1 K [5]. Indeed, for $V > V_c$, cavitation occurs earlier in the wave, and we expect thermal and vortex effects to be less important, and conditions of homogeneous cavitation at 1 K to be approached.

Conclusion: Using a SB gain spectrometer, we have measured the Brillouin frequencies of metastable states of superfluid ^4He around 1 K. Associated with local density measurements[6], our Brillouin frequency measurements enable us to give an estimation of the local pressure at the cavitation threshold in clear contradiction with extrapolated previous experimental estimates. This calls for an accurate experimental measurement of the EoS at $T \sim 1$ K by a simultaneous measurement of the density and the Brillouin frequency of the metastable states of superfluid ^4He . We also have measured Brillouin frequencies beyond the commonly defined cavitation threshold suggesting that thermal effects or vortices could play a significant role in the destabilization process.

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