

Optical measurement of the equation of state of bulk liquid helium-4 around 1 K

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Using a stimulated Brillouin gain spectroscopy technique, we have measured the equation of state of bulk liquid helium-4 at ~ 1 K between 0 and 10 bars. Our results are in very good agreement with previous measurements and with theoretical equations of state. However, our specific method allows one to determine it on space/timescales as narrow as $35 \mu\text{m}/190$ ns. This technique is of particular interest for solving a remaining debate about the value of the homogeneous cavitation pressure of liquid ^4He at $T \sim 1$ K.

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I. INTRODUCTION

Condensed helium-4 is a model material at low temperature and its phase transitions have been studied in much detail. In particular, the stability limits of the liquid phase with respect to the gas phase at negative pressure have been studied experimentally and theoretically (see, for instance, the review articles [1–3]). There is still a debate regarding the value of the pressure at which homogeneous cavitation occurs in liquid helium-4 at about 1 K. Indeed, Caupin *et al.* have developed an indirect method to measure the cavitation pressure of liquid helium-4 and found it to be between -8 and -10 bars at 1 K [4], which is compatible with the spinodal limit $P_{\text{spin}}(1 \text{ K}) \sim -9$ bars [5,6]. This experiment was reproduced by Qu *et al.* which arrived at the same result [7]. In addition, the latter authors have developed an apparatus capable of determining the local density of liquid helium-4 under tension and hence the cavitation density of liquid helium-4 [7]. Together with the self-consistent theoretical equations of state (EOS) of liquid helium-4 at negative pressures [6,8–10], the measurement of the cavitation density at 1 K gives a cavitation pressure of $-5.1(1)$ bars. This is much above P_{spin} and in clear contradiction with the Caupin *et al.* value, but agrees with a model taking into account the presence of quantized vortices [11]. In order to understand this discrepancy, it appears crucial to be able to measure experimentally the EOS of metastable liquid helium and thus check the validity of the theoretical one. However, this is a rather difficult task to achieve mostly because liquid helium-4 metastable states are produced in those experiments using focused MHz acoustic waves. The typical spatiotemporal domain over which the metastable state should be probed is then about $100 \mu\text{m}$ (size of the acoustic focus) and 500 ns (half period of the acoustic wave). Moreover, a nonintrusive technique is needed in order not to destabilize the metastable state. Spontaneous Brillouin scattering was previously used for determining the cavitation pressure of water [12] at room temperature, but cannot be used here since the acquisition time is expected to be prohibitive at 1 K. In this paper we show that we have been able to measure

the equation of (stable) liquid helium-4 at 1 K on such a narrow spatiotemporal domain using an optical method based on stimulated Brillouin gain spectroscopy. This is a crucial step on the route of solving the cavitation pressure debate mentioned above.

II. EXPERIMENTAL SETUP AND PROCEDURE

Brillouin scattering refers to the scattering of light by a transparent medium due to the coupling of incoming photons with phonons of the material [13]. The energy-momentum conservation in the photon/phonon collision imposes that the Brillouin scattered light is frequency shifted by the amount

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

where n is the refractive index, v is the 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. The frequency f_B is called the Brillouin frequency. Stimulated Brillouin (SB) gain spectroscopy is a pump/probe laser spectroscopy technique allowing one to measure the Brillouin frequency of *a priori* any transparent medium. When the frequency difference $f = f_2 - f_1$ between the crossing probe (f_2) and pump (f_1) laser beams is approaching f_B , energy is transferred from the low-frequency laser to the high-frequency one due to the electrostrictive coupling between the light fields and the liquid. Thus, the probe beam gains intensity if $f > 0$ and loses intensity if $f < 0$ ($|f| \sim f_B$). Monitoring the probe intensity as a function of f gives a resonance curve of central frequency $f_{B\pm}$ and width Γ_{\pm} , where \pm is the label for the positive and negative parts of the spectrum.

Recently, we have been able to obtain SB gain (loss) spectra of liquid helium-4 [14] using the experimental arrangement of Fig. 1. The experimental cell is a cubic-shaped block of oxygen-free high thermal conductivity copper with 1-in.-diam BK7 windows on each face perpendicular to the x or y axis of Fig. 1. It contains 30 cm^3 of liquid helium. A calibrated germanium resistor is embedded at the bottom of the experimental cell and allows one to measure the temperature of the liquid with 5 mK accuracy. The cell is cooled in a cryostat with four optical ports. The working

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TABLE I. Brillouin frequency of liquid helium-4 at $T = 0.960(5)$ K as a function of pressure.

P (bars)	0.00	0.48	1.06	1.56	2.07	2.51	3.09	3.59	3.84	4.27	4.60	5.03
f_B (MHz)	307.9	312.6	318.6	323.6	328.4	332.7	337.8	342.1	344.4	348.1	350.9	354.2
Δf_B (MHz)	0.1	0.1	0.1	0.2	0.2	0.2	0.1	0.1	0.1	0.1	0.1	0.1
P (bars)	5.38	5.82	6.21	6.64	7.05	7.54	7.98	8.55	8.93	9.59	10.09	
f_B (MHz)	357.3	360.5	363.9	366.9	370.3	373.8	377.0	381.0	383.7	388.3	391.5	
Δf_B (MHz)	0.1	0.1	0.2	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.2	

where $\gamma = C_P/C_V$ is the ratio between the specific heats of the material respectively at constant pressure and constant volume. We will admit that this relation holds in superfluid helium-4, meaning that we neglect an additional term due to the coupling between the first and second sound whose relative contribution to the sound velocity is of the order of a couple of ppm [20]. The refractive index n is a function of the density ρ via the Lorentz-Lorenz relation

$$\frac{n^2 - 1}{n^2 + 2} = \frac{4\pi}{3} \frac{\alpha}{M} \rho, \quad (3)$$

where M is the molar weight of ^4He (4.0026 g/mole [21]) and α is the molar polarizability which we assume to be independent of pressure for liquid helium-4 at 1 K [22]. In the limit $n \sim 1$, which is a very good approximation for liquid helium, this relation simply becomes

$$n^2(\rho) = 1 + \beta\rho, \quad (4)$$

with $\beta = \frac{4\pi\alpha}{M}$. Including Eqs. (2) and (3) in Eq. (1) and integrating it at a constant temperature T from a known initial state (ρ_0, P_0) to a final state (ρ, P) , one readily gets

$$\int_{\rho_0}^{\rho} \frac{d\rho'}{n(\rho')^2 \sin^2[\theta(\rho')/2]} = \frac{4}{\lambda_0^2} \int_{P_0}^P \gamma(P') \frac{dP'}{f_B^2(P')}. \quad (5)$$

$\theta(\rho)$ may depend upon ρ through the Snell-Descartes law. Actually, the effect is small: The corresponding relative variations of θ are less than 0.01%, much smaller than the accuracy with which we can determine θ_0 (0.1%). Thus $\theta(\rho) = \theta_0$ is considered as a constant. Its value was determined at saturated vapor pressure [$T = 0.960(5)$ K] from our measurement of the Brillouin frequency $f_B = 307.9(1)$ MHz and our measurement of the laser wavelength $\lambda_0 = 1064.46(2)$ nm with a calibrated wavelength meter, and from the value of the speed of the first sound $v_0 = 237.9(1)$ m/s [23] and the value of the refractive index $n_0 = 1.0277(5)$ computed using Eq. (4) for a density of $\rho_0 = 145.1202(3)$ kg/m³ [23]. This gives $\theta_0 = 84.2(1)^\circ$.

Another simplification in integrating Eq. (5) comes from the fact that for liquid helium-4 at 1 K, $\gamma(P)$ is to a good approximation independent of pressure and actually equals 1 to better than 0.04% in the range 0–10 bars at $T \sim 1$ K [24]. The uncertainty on the value of the function $\gamma(P)/f_B(P)^2$ to be integrated in the right member of Eq. (5) is then dominated by the uncertainty on f_B which is about 0.1%. In the frame of these reasonable approximations, Eq. (5) can simply be integrated.

III. RESULTS AND DISCUSSION

Our measurement of the Brillouin frequency of liquid helium-4 at $T = 0.96$ K as a function of pressure are listed in

Table I. The uncertainty Δf_B on the measurement is calculated from the uncertainty obtained on f_{B+} and f_{B-} by the fitting procedure.

The integration of Eq. (5) is done as follows. The left side is directly integrated analytically. For the right side, we preliminary fitted the data $1/f_B^2(P)$ by a polynomial function of P . A third-order polynomial $h(P) = a_0 + a_1P + a_2P^2 + a_3P^3$ gives a satisfactory fit (with a reduced chi square $\chi_{\text{red}}^2 = 1.6$), not significantly better for higher-order polynomials. The values of the coefficients we found are $a_0 = 1.0546(7) \times 10^{-5}$ MHz⁻², $a_1 = -6.92(5) \times 10^{-7}$ MHz⁻² bar⁻¹, $a_2 = 4.2(1) \times 10^{-8}$ MHz⁻² bar⁻², and $a_3 = -1.28(7) \times 10^{-9}$ MHz⁻² bar⁻³. Setting $H(P - P_0) = a_0(P - P_0) + \frac{a_1}{2}(P^2 - P_0^2) + \frac{a_2}{3}(P^3 - P_0^3) + \frac{a_3}{4}(P^4 - P_0^4)$, the EOS is given by

$$\log \left(\frac{1 + \beta\rho}{1 + \beta\rho_0} \right) = \left(\frac{4}{\lambda_0^2} \beta \sin^2(\theta_0/2) \right) H(P - P_0), \quad (6)$$

where log is the natural logarithm. In addition to the a_i coefficients mentioned above, we used the numerical values $P_0 = 10.4(5)$ Pa determined from our temperature measurement $T = 0.960(5)$ K and the properties of liquid helium at saturated vapor pressure [23], $\alpha = 0.1233(3)$ cm³/mole [25].

In order to evaluate the uncertainty on our determination of the $\rho(P)$ function, we have used a Monte Carlo method. We generated, from our measurements, several sets of data ($N \sim 5000$ trials) of the measured pressures and Brillouin frequencies randomly distributed according to their central values and uncertainties. Each set of data was fitted which allowed us to deduce $H(P - P_0)$. Simultaneously, all parameters of Eq. (6) are also randomly varied according to their own uncertainties. Setting a given value for the pressure P , the density $\rho(P)$ of one trial is computed using Eq. (6). Then the standard deviation of $\rho(P)$ of the N trials is computed and this quantity defines our error bar on the $\rho(P)$ value. We then change the value of pressure P for 3000 steps ranging between 0 and 10.1 bars and obtain the corresponding error bars.

Our results are displayed in Fig. 3 together with other equations of state of liquid helium-4 obtained by other methods. The error bars on the different measurements are imperceptible on the scale of the graph. Boghosian *et al.* have measured the dielectric constant of liquid helium-4 as a function of pressure using a resonant LC tunnel diode oscillator filled with the liquid. Assuming the validity of the Clausius-Mosotti relation for the pressure and temperature range they were studying, they were able to convert the dielectric constant measurement into a density measurement [26]. The uncertainty on their measurement of $\rho(P)$ is of about 0.1%. Abraham *et al.* have measured the change with pressure of the transit time of a sound wave traveling over ~ 1 cm distance. Assuming that,

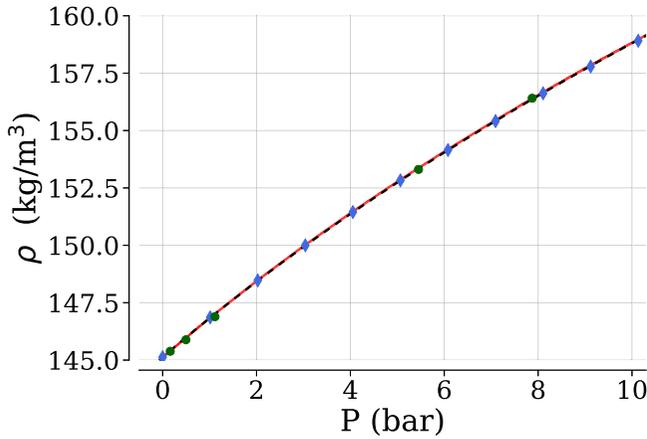


FIG. 3. Equation of state of bulk liquid helium-4. Solid red line: this work ($T = 0.96$ K); blue diamonds: Ref. [22] (experiments, 0.1 and 0.5 K); green circles: Ref. [26] (experiments, 1 K); solid black dashed line: Ref. [9] (theory, 0 K).

in their experimental conditions, the process was isothermal, they were able to determine the equation of state by integrating the isothermal sound velocity over pressure [22]. They performed their measurements at $T = 0.1$ K and $T = 0.5$ K and obtained the same result within the accuracy of their measurement (0.01%). We want to emphasize that these measurements were carried out on spatial and temporal scales orders of magnitude higher than ours. In Fig. 3 is also plotted a theoretical equation of state of liquid helium-4 at $T = 0$ K computed by Dalfovo *et al.* using a density functional method [9]. We should mention that other computational methods have been used to calculate the EOS in the $T \rightarrow 0$ limit [6,8] which all agree with Dalfovo's to within 1% but do not provide an explicit analytic form of the EOS. The agreement between the different measurements and the theory of Dalfovo *et al.* shows that the EOS of liquid helium-4 seems to be relatively independent on temperature between 0 and 1 K [10].

To quantify more the relative agreement between those results, we have computed and plotted in Fig. 4 the quantity $(\rho_m - \rho_d)/\rho_d$, where ρ_m is the density value of a given measurement and ρ_d the corresponding value of Dalfovo *et al.*'s theory. We have not assigned any error to Dalfovo *et al.*'s calculation. For our work, the shaded area represents the confidence interval corresponding to $\pm 1\sigma$ uncertainty. It is seen that all the results are self-consistent within less than 0.1%. If one focuses only on our work and the one of Abraham *et al.*, one sees that the two curves follow the same trend with respect to theory and that the difference between the central values is not exceeding 0.03%, corresponding to an agreement of less than 2σ between both. The slight systematic difference between the two measurements is most likely due to the temperature dependence of the EOS such as the one observed in Ref. [26].

IV. CONCLUSION

We have been able to measure the equation of state of liquid helium-4 by stimulated Brillouin gain spectroscopy. Our measurement, performed on a spatiotemporal scale as short as ($\sim 35 \mu\text{m}$, 190 ns), agrees with satisfactory accuracy

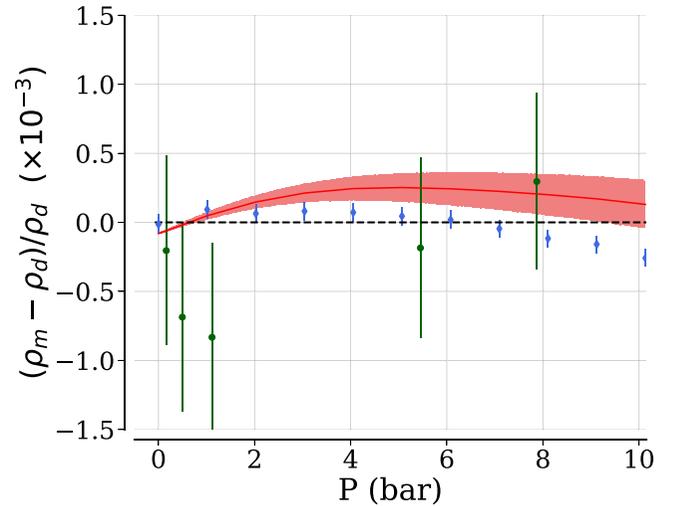


FIG. 4. Relative variations $(\rho_m - \rho_d)/\rho_d$ between the different measurements (ρ_m) and the theory of Dalfovo *et al.* (ρ_d) (see text). Red line: this work; blue diamonds: Ref. [22]; green circles: Ref. [26]. The dashed black zero line corresponds to Dalfovo *et al.*'s theory [9].

with previous measurements of the EOS performed on orders of magnitude greater length scales and timescales. This is an important result and paves the way to solving a remaining debate on the value of the cavitation pressure of liquid helium-4 at 1 K where a measurement of the EOS on space/timescales of ($\sim 100 \mu\text{m}$, ~ 500 ns) is needed. Our result shows that such a measurement is indeed doable. The experiment aiming at measuring the EOS of the metastable states of liquid helium-4 is under construction in our laboratory. Concretely, the setup and method described in this paper will be used to measure the local space/time Brillouin frequencies and simultaneously will be measured the local space/time density with the apparatus presented in Refs. [7,27]. However, there are two differences to consider in the case of the EOS determination of the metastable states ($P < 0$) compared to that of the stable states. The first one arises from the fact that the metastable states are produced at constant entropy (within an acoustic wave). But this is actually even simpler as $(\frac{\partial P}{\partial \rho})_S$ directly appears in Eq. (1) as $v^2 = (\frac{\partial P}{\partial \rho})_S$. The second one is that f_B will be measured and fitted, not as a function of P but as a function of the refractive index n (or density) in order to compute the corresponding pressure instead of fitting f_B as a function of pressure to compute the refractive index (or density). We shall finally notice that such a measurement could also be used to determine the EOS of metastable hcp solid helium-4 also produced by focused acoustic waves where an unexpected instability has been observed [3,28].

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