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

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Abstract 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 states. However our specific method allows for the first time to determine it on space / time scales 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.

1 Introduction

Condensed helium-4 is a model material at low temperature and its phase transitions have been studied in much details. 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, 2, 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 bar and -10 bar at 1 K [4], which is compatible with the spinodale limit $P_{\text{spin}}(1 \text{ K}) \sim -9$ bar [5, 6]. This experiment was reproduced by Qu *et al.* which arrived to 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 self consistent theoretical equation of states (EOS) of liquid helium-4 at negative pressures [8, 9, 6, 10], the measurement of the cavitation density at 1 K gives a cavitation pressure of $-5.1(1)$ bar. This is much above P_{spin} and in clear contradiction with 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 spatio-temporal 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 non-intrusive 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 article we show that we have been able to measure the equation of (stable) liquid helium-4 at 1 K on such a narrow spatio-temporal 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.

2 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:

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

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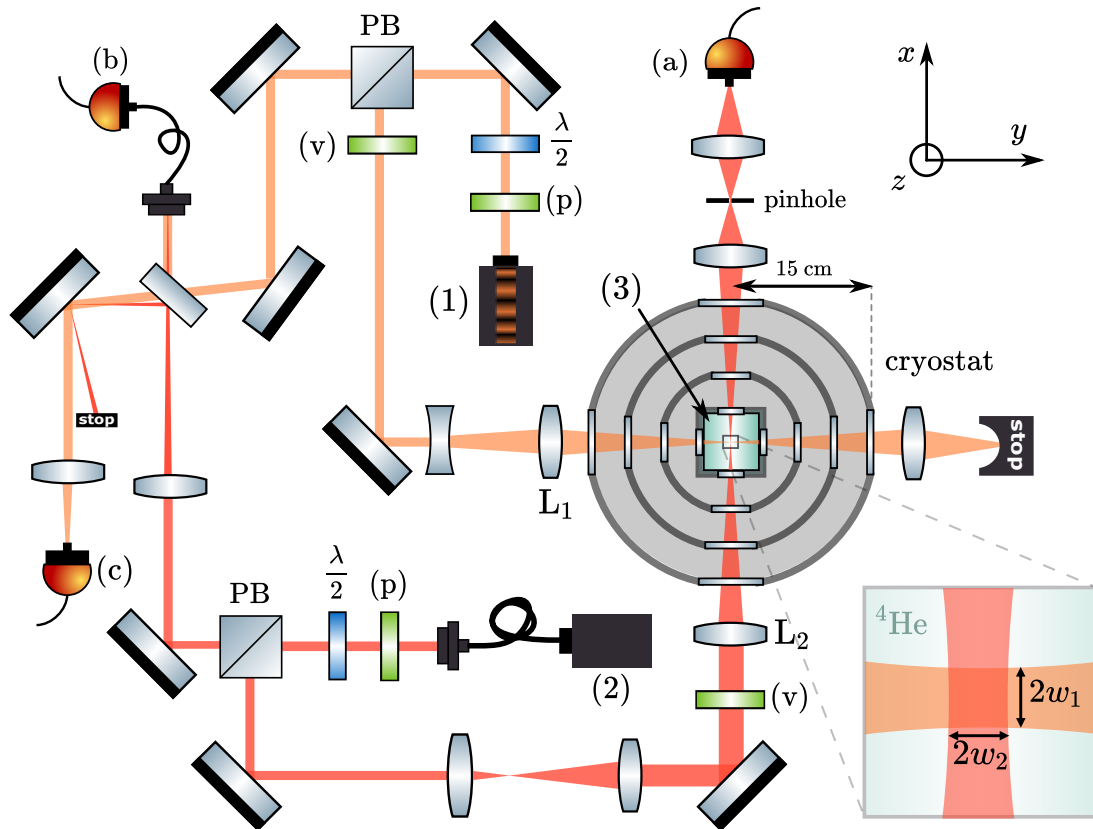


Figure 1: Schematic of the experimental setup. (1): Pump laser (pulsed Nd:YAG); (2): Probe laser (CW frequency tunable diode); (3): Sample (superfluid helium-4); (a) and (c): Pump and probe signal photodiodes; (b): Beating signal fiber-coupled photodiode; (p): Linear polarizer; (v): Vertical polarizer; $\lambda/2$: Half-wave plate; PB: Polarizing beamsplitter; OD: Optical Density; L_1 , L_2 : 150 mm, 200 mm focal length lenses; pinhole aperture: $\text{\O}50 \mu\text{m}$.

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 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 label for the positive and negative part 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 bloc of oxygen-free high thermal conductivity copper with 1 inch diameter 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 to measure the temperature of the liquid with 5 mK accuracy. The cell is cooled in a cryostat with four optical ports. The working temperature can be regulated from 0.9 K to 4 K. In the present experiment, the temperature is fixed at $T = 0.960(5)$ K. The experimental cell is connected to a buffer volume at room temperature, so that the pressure P is easily monitored with the help of a pressure sensor with a resolution of 10 mbar.

The pump laser is a pulsed single longitudinal mode Q-switched Nd:YAG laser of central wavelength ~ 1064 nm. Its pulse duration $\tau = 190$ ns (FWHM) gives the time scale on which the Brillouin frequency and hence the EOS will be measured. The typical instantaneous optical pump power used for this study is ~ 3.5 kW. The probe laser is a single frequency CW laser diode (linewidth ~ 1 MHz) of central wavelength ~ 1064 nm and average power of ~ 20 mW, frequency tunable over couple of GHz by modulating its feeding current.

Both the pump and probe laser beams are split into two secondary beams. One of these secondary beams is used to measure the frequency difference between the two lasers by beating them on photodiode (b) of Fig. 1. We measured

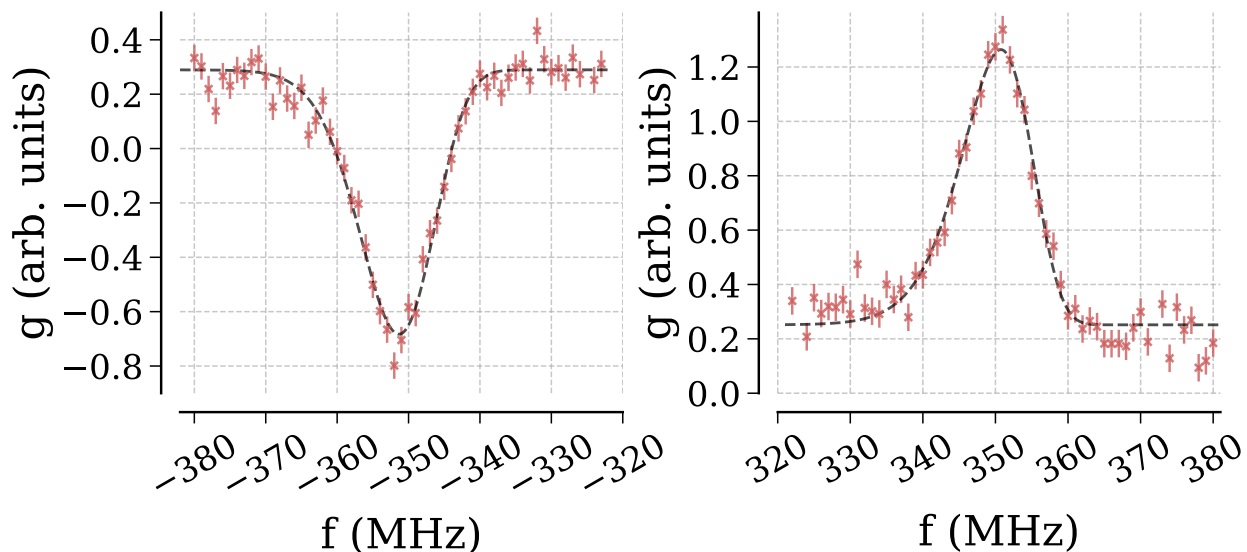


Figure 2: Stimulated Brillouin spectrum of liquid helium-4 ($T = 0.96$ K, $P = 4.60$ bar) obtained on a time scale of ~ 190 ns and a spatial scale of $\sim 35^3 \mu\text{m}^3$. Data points were adjusted by an asymmetric function (see [14]).

that the convolution linewidth of the beating lasers is about 3.5 MHz (FWHM) giving the spectral resolution of the experiment [15]. The other secondary beams, both vertically polarized, are sent towards the experimental cell where the SB gain spectroscopy will be performed. To achieve beam waists of about $20 \mu\text{m}$ in the center of the experimental cell, both probe and pump beams are first expanded and then focused into the cell with lenses all set outside the cryostat. In order to minimise the interaction volume between the lasers, those are crossed at an angle of about 90° . Using a needle head fixed at the bottom of the experimental cell and monitoring the transmission of the laser beams while translating them, we have been able to measure their waists to be $w_1 = 22(2) \mu\text{m}$ and $w_2 = 16(3) \mu\text{m}$ for the pump and probe beams respectively [14]. Those give the length scale on which the EOS will be measured. The probed bulk liquid volume is of about $(2w_1)(2w_2)^2 \sim 35^3 \mu\text{m}^3$. Photodiode (a) is AC coupled and records the signal of the probe laser after its interaction with the liquid and the pump beam. When f approaches f_{B+} , a positive peak of duration τ is recorded by the photodiode (a) due to the energy transfer from the pump pulse to the probe pulse. The peak is negative when f is approaching f_{B-} . The time integral of these peaks defines the value of the Brillouin gain g . Repeating this measurement for different values of f allows us to measure the Brillouin gain spectrum $g(f)$ of the liquid. Such a Brillouin gain spectrum for liquid helium-4 at $T = 0.96$ K and $P = 4.60$ bar is shown in Fig. 2. At a given pressure P , our measurement of the Brillouin frequency is given by $f_B(P) = (f_{B+} + f_{B-})/2$ where f_{B+} (f_{B-}) is the result of the fit of the positive (negative) part of the spectrum by an asymmetric pseudo gaussian function [16] of central frequency f_{B+} (f_{B-}) and width Γ_+ (Γ_-) [14].

As stated by Grubbs *et al.* [17], the observed linewidth is given by $\Gamma_{\text{exp}} = \sqrt{\Gamma_{\text{laser}}^2 + \Gamma_{\text{ang}}^2 + \Gamma_{\text{nat}}^2}$ where Γ_{laser} is the convolution linewidth of the lasers (3.5 MHz for us), Γ_{nat}^2 is the natural linewidth of the resonance depending on the phonon lifetime and the laser wavelength [18] (~ 1 MHz for us), and $\Gamma_{\text{ang}} = 2 \cos(\theta/2)v/(\pi w_{\text{eff}})$ is a purely geometrical contribution, where $w_{\text{eff}}^2 = (w_1 w_2)^2 / (w_1^2 + w_2^2) = (\lambda_0/n\pi)^2 / (\Theta_1^2 + \Theta_2^2)$, Θ_1 and Θ_2 being the (gaussian) beam divergences of the pump and probe beams. The physical origin of the geometrical contribution is the finite size of the laser beam interaction region resulting in a local spread of the excited acoustic wave vectors and thus of the corresponding Brillouin frequencies. This geometrical broadening is largely dominant in our case, of about 8-10 MHz (from 0 to 10 bar). The Grubbs formula gives an expected linewidth (FWHM) of about 9-11 MHz, not far from the 10-13 MHz we observe.

In this work we focus on the central value of the Brillouin frequency as a function of the pressure P at a given temperature T (0.96 K). Indeed the measurement of f_B , here obtained with a relative accuracy of about 0.1%, gives access to the EOS of liquid helium-4. For a material of density ρ at a pressure P , the (adiabatic) sound velocity v is linked to the isothermal compressibility of the material through the relation [19]:

P (Bar)	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 (Bar)	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	

Table 1: Brillouin frequency of liquid helium-4 at $T = 0.960(5)$ K as a function of pressure.

$$v^2 = \gamma \left(\frac{\partial P}{\partial \rho} \right)_T \quad (2)$$

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 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 ${}^4\text{He}$ (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 Eq. (2) and Eq. (3) in Eq. (1) and integrating it at 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 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 to 1 to better than 0.04% in the range 0 to 10 bar 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.

3 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 Tab. 1. 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 $a_0 = 1.0546(7) \cdot 10^{-5}$ MHz⁻², $a_1 = -6.92(5) \cdot 10^{-7}$ MHz⁻²bar⁻¹, $a_2 = 4.2(1) \cdot 10^{-8}$ MHz⁻²bar⁻², $a_3 = -1.28(7) \cdot 10^{-9}$ MHz⁻²bar⁻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 svp [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 set 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 a 3000 steps ranging between 0 and 10.1 bar and obtain the corresponding error bars.

Our results are displayed in Fig. 3 together with other equations of states 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, 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 emphasise 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 [8, 6] which all agree to Dalfovo's within 1 % but don't 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 like the one observed in Ref. [26].

4 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 spatio-temporal scale as short as (~ 35 μ m, 190 ns), agrees with satisfactory accuracy with previous measurements of the EOS performed on orders of magnitude greater length and time scales. This is an important result and paves the way of 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/time scales of (~ 100 μ 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 article 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 Ref. [27, 7]. 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 $\left(\frac{\partial P}{\partial \rho}\right)_S$ directly appears in Eq. (1) as $v^2 = \left(\frac{\partial P}{\partial \rho}\right)_S$. The second one is that f_B will be measured and fitted, not as a function of P but as 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

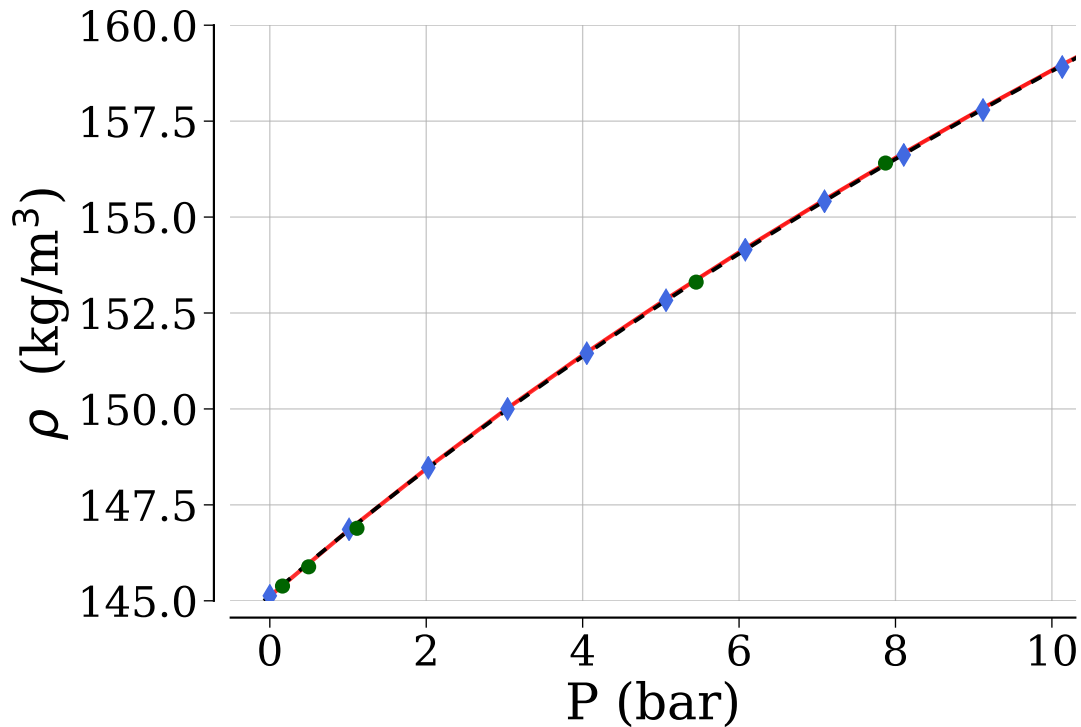


Figure 3: Equation of state of bulk liquid helium-4. Solid red line: this work ($T = 0.96$ K), blue diamonds: Ref. [22] (experiments, 0.1 K and 0.5 K), green circles: Ref. [26] (experiments, 1 K), solid black dash line: Ref. [9] (theory, 0 K)

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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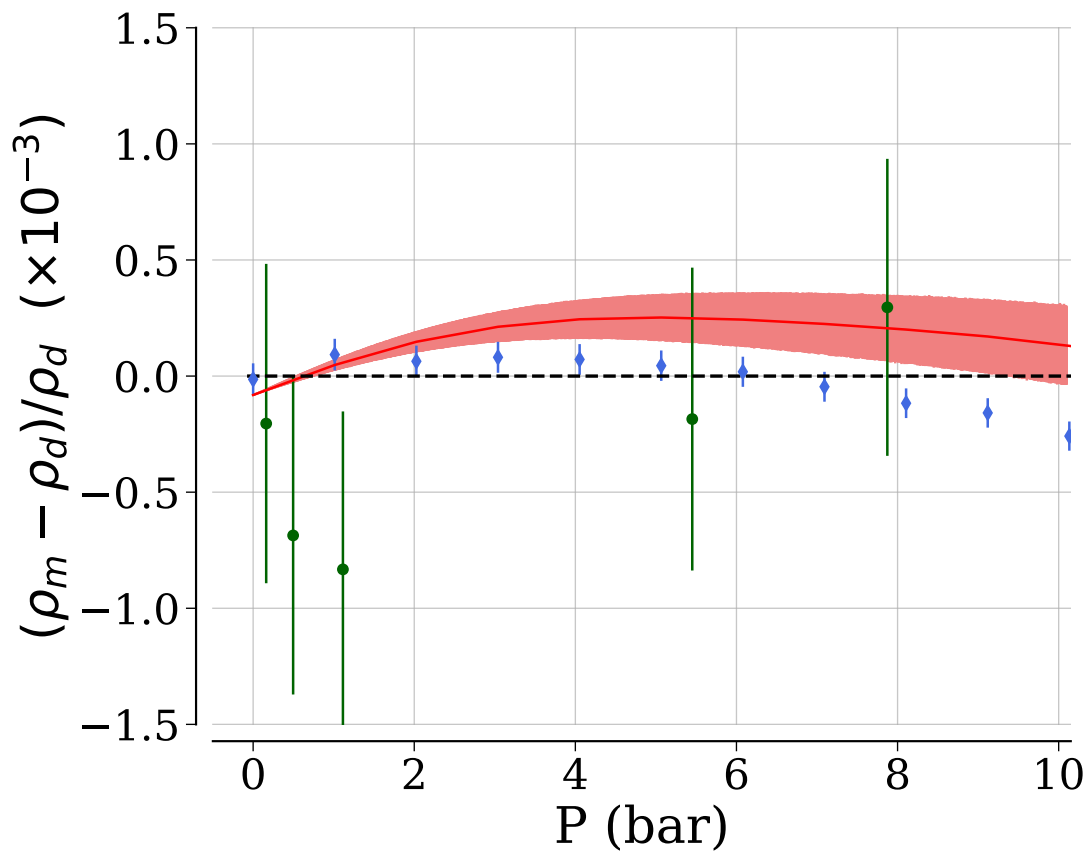


Figure 4: Relative variations $(\rho_m - \rho_d)/\rho_d$ between the different measurements (ρ_m) and the theory of Dalfovo *et al.* (ρ_d), see text. Red line: this work, blue diamonds: Ref. [22], green circles: Ref. [26]. The dashed black zero line corresponds to Dalfovo *et al.*'s theory [9].

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