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# Stimulated Brillouin Gain Spectroscopy of Superfluid Helium-4

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## Abstract

We report Stimulated Brillouin gain spectra of bulk superfluid helium-4. Stimulated Brillouin gain spectroscopy is a powerful optical method to probe the acoustic properties of liquid helium in a narrow spatio-temporal domain with moderate optical power and fast acquisition rates. We determine the velocity of first sound of superfluid helium-4 from the Stimulated Brillouin gain spectra between 0.90 K and 2.17 K and compare it to well known available data, which validate our experimental method.

## 1 Introduction: Brillouin scattering in liquid helium-4

Brillouin Scattering corresponds to the inelastic scattering of light by acoustic waves (phonons) in a transparent material [1]. When analysed, the frequency and the linewidth of the scattered light contain valuable information on the scattering material such as the speed of sound and the phonon lifetime. Energy-momentum conservation requires that photons scattered at an angle  $\theta$  from an incoming light interacting with the material are frequency-shifted by an amount :

$$\Omega_B = 2n\omega \frac{v}{c} \sin(\theta/2) \quad (1)$$

where  $n$  is the index of refraction of the material,  $\omega$  the angular frequency of the incoming light,  $v$  the speed of sound in the material,  $c$  the speed of light in vacuum.  $\Omega_B$  is called the angular Brillouin frequency. Thus knowing  $n$ ,  $\omega$  and  $\theta$  and measuring  $\Omega_B$  provide a way to determine the speed of sound in the sample. The spread in angular frequency, called Brillouin linewidth and noted  $\Gamma_B$ , is directly related to the phonon lifetime.

Brillouin scattering is therefore a powerful tool to optically probe the acoustic properties of transparent media. It has naturally been applied to liquid helium-4 in different configurations.

**Spontaneous Brillouin scattering:** The first way to determine Brillouin frequency and linewidth in liquid helium-4 is to perform a spontaneous Brillouin scattering experiment. It consists of focusing a single CW laser beam into the liquid and of collecting the light scattered at angle  $\theta$  by the (scarce) thermal phonons of the liquid. The scattered light is then frequency analysed to obtain the corresponding spectrum. Pike *et al.* were the first to publish spontaneous Brillouin spectra of liquid helium-4 in 1970[2]. They used an Argon laser ( $\lambda = 488$  nm) and a Fabry-Pérot interferometer as a frequency analyser to measure Brillouin frequencies of about 670 MHz. The same year, St Peters *et al.* have published sound velocities and sound attenuation coefficients of superfluid helium-4 under its saturated vapour pressure between 1.25 K and  $T_\lambda$  obtained by spontaneous Brillouin scattering [3]. The probed phonons were about 750 MHz where the ruby laser light was spectrally analysed using again a Fabry-Pérot interferometer. Spontaneous Brillouin scattering was also actively used in pressurised liquid <sup>4</sup>He or in mixtures of <sup>3</sup>He and <sup>4</sup>He, in particular to get a Brillouin signature of second sound propagation [4, 5, 6, 7, 8].

The main difficulty in obtaining spontaneous spectra in liquid helium-4 is the very low acquisition rate as the one for instance mentioned by Pike *et al.* of about 30 minutes to get a single spectrum [2]. It has the advantage to inject into the sample laser power of typically hundreds of mW, much lower than the one needed for Stimulated Brillouin scattering experiments which we shall describe now.

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**Stimulated Brillouin scattering:** The second optical way to probe acoustic properties of liquid helium is to perform a stimulated Brillouin scattering experiment. When the incoming light field is intense enough (above a given power threshold), the light itself can generate phonons in the material. This is due to the nonlinear coupling between the light field and the material: acoustic waves are driven by the interference of the incoming and spontaneous retro-reflected light fields, through the process of electrostriction. Then an amplification of the light scattered at  $\theta = 180^\circ$  naturally occurs: this is the Stimulated Brillouin Scattering (SBS) effect in the so-called "generator" configuration. In the limit of constant incoming intensity  $I_1$ , the intensity of the retro-reflected light  $I_2(L)$  is given by:

$$I_2(L) = I_2(0) \exp(g_0 I_1 L) \quad (2)$$

where  $I_2(0)$  corresponds to the spontaneous scattered light intensity,  $L$  is the typical interaction length between the optical and acoustic fields and  $g_0$  the so-called Brillouin gain factor expressed in m/W. The latter parameter depends on the photoelastic/absorptive properties of the medium and on the phonon lifetime. It is generally admitted that  $I_2(0)$  is given by  $f \cdot I_1$  where  $f \sim 10^{-12}$  in conventional materials [9] and probably lower in liquid helium-4. Thereby, in order to obtain measurable ratios  $I_2(L)/I_1$ , the necessary incoming power should lie in the MW range, which requires the use of pulsed laser (typically with a pulse duration of tens or hundreds of nanoseconds).

Stimulated Brillouin scattering in liquid helium-4 was first observed by Winterling *et al.* [10], quickly followed by Abrikosova *et al.* [11, 12]. Winterling *et al.* determined the threshold power for SBS in liquid helium-4 to be about 3 MW [10, 13]. The measurement of the Brillouin frequencies ( $\sim 700$  MHz) was done using a Fabry-Pérot interferometer. They deduced first sound velocities from their Brillouin frequencies, in good agreement with previous standard 12 MHz ultrasonic measurements showing that there was no dispersion in the velocity of first sound of liquid helium-4 between 12 MHz and 700 MHz. Just after this study, the same group used stimulated Brillouin scattering to measure hypersonic attenuation in liquid helium-4 at about 650 MHz between 1.2 and 4.2 K [14]. For that purpose, they measured, as a function of temperature, the intensity of a second light pulse (probe pulse), delayed by a given time from the initial stimulated Brillouin pulse creating the phonons. A few years later, Berberich *et al.* improved the latter method by monitoring the temporal decay of the probe pulse intensity, allowing a more accurate determination of the phonon lifetime. This way, they were able to measure the velocity and attenuation of phonons excited at about 700 MHz in superfluid helium-4 at saturated vapour pressure and at 23 bar [15].

The main advantage of using stimulated Brillouin scattering consists of reducing the acquisition rate, as a single or a couple of laser pulses are sufficient to obtain the Brillouin frequency and linewidth. The price to pay though is the need to inject MW pulses of light into the liquid. This sets a limit for using SBS in liquid helium-4 with a tightly focused laser as optical breakdown [16, 17] might occur before the generation of any SBS signal.

**Stimulated Brillouin gain spectroscopy:** There is actually a third way to obtain a Brillouin spectrum of a transparent medium. This consists of applying stimulated Brillouin scattering in the so-called "amplifier" configuration. In that case, two light fields are overlapped in the medium. One is referred as the pump field (intensity  $I_1$  (W/m<sup>2</sup>), angular frequency  $\omega_1$ ) and the other one as the probe field ( $I_2$ ,  $\omega_2$ ). When  $\Omega = \omega_2 - \omega_1$  is approaching  $\Omega_B$ , the probe intensity after interaction with the pump and the medium over a distance  $L$  follows an exponential growth ( $\Omega > 0$ ) governed by Eq. (2), where  $g_0$  has to be replaced by a resonant function  $g(\Omega)$ . It must be noted that if  $\Omega < 0$ , the Brillouin gain factor is negative and energy is transferred from the probe to the pump field, resulting in an exponential decay of the probe field intensity. The Brillouin gain factor  $g(\Omega)$  is expected to be a Lorentzian function of  $\Omega$  of central angular frequency  $\Omega_B$  and linewidth  $\Gamma_B$ , where  $\Gamma_B$  is given by the phonon lifetime [9]. Stimulated Brillouin Gain (Loss) Spectroscopy (SBgs) consists of measuring the  $g(\Omega)$  function in order to access  $\Omega_B$  and  $\Gamma_B$ . To that purpose, the ratio  $I_2(L)/I_2(0)$  is measured as a function of  $\Omega$  by tuning one of the laser frequencies.

Stimulated Brillouin gain spectroscopy combines the advantages of each of the precedent configurations. First, being a stimulated scattering based technique, acquisition rates of Brillouin spectra are orders of magnitude greater than the ones obtained with spontaneous Brillouin scattering. Second, significant signals can be obtained with orders of magnitude less incoming optical power than in the single laser stimulated case. This is simply because the initial scattered intensity  $I_2(0)$  in Eq. (2) corresponds directly to the probe intensity and is not constrained by the relation  $I_2(0) = f \cdot I_1$ .

Stimulated Brillouin gain spectroscopy has been applied to numerous materials (see for instance [18, 19, 20]) and is extremely active nowadays in the area of probing acoustic properties of biological samples [21, 22] where low optical powers and fast acquisition rates are mandatory.

However, until now, it has not been applied to the case of liquid helium-4. In this article, we report stimulated Brillouin gain spectra obtained in bulk superfluid helium-4 at saturated vapour pressure between 0.90 K and 2.17 K. The spectra are obtained on space/time scales of about 40  $\mu\text{m}$  and 190 ns. This is of interest for probing the acoustic properties of liquid helium-4 subjected to repetitive fast transient phenomena on small length scales.

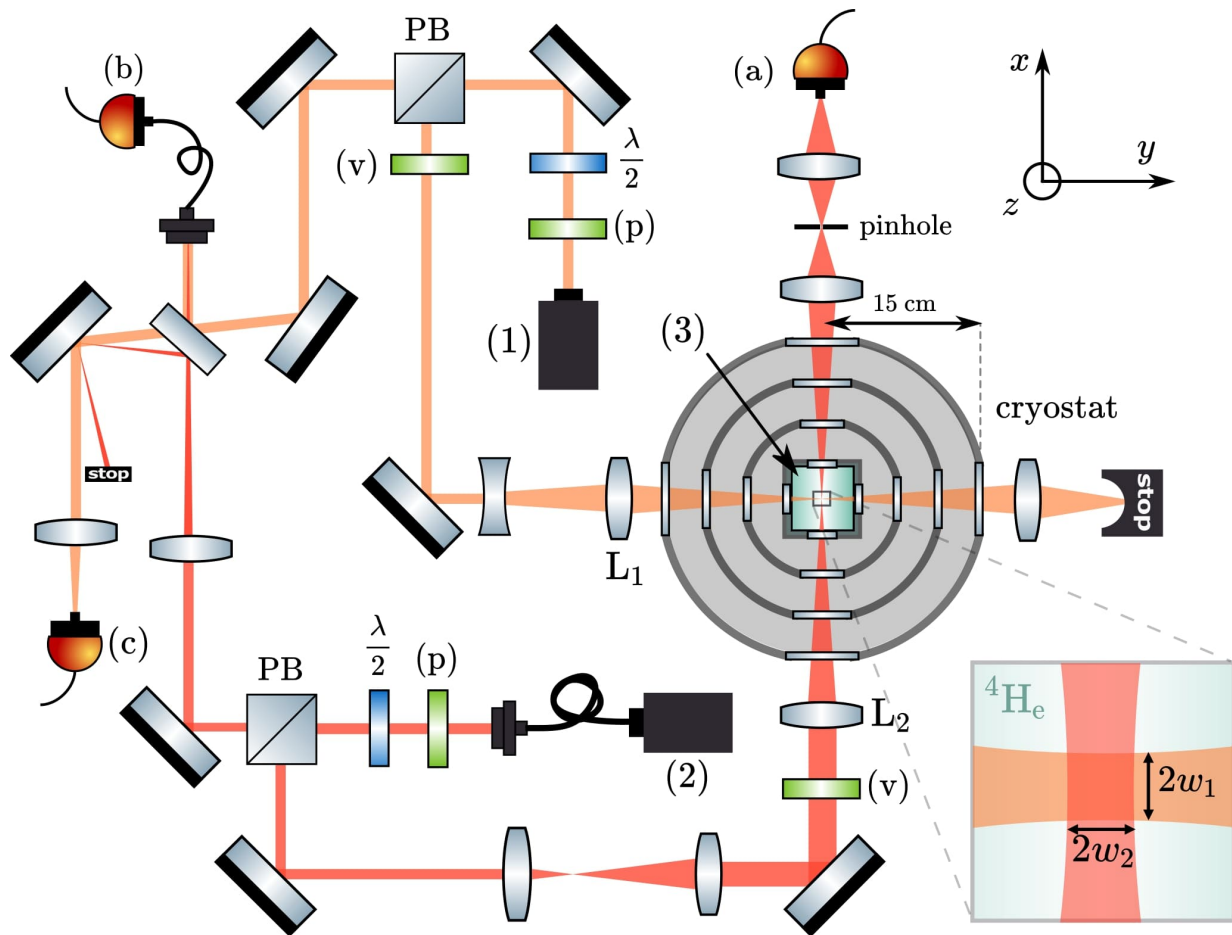


Figure 1: Schematic of the experimental setup. (1): Pump laser (pulsed Nd:YAG); (2): Probe laser (CW frequency tunable diode); (3): Sample (superfluid helium-4); (a) and (c): Pump and probe signal photodiodes; (b): Beating signal fiber-coupled photodiode; (p): Linear polarizer; (v): Vertical polarizer;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

## 2 Experimental setup and procedure

The experimental setup is shown in Fig. 1. The experimental cell containing liquid helium-4 is cooled in a He-4 pumped cryostat with optical access from four directions. The working temperature can be regulated from 0.9 K to 4.2 K.

The SBgs pump laser ( $\omega_1$ ) is a pulsed single longitudinal mode Q-switched Nd:YAG laser of central wavelength 1064 nm. The Full Width at Half Maximum (FWHM) pulse duration  $\tau \sim 190$  ns defines the time scale on which the SBgs will be performed and hence on which the acoustic properties of liquid helium-4 are probed. The typical instantaneous power used for this study is 3.5 kW, corresponding to a pulse energy of about 0.7 mJ.

The probe laser ( $\omega_2$ ) is a  $\sim 20$  mW tunable single frequency (1 MHz linewidth) CW Distributed Feedback Laser diode of central wavelength 1064 nm. Its frequency can be tuned over couples of GHz by scanning its feeding current while maintaining its temperature fixed using a standard Peltier PID loop scheme.

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) (5 GHz bandwidth). We measured the convolution linewidth of the beating lasers to be about 3.5 MHz (FWHM) giving the spectral resolution of the experiment. Assuming that the linewidth of the probe is about 1 MHz as stated by the manufacturer, this ensures the spectral-monochrome character of the pump laser, whose spectral linewidth is indeed of the order of  $1/\tau$ . Another fraction of the pump beam is picked up and sent to a fast DC-coupled photodiode (photodiode (c)) in order to monitor the power of the pump beam.

The two other secondary beams, both vertically polarized are focused in the experimental cell and overlapped at a crossing angle  $\theta \sim 90^\circ$  [23] using converging lenses of 150 mm and 200 mm focal lengths. Prior to that, the pump and probe beams are expanded with telescopes in order to match their diameters at focus. The probe intensity is

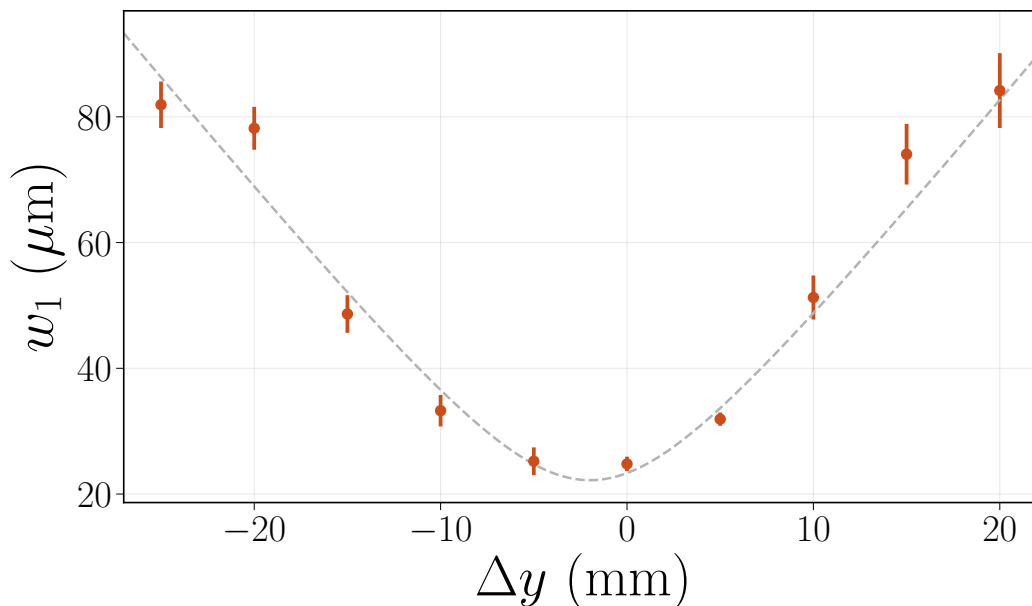


Figure 2: Measurement of the pump beam diameter (inside superfluid helium-4) at the position of the alignment needle as a function of the position of the focusing lens  $L_1$  along the pump beam propagation direction ( $y$  axis in Fig. 1).  $\Delta y = 0$  mm is an arbitrary initial position of  $L_1$  along the  $y$ -axis.

monitored using photodiode (a). In order to reduce the parasitic signals on photodiode (a) due to residual reflections of the pump beam into the cell, the probe beam is spatially filtered after passing through the cryostat using a 100 mm focal length lens associated with a  $\text{\O}50$   $\mu\text{m}$  pinhole aperture. The probe beam signal is then electronically filtered (high-pass filter, cutoff frequency of 50 kHz) to get rid of the slow signal variations due to the slight deviation of the beam by mechanical vibrations. This filter also eliminates the DC component of the probe signal which is typically of the order of 7.5 V.

An alignment head (not shown in the figure) consisting of a needle tip with a base diameter of 0.69 mm has been fixed at the bottom of the experimental cell. It was used both to easily overlap the pump and probe beams and to measure their respective waists. More precisely in the case of the pump beam, the focusing lens  $L_1$ , is mounted on translation stages such as the focusing point of the laser beam can be moved in the three directions of space. By intercepting the laser beam with the alignment head and by studying the transmission signal, we were able to evaluate the diameter of the probe beam using the standard knife-edge method [24]. Such measurements, for the pump beam, are presented in Fig. 2: the waist of the beam is determined from a fit of the diameters along the propagation axis assuming gaussian beam propagation. For the pump and probe beams we found respectively  $w_1 = 22 \pm 2$   $\mu\text{m}$  and  $w_2 = 16 \pm 3$   $\mu\text{m}$ .

After this measurement, each beam waist is placed on the tip of the alignment needle. We then translated each beam upward by 100  $\mu\text{m}$  in order to avoid any residual reflection on the needle. The lasers are now focused in the bulk of the liquid and their overlapping volume is about  $(2w_1)(2w_2)^2 \sim 35^3$  ( $\mu\text{m}$ )<sup>3</sup>. This is the spatial scale on which the SBgs will be performed and on which the acoustic properties of liquid helium-4 are probed.

### 3 Experimental results and discussion

SBgs spectra are obtained as follows. For different values of the feeding current of the probe laser corresponding to different values of  $\Omega = \omega_2 - \omega_1$ , the AC component of the temporal intensity of the probe pulse is recorded by photodiode (a). Three such signals measured at  $T = 0.98$  K are shown in Fig. 3. In Fig. 3a),  $\Omega \approx \Omega_B$  and a positive peak of duration 190 ns is detected. This peak is the signature of the SBS gain effect and corresponds to the energy transfer from the low frequency field (in that case the pump  $\omega_1$ ) to the high frequency field (in that case the probe  $\omega_2$ ). The energy transfer naturally occurs only when the pump and probe beams are interacting and hence only during  $\tau \sim 190$  ns. Fig. 3c) corresponds to a negative value of  $\Omega$  so that  $\Omega \approx -\Omega_B$ . In that case, the energy is transferred from the probe (low frequency field) to the pump (high frequency field), leading to a negative peak. In Fig. 3b),

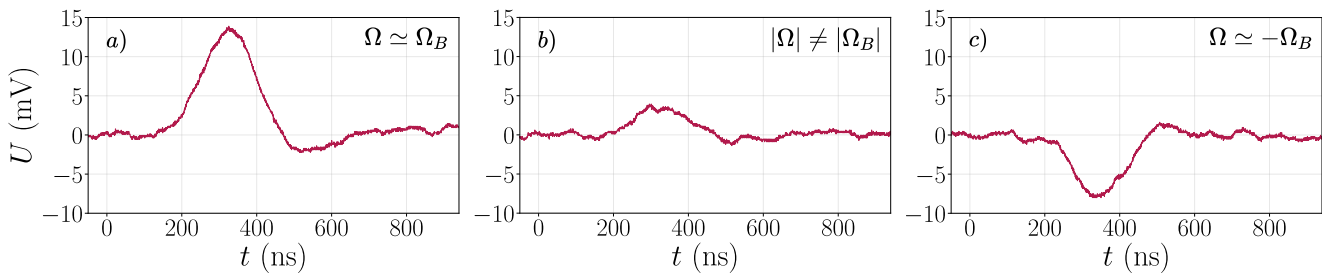


Figure 3: AC coupled photodiode (a) probe signal recordings at  $T = 0.98$  K in case: a)  $\Omega \approx \Omega_B$ , b)  $\Omega \neq \Omega_B$ , c)  $\Omega \approx -\Omega_B$ . The positive peak in a) and negative peak in c) are signatures of the SBS gain (loss) effect. The small positive peak in b) is a background signal and is independent of  $\Omega$ .

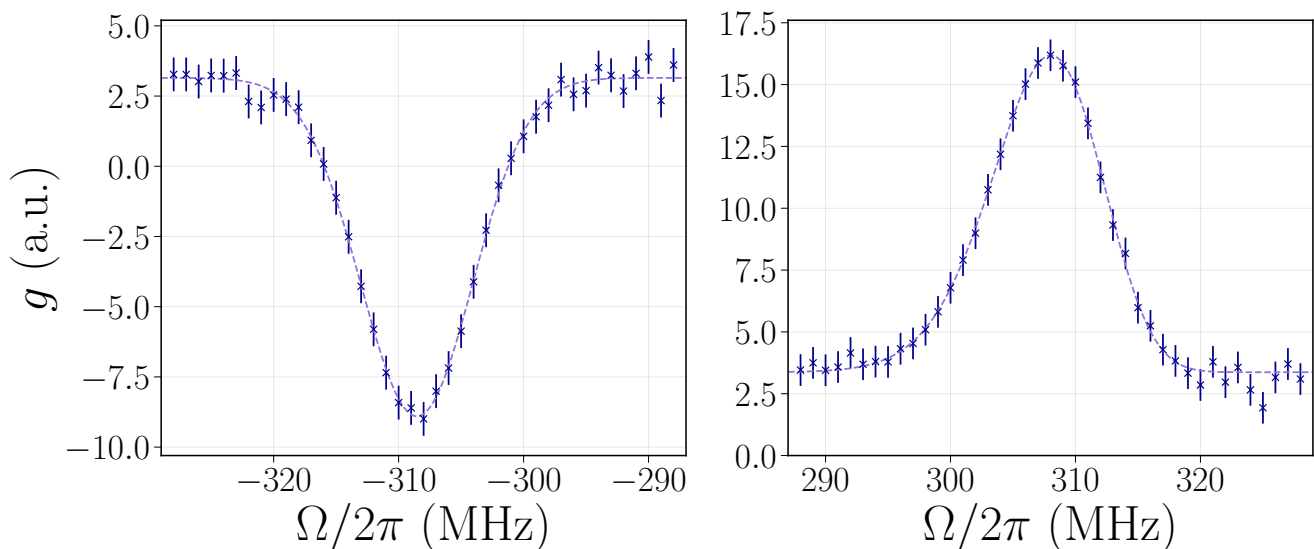


Figure 4: Stimulated Brillouin gain spectra of liquid helium-4 at  $T=0.98$  K (saturated vapour pressure) for  $\Omega < 0$  (left) and  $\Omega > 0$  (right). Data points were adjusted by an asymmetric function (see text).

the lasers are far detuned from the Brillouin frequency ( $\Omega \neq \Omega_B$ ), only a small parasitic signal from the pump beam reflections in the cell is detected. The amplitude of this background signal is independent of  $\Omega$  and just results in an additional offset in the Stimulated Brillouin gain spectra measurements.

We define the experimental Brillouin gain  $g(\Omega)$  in arbitrary units as being the time integral of such signals divided by the pump intensity to normalize it against laser intensity fluctuations. Applying this definition to the various signals obtained for various values of  $\Omega$ , we obtain the SBGs spectra. Data points are averaged in one MHz increments (less than the 3.5 MHz spectral resolution of the beating lasers), the error bars being given by the corresponding standard deviation of the averaged measurements. Two Brillouin gain spectra of liquid helium-4 at  $T=0.98$  K (saturated vapour pressure) are presented in Fig. 4 for  $\Omega < 0$  and  $\Omega > 0$ .

We first comment on the observed line shape. The linewidth is of the order of 10 MHz (FWHM). This is much larger than what is expected from the inverse phonon lifetime contribution. Setting  $\Delta\nu_B = \Gamma_B/2\pi = 1/(2\pi\tau_p)$  where  $\tau_p$  is the phonon lifetime, one has  $\Delta\nu_B \propto \Omega_B^2$  where  $\Omega_B$  is the angular Brillouin frequency and hence the excited phonon angular frequency. At about 1 K, Berberich *et al.* have measured  $\Delta\nu_B \sim 2.4$  MHz for  $\Omega_B/2\pi \sim 700$  MHz [15]. Using this value to estimate the expected value of  $\Delta\nu_B$  in our case, one finds  $\Delta\nu_B \sim 0.4$  MHz (as we have excited phonons at about  $\Omega_B/2\pi \sim 300$  MHz), lower than the observed 10 MHz linewidth. Indeed, the measured linewidth not only depends on the Brillouin linewidth but also on the laser convolution linewidth  $\Delta\nu_{\text{laser}}$  and on a geometrical linewidth  $\Delta\nu_{\text{ang}}$ . Studying the angle dependence of the observed linewidth in Brillouin scattering experiment, Grubbs *et al.* argued that the observed linewidth must be given by [25]:

$$\Delta\nu = \sqrt{\Delta\nu_{\text{laser}}^2 + \Delta\nu_{\text{ang}}^2 + \Delta\nu_B^2} \quad (3)$$

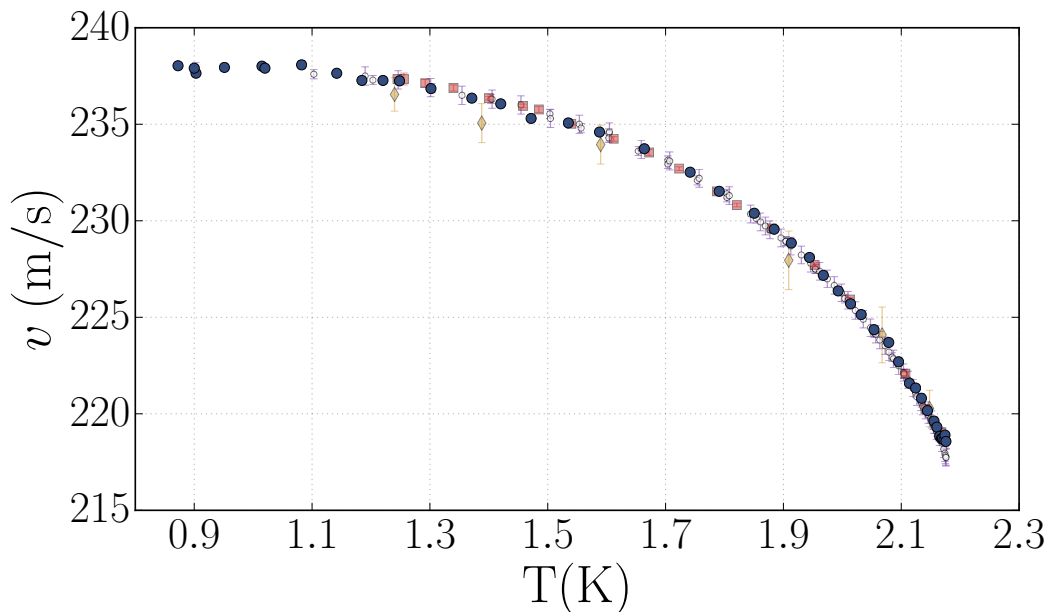


Figure 5: First sound velocity in superfluid helium-4 as a function of temperature. Solid circles: our measurement based on Stimulated Brillouin gain spectroscopy (error bars are smaller than the symbols). The temperature of the liquid is measured using a calibrated germanium resistor embedded at the bottom of the experimental cell. Open circles: tabulated values from [27]. Squares and Diamonds: measurements obtained from Spontaneous Brillouin Spectroscopy in Refs. [3] and [28], respectively.

where  $\Delta\nu_{\text{ang}} = 2v \cos(\theta/2)/(\pi w_{\text{eff}})$  for pure gaussian beams with  $v$  the speed of sound and  $w_{\text{eff}}^2 = (w_1 w_2)^2 / (w_1^2 + w_2^2)$ ,  $w_1$  and  $w_2$  being the laser waists. The physical origin for this geometrical broadening 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. In our configuration with  $\theta \sim 90^\circ$  and  $w_{\text{eff}} = 13 \mu\text{m}$ , one finds  $\Delta\nu_{\text{ang}} \sim 8.3 \text{ MHz}$ . Adding  $\Delta\nu_{\text{laser}} \sim 3.5 \text{ MHz}$  and  $\Delta\nu_B \sim 0.4 \text{ MHz}$ , Eq. (3) predicts  $\Delta\nu \sim 9 \text{ MHz}$ , close to what we actually observe.

As  $\Delta\nu_{\text{laser}} \gg \Delta\nu_B$  and as the spectrum of the beating lasers is gaussian, we initially fitted the SBg spectra by a gaussian profile of central angular frequency  $\Omega_{B_+}$  ( $\Omega_{B_-}$ ) and FWHM  $\Gamma_+$  ( $\Gamma_-$ ), where  $\pm$  labels for the positive part ( $\Omega > 0$ ) and negative part ( $\Omega < 0$ ) of the spectrum. Doing so, we however observed a systematic difference in the values obtained for  $\Omega_{B_+}$  and  $\Omega_{B_-}$ . It is typically of about  $(\Omega_{B_+} - \Omega_{B_-})/2\pi \sim 1 \text{ MHz}$ , quite important in regard to the error bars on each independent fit with  $\Delta\Omega_{B_\pm}/2\pi \sim 0.2 \text{ MHz}$ . A careful analysis of the gaussian fits shows that they fail in determining precisely the central frequency since the line shape is slightly asymmetric. Better results can be obtained by fitting by an asymmetric gaussian profile, as used commonly in spectroscopy experiments [26], where one allows the width of the gaussian function to vary with the frequency according to :

$$\Gamma(\Omega) = \frac{2\Gamma_0}{1 + \exp(a(\Omega - \Omega_B))} \quad (4)$$

where the  $a$  parameter is a measure of the asymmetry. By this method, the systematic difference  $(\Omega_{B_+} - \Omega_{B_-})/2\pi$  was reduced to  $0.4 \text{ MHz}$ , which is of the order of magnitude of the error bars. The physical grounds of the residual difference might be of interest but is out of the scope of the present article. In this work, we define the measured Brillouin frequency as:  $\Omega_B = (\Omega_{B_+} + \Omega_{B_-})/2$ .

Using Eq. (1), the measurement of  $\Omega_B(T)$  should give the first sound velocity at saturated vapour pressure provided that  $\theta$  and the refractive index  $n(T)$  are known. This latter one is computed using the Clausius-Mossotti relation and the tabulated values of the density at saturated vapour pressure [27]. The value of  $\theta$  was determined from a single measurement of  $\Omega_B$  at  $T = 0.96 \text{ K}$  (svp) and from the corresponding tabulated value of the speed of sound ( $237.9(1) \text{ m/s}$  [27]). This gives  $\theta = 84.1(1)^\circ$ .

Fig. 5 shows the deduced speed of first sound from our SBgs measurements at svp between  $0.90 \text{ K}$  and  $2.17 \text{ K}$ . It is compared to tabulated values [27] and to previous measurements of the speed of sound from Spontaneous Brillouin scattering experiments [3, 28]. As can be seen in the figure, our values compare well to others, the difference being of the order of  $0.2\%$ .



## 4 Conclusion

We have reported the first Stimulated Brillouin gain spectra obtained in bulk superfluid helium-4. This pump/probe spectroscopy technique enables one to optically measure the velocity of first sound of liquid helium-4 using moderate optical power and fast acquisition rates. Provided that one manages to resolve the natural Brillouin spectrum linewidths, the technique can also be used to study bulk superfluid helium sound attenuation coefficients. Our specific configuration (laser crossing angle of  $\sim 90^\circ$ ), as well as the short pump pulse duration and the tight laser focusing, allows us to probe the sound velocity in a narrow spatio-temporal domain ( $40 \mu\text{m}$  and  $190 \text{ ns}$ ). This is of interest as this technique appears as a promising tool for measuring the first sound velocity and hence the compressibility of the metastable states (negative pressure states) of liquid helium-4 produced by focused MHz acoustic waves on space/time scales of about  $100 \mu\text{m}$  and  $500 \text{ ns}$  [29, 30].

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