# Pressure dependence of the low-momentum phonon dispersion relation in liquid <sup>4</sup>He<sup>+</sup>

W. R. Junker and C. Elbaum

Department of Physics, Brown University, Providence, Rhode Island 02912

(Received 14 July 1976)

The pressure dependence of the low-momentum phonon dispersion relation in liquid <sup>4</sup>He has been determined from an analysis of experiments on the interactions between sound waves and thermal phonons. Specifically, changes  $\Delta C/C$  in sound velocity were studied in the frequency range 1-15 MHz for pressures  $0 \le P \le 20$ atm, at temperatures  $0.1 \le T \le 0.45$  K. The results were found to be in agreement with phonon dispersion, for small momenta, of the form  $\epsilon = Cp(1 + \gamma p^{2} \cdots)$ , with  $\gamma$  positive at low pressures. At saturated vapor pressure, the present study yields a value  $\gamma = (16 \pm 1) \times 10^{37}$  cgs;  $\gamma$  decreases linearly with increasing density and reaches zero at a density corresponding to a pressure of ~ 18 atm.

## I. INTRODUCTION

The nature of the low-momentum phonon dispersion relation in liquid <sup>4</sup>He has been the subject of considerable interest and speculation in recent years, especially since this region (for wave vectors  $q \leq 0.2 \text{ Å}^{-1}$ ) is not measured reliably by inelastic neutron scattering. This interest was extended by a suggestion of Maris and Massey<sup>1</sup> that measured ultrasonic attenuation in liquid <sup>4</sup>He is well accounted for through the use of a dispersion relation of the form

$$\epsilon = Cp(1 + \gamma p^2 + \cdots), \qquad (1)$$

with  $\gamma > 0$  (where  $\epsilon$  is energy, p is momentum, and C is the sound velocity). In fact, it has since been shown that using this assumption yields better qualitative and quantitative agreement between calculated and measured ultrasonic attenuation and velocity changes than was obtained with previous theories. These theories had used the same form of dispersion relation, but with  $\gamma < 0$ . The earlier calculations of attenuation which yielded results closest to experiments were based on three-phonon interactions (acoustic phonon + thermal phonon - thermal phonon). With  $\gamma < 0$ , however, such an interaction is forbidden by energy and momentum conservation requirements. To skirt the shortcoming of nonconservation, the concept of lifetime broadening was introduced, with the result of relaxing the strict conservation requirement. A direct consequence of assuming the new form of the dispersion relation ( $\gamma > 0$ ) is that the threephonon interaction becomes explicitly permitted within conservation of energy and momentum. By the same token the interaction between thermal and acoustic phonons can no longer be characterized by a single collision time. The theoretical solution for the attenuation and velocity change thus becomes more complicated to calculate.

The present study was motivated by the fact that

the shape of the low-momentum phonon dispersion relation in liquid <sup>4</sup>He can be determined indirectly from an analysis of the interactions between sound waves and thermal phonons. In particular, measured changes,  $\Delta C$ , in the sound velocity C as a function of frequency lend themselves to such an analysis through the use of the Boltzmann transport equation.

A calculation of both the attenuation and velocity change, utilizing a dispersion relation of the above form, in which  $\gamma > 0$ , was carried out by Maris.<sup>2</sup> This calculation involved a numerical solution of the Boltzmann equation. It was performed for a temperature of 0.35 K and for frequencies f between 1 and 1000 MHz. The calculated attenuations were in good agreement with the measurements. In addition, this calculation produced a complicated variation of the velocity change  $\Delta C/C$  with frequency. Below 10 MHz there appeared a maximum in  $\Delta C/C$ , while at a frequency near 100 MHz  $\Delta C/C$  exhibited a minimum. For frequencies above 100 MHz,  $\Delta C/C$  increased monotonically to a limiting value. Likewise, at very low frequency  $\Delta C/C$  decreased to a limiting value. Such a dependence of  $\Delta C/C$  on frequency did not result from calculations utilizing  $\gamma < 0$  and a single collision time; these theories produced a  $\Delta C/C$  which monotonically increased with frequency from a low-frequency limit to a high-frequency limit.

Indication of unexpected dependence of  $\Delta C/C$  on frequency had been observed in experiments. In particular, measurement of  $\Delta C/C$  for frequencies below 12 MHz had been performed by Whitney and Chase.<sup>3</sup> Their results for temperatures above 0.4 K suggested that  $\Delta C/C$  increased with increasing frequency as had been predicted by then existing theory. Measurements at frequencies above 12 MHz performed by Abraham *et al.*<sup>4</sup> suggested a different frequency dependence; they found that  $\Delta C/C$  became smaller for higher frequencies. Measurements by Roach *et al.*,<sup>5</sup> again for f > 12

162

15

MHz, suggested that the situation was even more complicated. They found that for temperatures below 0.212 K,  $\Delta C/C$  increased with increasing frequency, while above 0.3 K  $\Delta C/C$  was that found by Abraham *et al.*<sup>4</sup>

An accurate measurement of  $\Delta C/C$  within the frequency range below 15 MHz and for temperature less than 0.4 K, combined with existing data, could then be used to test the validity of the calculation of  $\Delta C/C$  with  $\gamma > 0$  and, in addition, shed light on the magnitude of  $\gamma$ .

With this in mind we undertook such experiments. In particular, we have performed measurements as a function of temperature T, frequency f, and pressure P, for  $0.1 \le T \le 0.5$  K,  $1 \le f \le 15$  MHz, and  $P_s \le P \le 20$  atm, where  $P_s$  stands for saturated vapor pressure (SVP).

In what follows, Sec. II is devoted to experimental technique, while in Sec. III we describe and discuss the results. Our conclusions are given in Sec. IV.

# **II. EXPERIMENTAL TECHNIQUE**

#### A. Experimental chamber

The experimental and mixing chambers, in which our experiments were performed, were one assembly. As is shown in Fig. 1, the mixing chamber was an annular space which surrounded the cylindrical experimental chamber. To improve thermal contact to the <sup>4</sup>He inside the experimental



FIG. 1. Sample and mixing chamber.

chamber, copper braid was silver soldered to the inner wall of the mixing chamber. One end of the experimental chamber was soldered into position with low-melting-point solder which permitted the removal of the end cap without damaging the joints of the mixing chamber. To facilitate the removal of the transducers, the remaining end was sealed by means of an indium *O* ring. For experiments at elevated pressure, a superfluid leak-tight valve was added to the chamber to permit measurement at constant volume.

The interior of the experimental chamber contained the ultrasonic cell, consisting of two X-cut quartz transducers separated by a Pyrex glass spacer whose ends were ground flat and parallel, to within better than  $2 \times 10^{-4}$  rad. The surfaces of the spacer were coated with a conducting film to facilitate the grounding of the interior faces of the transducers and to damp the sound waves transmitted through the glass spacer. Above the top transducer, sufficient volume was incorporated to insure that the chamber could not be easily overfilled during the measurements at SVP.

The <sup>4</sup>He for the experiments was condensed into the chamber from a cylinder of <sup>4</sup>He gas, after passing through a liquid-nitrogen sieve trap which removed contamination, such as air, water, or oil vapor. No attempt, however, was made to decrease the content of <sup>3</sup>He below that naturally present in the <sup>4</sup>He gas.

Measurements of the sample temperature was accomplished by three thermometers. Two of these were mounted in copper blocks which were fastened through a layer of varnish to the outside of the mixing chamber. The third thermometer was mounted inside the <sup>4</sup>He sample chamber. This thermometer cooled to the same ultimate temperature that was indicated by the thermometers on the outside of the chamber and showed no indication of warming when the ultrasonic pulses were applied to the sample. The sensitivity and reproducibility of our temperature measurements is better than one part in 10<sup>3</sup>; the uncertainty in the absolute value (calibration) is much larger and is estimated to be 1%.

#### B. Velocity measuring system

The system used to determine the change in velocity is based on a pulsed ultrasonic interferometric technique of a type described by Blume.<sup>6</sup> In this system, the frequency of a free-running oscillator is locked to the reciprocal of the round trip time of an echo in the sample. Under such a scheme, the velocity change manifests itself as a change in the frequency of the oscillator. The basic system is summarized in Fig. 2. We have a



FIG. 2. Block diagram of ultrasonic interferometer used for sound-velocity measurements.

voltage-controlled oscillator from which we gate the signal that is applied to the sample. Sufficient time is allowed between the gated pulses so that there is not overlapping of the echo trains. The signal received from the sample is amplified by the receiver and applied to a phase detector, the reference signal for which is taken from the same voltage-controlled oscillator. The phase-detected signal is then fed into a box-car integrator. The portion of the echo of interest is then sampled. averaged, and converted into a dc voltage. This voltage is applied to the control port of the voltage-controlled oscillator, completing the loop. The phase-detected echo of interest is set at quadrature and the system maintains this phase relation. Once in operation, the system "lock" condition is  $ft_n = \text{const}$ , where f is the oscillator frequency and  $t_n$  is the time delay of the received echo. If  $t_n$  is determined solely by the propagation delay in the sample, then

$$\Delta f/f = \Delta C/C , \qquad (2)$$

where  $\Delta f$  and  $\Delta C$  are the changes in frequency of the oscillator and velocity of sound in the sample, respectively. Under these conditions the velocitychange measurement reduces to the measurement of a frequency change.

The total delay of the signal is given by

$$t_n = t_s + t_t + t_\gamma + t_\phi , \qquad (3)$$

where  $t_s$  is the actual transit time in the sample,  $t_t$  is the propagation delay through cables and receiver,  $t_{\gamma}$  is the delay caused by phase shifts at the sample-transducer interface, and  $t_{\phi}$  is the delay caused by phase shifts in tuned elements of the system. For our experiment, we can neglect  $t_{\gamma}$ due to the large acoustic impedance mismatch between the sample and transducer, so that  $t_n = t_s$  $+t_t + t_{\phi}$ . To see how these additional times affect the results, we note that

$$\frac{\Delta f}{f} = \frac{\Delta t_s + \Delta t_t + \Delta t_{\phi}}{t_s + t_t + t_{\phi}} .$$
(4)

We also assume that the propagation delay through nontuned portions of the system is independent of frequency and experimental conditions, so that  $\Delta t_t = 0$ . We write the expressions for  $t_s$  and  $t_{\phi}$ 

$$t_s = l/C , (5)$$

where l is the sample length, and for each tuned element

$$t_{\phi} = \frac{Q}{\pi} \frac{f - f_r}{f f_r} , \qquad (6)$$

where Q is the quality factor of the tuned element,  $f_r$  is the resonant frequency of the element, and f is the frequency at which the system is operated. The change in  $t_{\phi}$  due to the change in operating frequency  $\Delta f$  is then

Equation (4) is then written

$$\frac{\Delta f}{f} \left( 1 + \frac{t_t}{t_s} + \frac{Q}{\pi f_r t_s} \right) = \frac{\Delta C}{C} , \qquad (7)$$

so that our measurements of  $\Delta f/f$  must be multiplied by

$$\left(1 + \frac{t_t}{t_s} + \frac{Q}{\pi f_r t_s}\right) \tag{8}$$

to give accurate measurements of  $\Delta C/C$ .

To obtain a numerical value for this correction, we assumed that  $t_s$  is known, since the velocity of sound in <sup>4</sup>He is well documented, and we can measure the spacing between our transducers. We then simulate a velocity change with a delay line. Knowing the amount of the delay and the transit time in the sample, we calculate the expected  $\Delta f / f$ . The measured  $\Delta f / f$  differs from the calculated value by the correction factor.

For our measurements at SVP the correction factors were found to depend on the particular set of transducers used in the measurements. In particular those measurements utilized two sets of transducers with resonant frequencies of 1 and 3 MHz. The appropriate correction factors  $(\eta)$  were thus

$$\eta_1 = 1.15 \quad (f_r = 1 \text{ MHz}),$$
  
 $\eta_3 = 1.09 \quad (f_r = 3 \text{ MHz}),$ 

and were essentially independent of frequency for the particular set of transducers used. For measurements at elevated pressure the experimental conditions were such that corrections were different for each measuring frequency.

<u>15</u>

Under certain circumstances the velocity measurements exhibit a spurious dependence on the amplitude of the signal. One source of this difficulty is a direct consequence of the operation of the interferometer, that can be thought of as a servo-system in which the frequency is controlled by the transit time. This type of servo-system requires an offset between the frequency and transit time to maintain control. The magnitude of the offset is determined by the loop gain of the system. A change in the loop gain will cause a shift in the offset and a spurious change in the frequency. Such a change in loop gain occurs whenever there is a change in the attenuation of the medium measured. Thus, when the attenuation change was large enough to cause a measurable effect on the velocity change, the receiver was gain controlled.

15

### **III. RESULTS AND DISCUSSION**

#### A. Experimental results

Measurements of  $\Delta C/C$  were conducted as a function of temperature for each frequency, and for comparison with the results of recent calculations the data are presented as the dependence of  $\Delta C/C$  on frequency at a given temperature. To facilitate further this comparison, the data at SVP for temperatures near 0.25, 0.30, 0.35, 0.400, and 0.450 K have been interpolated to these temperatures. This procedure was carred out both graphically and by means of an analytical interpolation, assuming  $T^4$  dependence of  $\Delta C/C$  (at a given frequency). The results from both these techniques are in close agreement with each other.

The calculation of Maris<sup>7</sup> and the measurements of Roach *et al.*<sup>5</sup> with which we make comparisons, are at constant volume. The present vapor-pressure data were, therefore, converted to constant volume through the Grüneisen relation.

We have referenced our measurements of the velocity change to T = 0. For our purposes, T = 0 is taken to be some temperature T < 0.1 K which within the resolution of the present measurements is well inside the range where the sound velocity is independent of temperature. Comparison of measurements at different frequencies relies on the velocity of sound at T = 0 being independent of frequency. Furthermore, within the frequency range of ultrasonic waves considered here the departure of the phonon dispersion relation from linearity is negligible. Thus, measured dependence of  $\Delta C/C$  on frequency does not require any corrections for the phonon dispersion.

The results of the measurements of  $\Delta C/C$  for frequencies of 1, 3, 5, 9, and 15 MHz, at various temperatures, are plotted for saturated vapor pressure in Fig. 3. Included in these graphs are



FIG. 3. (a) Measured sound-velocity change  $\Delta C/C$  as a function of frequency, at saturated vapor pressure, for temperatures of 0.350 and 0.400 K. (b) Same as (a) for temperatures of 0.250 and 0.300 K. In both parts the points at 12, 15, 36, 45, 84, and 105 MHz are from Roach *et al.* (Ref. 5). The solid lines are drawn freehand through the experimental points.

the results of Roach *et al.*<sup>5</sup> For our measurement, the resolution of  $\Delta C/C$  (the uncertainty in  $\Delta C/C$ due to uncertainty in  $\Delta f/f$ ) is less than the scatter between repeated measurements of  $\Delta C/C$ , so that the worst error assumed for the present data is that of the scatter between measurements.<sup>8</sup>

The salient features can be summarized as follows. Below 15 MHz there exists a maximum in  $\Delta C/C$  as a function of frequency. The maximum moves toward higher frequencies as the temperature increases. These results are not consistent with a simple frequency dependence of  $\Delta C/C$  as suggested by calculations using the single collision time approximation.<sup>9</sup> Rather, a description as proposed by Maris,<sup>2</sup> is in qualitative agreement with the data, supporting the assumption that  $\gamma > 0$ . A more complete comparison with calculations is presented below.

We now examine the effect upon  $\Delta C/C$  of increased pressure above the <sup>4</sup>He. Measurements have been carried out for pressures of 4, 8.4, 10, 12, 15, and 20 atm over the same temperature range as was used in the measurements at SVP. The maximum frequency at which the measurements have been performed is 13 MHz. (As a result of the lowered attenuation, at elevated pressure, it was possible to use the 1-MHz fundamental-frequency quartz transducer to obtain measurements with acceptable sensitivity up to a frequency of 13 MHz.) The results of the  $\Delta C/C$  measurements are shown for the different pressures in Figs. 4-8. Several features are obvious. First, as the pressure increases, there is a general decrease in the magnitude of  $\Delta C/C$  for any particular temperature and frequency. Also, the maximum in  $\Delta C/C$  as a function of frequency persists in the frequency region similar to that for saturated vapor pres-



FIG. 4. Measured sound-velocity change  $\Delta C/C$  as a function of frequency, at 4 atm, for several temperatures, as indicated. The uncertainties are less than, or at most equal to the size of the symbols, except at 1 MHz where for each temperature two separate points appear; these represent the maximum scatter occurring at this frequency only, because of limitations in the equipment used.



FIG. 5. Same as Fig. 4, at 8.4 atm. Data at 12, 15, 36, 45, 84, and 105 MHz are from Roach *et al*. (Ref. 5).

sure, finally disappearing for pressures above 12 atm. Again, the position of this maximum shifts to higher frequencies for increased temperature.

### B. Discussion

Ultrasonic studies, unlike, for example, inelastic-neutron-scattering studies, can only lead to an understanding of the phonon dispersion relation within the framework of a theoretical model which





166



FIG. 7. Same as Fig. 4, at 12 atm.

explicitly incorporates a dependence upon the dispersion relation. The model which we consider has been proposed by Maris,<sup>7</sup> who used a dispersion relation of the form

$$\epsilon = Cp \left[ 1 + \gamma p^2 \left( \frac{1 - (p/p_A)^2}{1 + (p/p_B)^2} \right) \right], \tag{9}$$



FIG. 8. (a) Same as Fig. 5, at 15 atm; points for frequencies above 13 MHz are the data of Roach *et al.* (Ref. 5), obtained at 15.5 atm. (b) Same as Fig. 4, at 20 atm.

where  $\gamma$ ,  $p_A$ , and  $p_B$  are adjustable parameters. This form was selected because a simple polynomial dispersion model, containing only linear and cubic terms, does not provide a satisfactory representation of the higher-energy phonons which are involved in the scattering processes. (A polynomial expansion including higher-order terms could provide a complete description of the phonon dispersion relation, however, such expansions tend to be cumbersome to use and determining the coefficients of the higher-power terms is tedious.) Equation (9) allows for the adjustment of the terms  $p_A$  and  $p_B$  to provide agreement for high-momentum phonons with neutron scattering data, while leaving the parameter  $\gamma$  to be adjusted for agreement between calculations and present measurements. The calculations of Maris<sup>7</sup> for  $\gamma = 8$  and  $\gamma = 10 \times 10^{37}$  cgs units, along with our data and those of Roach et al.<sup>5</sup> are shown in Fig. 9 for temperatures 0.25 and 0.35 K. The calculations are seen to be in qualitative agreement with the measurements. Also, as the temperature increases, the position of the maximum in the velocity-change curve shifts to higher frequency. Quantitative agreement, however, is lacking. We have calcu-



FIG. 9. Comparison of experimental results with calculations based on Eq. (9), for saturated vapor pressure.

lated, therefore, the velocity change as a function of frequency, with the use of the computer program developed by Maris, for temperatures of 0.25 K and  $\gamma = 15 \times 10^{37}$  cgs; the results are also shown in Fig. 9(a). We note that agreement is improved; however, a slightly larger value of  $\gamma$  may provide

more satisfactory agreement.<sup>10</sup> Recently, Meier, Beck, and Beck<sup>11</sup> and Wehner<sup>12</sup> have also calculated the change in velocity as a function of frequency. The calculation by Wehner<sup>12</sup> provides a reasonable fit to the data of Roach et al.<sup>5</sup> over the temperature region 0.2-0.4 K, with  $\gamma = 18 \times 10^{37}$  cgs. On the other hand, the calculation of Meier, Beck, and Beck<sup>11</sup> best fits our data, and that of Roach et al.<sup>5</sup> for the temperatures 0.25 and 0.35 K with  $\gamma = 10 \times 10^{37}$  cgs. (In a broader theoretical context, the observed dependence of  $\Delta C/C$  on frequency has been treated by a number of investigators in terms of a resonant interaction between the ultrasonic and thermal phonon populations.<sup>11-14</sup> While the results of these treatments differ in details, they are very similar in their qualitative features, and we have selected for direct comparison with our experiments those which lend themselves most readily to this operation.)

Measurements of the thermodynamic properties of <sup>4</sup>He can also be used to deduce features of the dispersion relation. The thermodynamic quantity of most direct interest for the phonon dispersion relation is the heat capacity of the liquid, whose measurement by Philips, Waterfield, and Hoffer<sup>15</sup> (PWH) provided the first experimental evidence that  $\gamma > 0$ . In their analysis, PWH found that the best fit to the data was provided by a dispersion relation in which  $\gamma = 4.1 \times 10^{37}$  cgs. A more recent analysis by Brooks and Donnelly<sup>16</sup> has shown, however, that it is possible to obtain satisfactory agreement with PWH when  $\gamma$  is significantly larger (up to  $23 \times 10^{37}$  cgs), depending on what is assumed for the values of the coefficients of the higher-order terms in a polynomial form of the dispersion relation.

Another thermodynamic quantity which provides information about the dispersion relation is the thermal-expansion coefficient. The analysis of this type of data, however, requires a knowledge of the density dependence of the coefficients in the dispersion relation. The most sensitive measurement of this type has been performed by Berthold *et al.*<sup>17</sup> From their experiment, they concluded that  $\gamma = (15.3 \pm 3.26) \times 10^{37}$  cgs units, in close agreement with our earlier results.<sup>10</sup>

Beyond the experimental determinations, there have been some theoretical attempts to calculate the dispersion relation, a recent example of which is the calculation by Aldrich.<sup>18</sup> His results show

that in the low-momentum expansion of the dispersion relation the parameter  $\gamma$  must be positive at saturated vapor pressure, and a term in  $p^4$  is present in Eq. (1). If this latter term does indeed exist (experimental evidence to date neither proves nor disproves its existence), the dispersion relation as proposed by Maris,<sup>14</sup> Eq. (9) would be incomplete, since it involves an expansion in terms of odd powers of momentum.

Additional experiments which provide in principle a direct measure of the phonon dispersion relation involve the use of heat pulses. An important quantity on which heat-pulse experiments can supply information is the cutoff energy  $\epsilon_c$ .

If the three-phonon decay process is the dominant contribution to the attenuation of high-frequency phonons, it is expected that phonons with energy  $\epsilon$  smaller than the cutoff value  $\epsilon_c$  will be highly damped, while phonons of  $\epsilon \geq \epsilon_c$  will propagate. Thus, a sharp increase in the number of phonons reaching the detector will occur for energy greater than  $\epsilon_c$ . Sluckin and Bowley<sup>19</sup> have shown that the sharpness of the attenuation cutoff is dependent on the form of the dispersion relation. Dynes and Narayanamurti (DN)<sup>20</sup> have measured this cutoff energy  $\epsilon_c$ , and for saturated vapor pressure they found the value  $9.5 \pm 0.5$  K.

By combining measurements of the cutoff energy with measurement of the group velocity at cutoff, it is possible to calculate the dispersion parameter  $\gamma$ , and in a relation of the form

$$\epsilon = Cp(1 + \gamma p^2 - \delta p^4) . \tag{10}$$

 $\mathrm{DN}^{20}$  found a value  $\gamma = 1.2 \times 10^{37}$  cgs, which is significantly below that obtained by other experimental techniques. The source of this discrepancy in  $\gamma$  is not clear at present; it could be the result of either an overestimation of  $\epsilon_c$  or a shortcoming of the particular form of the dispersion relations used.

As pressure is increased, the cutoff energy decreases until for some pressure it vanishes. Above this pressure, it is assumed that the dispersion parameter  $\gamma$  becomes negative and the three-phonon process is forbidden for all energy phonons. DN<sup>20</sup> have measured the cutoff energy as a function of pressure and observed that the characteristics of cutoff behavior are no longer observed for pressures in excess of about 16 atm. To obtain the pressure dependence of the phonon dispersion relation from our measurements we continue to use Eq. (9). By selecting values of  $\gamma$ it is then possible to determine corresponding values of  $p_A$  and  $p_B$  from the measurements of the cutoff energy by DN,<sup>21</sup> and of the phonon phase velocity, at q = 1 Å<sup>-1</sup>, obtained from inelastic neutron scattering.<sup>22</sup> These sets of values, of  $\gamma$ ,  $p_A$ ,  $p_B$ ,

are then used for computing  $\Delta C/C$  as a function of frequency. The curves thus obtained are compared with the experimental results, and the value of  $\gamma$  giving the best fit is selected as representing, for each pressure, the coefficient of the  $p^3$  term in the dispersion relation. Figures 10-13 show the computed and measured  $\Delta C/C$ . For these curves we take the resonance to occur in the frequency region where the velocity change is decreasing fastest with frequency. As  $\gamma$  is increased, the resonance shifts towards lower frequency. With this in mind we examine first the measurements at 8.4 atm and 0.256 K. Here we have calculated the dependence of the velocity change on frequency for  $\gamma = 3$ , 5, and  $10 \times 10^{37}$  cgs [Fig. 10(a)]. From the graph we observe that the experimental points lie between the curve for  $\gamma = 5$  and  $10 \times 10^{37}$  cgs, being somewhat closer to  $5 \times 10^{37}$  cgs. Subsequently [Fig. 10(b)], we calculated the velocity change as a function of frequency for 0.348 K, for  $\gamma = 3$ , 5, and  $7 \times 10^{37}$  cgs. Here the results again suggest



FIG. 10. Same as Fig. 9, for 8.4 atm. LFL and HFL represent the low- and high-frequency limits, respectively, for the calculated values. Generally LFL is independent of  $\gamma$ , whereas HFL varies with  $\gamma$  in the manner indicated.



FIG. 11. Same as Fig. 9, for 4 atm. LFL and HFL represent the low- and high-frequency limits, respectively, for the calculated values. Both LFL and HFL vary with  $\gamma$  in the manner indicated.

that  $\gamma$  is somewhat larger than  $5 \times 10^{37}$  cgs. These results indicate that  $\gamma$  lies in the range  $(5-7) \times 10^{37}$  cgs<sup>23</sup> for this pressure.

For 15 atm we performed calculations at 0.348 K and  $\gamma = 1$  and  $3 \times 10^{37}$  cgs (Fig. 13). Here we find  $\gamma$  to lie in the region  $(1.5-2.5) \times 10^{37}$  cgs units. For the other pressures involved in the experiments, selecting the calculated curve which reflects the most appropriate fit to the data becomes



FIG. 12. Same as Fig. 9, for 12 atm. LFL and HFL represent the low- and high-frequency limits, respectively, for the calculated values.



FIG. 13. Same as Fig. 9, for 15 atm; squares—present data, circles—data of Roach *et al.* (Ref. 5) obtained at 15.5 atm. LFL and HFL represent the low- and highfrequency limits, respectively, for the calculated values. LFL is independent of  $\gamma$ , whereas HFL varies with  $\gamma$  in the manner indicated.

more involved, since we do not have measurements at frequencies above 13 MHz. In general, we rely on the evolution of  $\Delta C/C$  with temperature to arrive at the appropriate range of values for  $\gamma$ .

To check the reliability of our estimates of the parameter  $\gamma$  we determined the effect of the terms  $p_{A}$  and  $p_{B}$  on the calculation of  $\Delta C/C$ . The pressure of 12 atm and temperature of 0.348 K was selected since the cutoff energy was well within the range of phonon energies considered in determining the velocity change. A shift in the cutoff energy would then change significantly the number of phonons which could contribute to the velocity change. We altered the values of  $p_A$  and  $p_B$  in such a manner as to increase the cutoff energy by 10%(this is the upper limit of the error bars in the measurements of DN<sup>21</sup>). The results of this calculation are included in Fig. 12 as the dashed line. We see that the qualitative shape of the curve and the location of the resonance are not affected by this change, while the calculated values of  $\Delta C/C$ at high and low frequencies are slightly reduced. Thus, our estimates of  $\gamma$  are not strongly dependent on  $p_A$  and  $p_B$  in the region covered in this test.

In Fig. 14, we show the results of our estimates of the values of the parameter  $\gamma$  for the densities (pressure) covered in the experiments. The pressure corresponding to the density at which  $\gamma$ reaches zero is approximately 18 atm. This pressure is in close agreement with that found by



FIG. 14. Dispersion parameter  $\gamma$  as a function of density difference  $\rho$ - $\rho_{SVP}$ , where  $\rho_{SVP}$  is the density at saturated vapor pressure.

Mills *et al.*<sup>24</sup> and DN.<sup>21</sup> These authors indicate that  $\gamma$  vanishes at 17 and 20 atm, respectively. This pressure region is somewhat higher than that originally suggested by the heat-capacity measurement of PWH<sup>15</sup> (8–10 atm), however, the latter measurements are sensitive to high-momentum phonons and may underestimate the value of  $\gamma$  and thus the pressure at which it goes through zero.

Recently, Svensson et al.<sup>25</sup> have measured the phonon dispersion relation by inelastic neutron scattering, at a series of pressures between SVP and the melting curve. To test the accuracy of the phonon dispersion relation used here in reproducing their measured dispersion curve, we compared the results of Eq. (9), with  $\gamma = 3 \times 10^{37}$ , to the neutron scattering measurements at 15 atm. The comparison shows (Fig. 15) that for wave vectors  $q < 0.5 \text{ Å}^{-1}$ , Eq. (9) provides a quite accurate representation of the dispersion curve, while for  $0.5 \le q \le 1$  Å<sup>-1</sup> this representation tends to overestimate the phonon phase velocity by only a few percent. Since the region of energies used in calculating  $\Delta C/C$  involves phonons with wave vector  $q \leq 0.3$  Å<sup>-1</sup>, the fact that the results of Eq. (9) deviate slightly from the dispersion curve for q > 0.5Å<sup>-1</sup> is not considered serious. We thus note the consistency of our approach with the inelasticneutron-scattering data. Our conclusion, however, differs from the analysis of Svensson et al.<sup>25</sup> of their measurements. They fit their experimental results to a dispersion relation given by Eq. (10). With the use of C as an adjustable parameter, taking on values up to 7% in excess of established results, they find that  $\gamma$  increases as the pressure increases. Clearly this is not consistent with our results. They suggest that the inability of their fits to reporduce the measured



FIG. 15. Comparison, for 15 atm, of phonon velocity (E/q) as a function of wave vector q, obtained from inelastic-neutron-scattering data (Ref. 25) (vertical bars), with the same quantity calculated from Eq. (9) by the procedure followed in the present study (solid line).

sound velocity C is an indication of the inability of Eq. (10) to describe fully the dispersion curve over a wide range of momenta; such is possibly the case.

<sup>†</sup>Research supported in part by the National Science Foundation through the Materials Research Laboratory at Brown University.

- <sup>1</sup>H. J. Maris and W. E. Massey, Phys. Rev. Lett. <u>25</u>, 220 (1970).
- <sup>2</sup>H. J. Maris, Phys. Rev. Lett. 28, 277 (1972).
- <sup>3</sup>W. M. Whitney and C. E. Chase, Phys. Rev. <u>158</u>, 200 (1967).
- <sup>4</sup>B. M. Abraham, Y. Eckstein, J. B. Ketterson, and M. Kuchnir, Phys. Rev. Lett. <u>19</u>, 690 (1967); B. M. Abraham, Y. Eckstein, J. B. Ketterson, M. Kuchnir, and J. Vignos, Phys. Rev. <u>181</u>, 347 (1969).
- <sup>5</sup>P. R. Roach, J. B. Ketterson, B. M. Abraham, and M. Kuchnir, J. Low Temp. Phys. 9, 105 (1972).
- <sup>6</sup>R. J. Blume, Rev. Sci. Instrum. <u>34</u>, 1400 (1963).

#### **IV. CONCLUSIONS**

We have measured the changes in sound velocity  $(\Delta C/C)$  of liquid <sup>4</sup>He in the frequency range 1-15 MHz, as a function of temperature with  $0.45 \ge T \ge 0.1$  K, and for pressures between saturated vapor pressure and 20 atm. Our measurements, coupled with those of Roach *et al.*,<sup>5</sup> show that  $\Delta C/C$  as a function of frequency displays a resonancelike structure, consistent with the assumption that for small-momentum phonons  $\gamma$  is positive in a dispersion relation of the form  $\epsilon = Cp(1 + \gamma p^2)$ .

In particular, we have compared our experimental results with the ones which we have calculated using the computer programs developed by Maris.<sup>7</sup> In these calculations, the low-momentum-phonon dispersion relation given by Eq. (9) has been used in determining the phonon interactions in the liquid. The calculated curves are compared to the measured values of  $\Delta C/C$  and the results that provide the best agreement with the data are selected. This yields a value of the dispersion parameter  $\gamma$ . We have thus found that the dispersion parameter  $\gamma$  is positive at saturated vapor pressure and has a value between  $15 \times 10^{37}$  and  $17 \times 10^{37}$  cgs. As pressure is increased,  $\gamma$  decreases linearly with increasing density, going to zero at a density corresponding to approximately 18 atm.

It is concluded, therefore, that the parameter  $\gamma$  is indeed positive up to a pressure of approximately 18 atm. As a direct consequence of this finding it is further concluded that for low-momentum phonons, three-phonon processes which explicitly conserve energy and momentum are permitted in liquid <sup>4</sup>He and that transport phenomena such as ultrasonic velocity reflect this property. Finally, it is noted that the phonon dispersion relation used here is consistent with the recently obtained results of inelastic neutron scattering at high pressure.<sup>25</sup>

<sup>7</sup>H. J. Maris, Phys. Rev. A 8, 2629 (1973).

<sup>8</sup>We note that the quantitative agreement between our measurements and those of Roach *et al.* (Ref. 5) is good. Any discrepancies could be the result of several factors. The correction that we have applied to our data, particularly the Q correction, could be in error by a few percent, and differences in the temperature scales used in the two experiments could amount to a few percent: this is perhaps the most likely possibility. A discrepancy of approximately 2% between the temperature scales would result (assuming a  $T^4$  dependence for  $\Delta C/C$ ) in a disagreement of 8% in the measured velocity change.

<sup>&</sup>lt;sup>9</sup>Y. Disatnik, Phys. Rev. <u>158</u>, 162 (1967); I. M. Kahalatnikov and D. M. Chernikova, Zh. Eksp. Teor. Fiz.

Pis'ma Red. 2, 566 (1965) [JETP Lett. 2, 351 (1965)]; I. M. Khalatnikov and D. M. Chernikova, Zh. Eksp.

- Teor. Fiz. 49, 1957 (1965) [Sov. Phys.-JETP 22, 1336 (1966)]; C. J. Pethick and D. Ter Haar, Physica <u>32</u>, 1905 (1966).
- <sup>10</sup>W. R. Junker and C. Elbaum, Phys. Rev. Lett. <u>34</u>, 186 (1974).
- <sup>11</sup>P. F. Meier, H. Beck, and R. Beck, Phys. Rev. B <u>12</u>, 3745 (1975).
- <sup>12</sup>R. K. Wehner, Phys. Rev. A 9, 2625 (1974).
- <sup>13</sup>P. F. Meier and H. Beck, Phys. Rev. A 8, 569 (1973).
   <sup>14</sup>H. J. Maris, Phys. Rev. A 9, 1412 (1974); D. Benin, Phys. Rev. B 13, 1105 (1976).
- <sup>15</sup>N. E. Phillips, C. G. Waterfield, and J. K. Hoffer, Phys. Rev. Lett. 25, 1260 (1970).
- <sup>16</sup>J. S. Brooks and  $\overline{R}$ . J. Donnelly, Phys. Lett. A <u>46</u>, 111 (1973).
- <sup>17</sup>J. E. Berthold, H. N. Hanson, H. J. Maris, and G. M. Seidel, Phys. Rev. B (to be published).
- <sup>18</sup>C. H. Aldrich, thesis (University of Illinois, Urbana,

1974) (unpublished).

- <sup>19</sup>T. J. Sluckin and R. M. Bowley, Proc. Phys. Soc. Lond. 7, 1779 (1974).
- $^{20}\overline{R}.$  C. Dynes and V. Narayanamurti, Phys. Rev. Lett. 33, 1195 (1974).
- <sup>21</sup><del>R.</del> C. Dynes and V. Narayanamurti, Phys. Rev. B <u>12</u>, 1720 (1975).
- <sup>22</sup>E. C. Svensson, A. O. B. Woods, and P. Martel, Phys. Rev. Lett. 29, 1148 (1972).
- <sup>23</sup>We note that at frequencies below 10 MHz the calculated curve tends to overestimate the velocity change. This discrepancy is attributed to the observed fact that the computations converge slowly at low frequencies; in order to obtain results of comparable accuracy at all frequencies covered, prohibitively long computation times are required.
- <sup>24</sup>N. G. Mills, R. A. Sherlock, and A. F. G. Wyatt, Phys. Rev. Lett. 32, 978 (1974).
- <sup>25</sup>C. E. Svensson, P. Martel, and A. D. B. Woods, Phys. Lett. A 55, 151 (1975).