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Investigation of the phonon dispersion relation in liquid ^{4}He by thermal-expansion measurements*

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The thermal expansion of superfluid ^{4}He under the saturated vapor pressure has been measured in the temperature range 0.1–0.62 K. The expansion was found by filling a superconducting resonant cavity with the liquid and measuring the resonant frequency as a function of temperature. The density was related to the dielectric constant through the Clausius-Mossotti relation. Sensitivity to changes in fractional density of 4×10^{-9} was achieved. The thermal-expansion data were used to obtain quantitative information about the dispersion relation for thermal phonons. In addition to the thermal-expansion measurements, tests of the validity of the Clausius-Mossotti relation were made.

I. INTRODUCTION

In the last few years there has been much discussion and controversy concerning the dispersion relation for phonons in liquid ^{4}He. The interest in this subject occurs because transport processes involving phonons depend in a very sensitive way upon the deviations of the phonon dispersion relation from linearity.[1] It was originally assumed that for small phonon momentum p the energy ϵ was given by a power series of the form[2]

$$\epsilon = c_0 p(1+\alpha_2 p^2/\hbar^2 + \alpha_4 p^4/\hbar^4 + \cdots), \tag{1}$$

where c_0 is the phonon velocity in the limit of very small momentum, and is equal to the velocity of sound. The parameter α_2 was believed to be negative but it is now known from the analysis of several experiments[3-8] that α_2 must actually be positive. This results in a complicated dependence of the phonon phase velocity

$$c(p) \equiv \epsilon/p \tag{2}$$

on momentum. For very small momentum $c(p)$ is equal to c_0. The velocity increases with increasing p and reaches a maximum value (probably at around 0.3 Å^{-1}) and then decreases rapidly for larger p. The increase in the velocity for small momentum is referred to as anomalous dispersion.

Although the qualitative features of the dispersion are well established, quantitative information is still lacking. In this paper we attempt to obtain information about the dispersion relation by studying the thermal expansion of the liquid in the temperature range 0.1–0.6 K. The basic idea behind this work is as follows. If the dispersion curve were exactly linear, the thermal expansion would vary with temperature exactly as T^3. Deviations of the dispersion relation from linearity give rise to deviations of the thermal expansion from T^3 behavior. Hence, at least in principle, a measurement of the thermal expansion can be used to study the phonon dispersion. In practice the main problem is that very accurate measurements of the thermal expansion are required. Measurements have to be made below about 0.6 K so that the roton contribution to the thermal expansion remains small. At 0.6 K the magnitude of the T^3 term in the thermal expansion is only 2×10^{-4} K^{-1}. To study the deviations from T^3 behavior[9] one therefore needs to be able to measure a thermal-expansion coefficient of this magnitude to an accuracy of at least 1%. In our experiments we have measured the thermal expansion by filling a niobium superconducting microwave cavity with liquid helium. As the temperature is increased, the liquid expands and flows out of the cavity. The dielectric constant of the liquid in the cavity is reduced and this causes a shift in the resonant frequency of the cavity. By measuring the frequency shift we have been able to detect fractional changes in density of 4×10^{-9}. In addition, we have been able to use the same cavity to measure the density of ^{3}He vapor in equilibrium with bulk liquid. In this way it is possible to use the cavity as a very accurate thermometer in the temperature range 0.25–0.6 K.

II. EXPERIMENTAL METHOD

A. Microwave cavity

The cavity used in these experiments was a cylindrical cavity fabricated from electron-beam-melted high-purity niobium. It was operated in the TE$_{011}$ mode, and resonated at 9.467 GHz when empty. As shown schematically in Fig. 1, the cavity consisted of three separate pieces (two end plates and a tube). These were held together by screws. In the TE$_{011}$ mode of oscillation there is no current flow across the joints between the tube

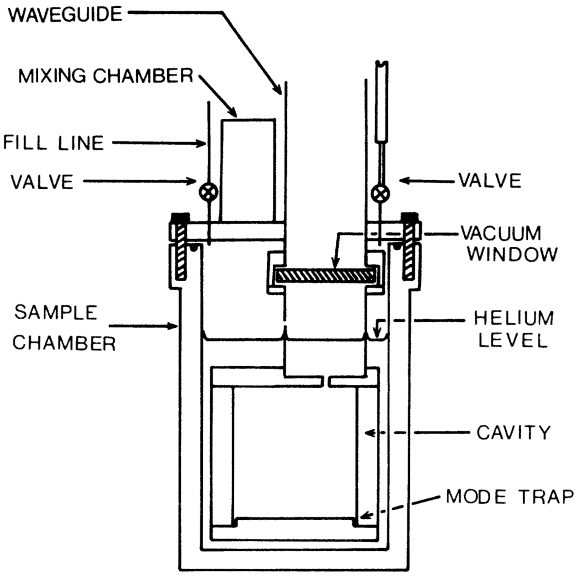

FIG. 1. Schematic diagram of the sample chamber and cavity.

and the end plates. This is a considerable advantage since it is not necessary to have good electrical contact at these joints in order to have a high Q. The TE_{011} mode also has the property that the electric field (both normal and tangential components) vanishes at all points on the cavity walls. Thus there is no dielectric loss or shift in resonant frequency due to a thin layer of adsorbed gas or dirt on the surfaces. For a perfect cylinder the TE_{011} mode is degenerate with the TM_{111} mode, and thus for a cylinder in which the geometry is nearly perfect one expects considerable coupling between the two modes. To reduce this coupling the geometry was intentionally altered by cutting a "mode-trap" groove as shown in Fig. 1.

To obtain a high Q it is essential that the niobium surface be smooth and undamaged. The cavity pieces were machined on a lathe using Freon TF as a coolant,[10] and were then mechanically polished with silicon carbide of varying degrees of coarseness, ending with No. 600 grit. Next, the surfaces were electropolished by an oscillating electrolysis method similar to that developed at Siemens.[11] Finally, the niobium was anodized[11] in a (10–15)%-volume NH_3 solution for about 20 min. The current density was roughly 3 mA/cm^2. The effect of anodizing is to produce a layer of Nb_2O_5 on the surface. It is believed that this displaces the cavity surface from the position of the original niobium surface to a deeper level where the niobium is less likely to be contaminated. The highest unloaded Q that was obtained in these experiments was 2.7×10^8, and this was essentially independent of temperature below 1 K. This Q is considerably lower than the highest previously reported Q for niobium cavities,[12] but as will be discussed later is quite adequate for the present experiments.

The cavity was coupled to the waveguide by an iris of diameter 0.345 cm in a wall of thickness 0.254 cm. This gave a loaded Q of 1.7×10^8 at low temperatures and a coupling constant β of 0.6.

B. Cryogenics

The sample chamber (Fig. 1) consisted of a copper top plate attached to the mixing chamber of a dilution refrigerator, and a thick-walled brass can. The cavity was supported by means of a waveguide section (including a vacuum window) which extended through the top plate. The can was sealed to the top plate by a screw-clamped indium O-ring seal that remained super-leak-tight up to pressures of at least 25 atm. At the upper end of the waveguide section was a waveguide to coaxial convertor. Coaxial cable was used from this point up the cryostat to room temperature.

The effect of a hydrostatic pressure head is quite serious in accurate thermal-expansion measurements. As discussed in Sec. III, we were able to measure the cavity resonant frequency with an accuracy such that fractional changes in liquid-helium density of 4×10^{-9} could be resolved. A density change of this magnitude can also be induced by a hydrostatic pressure of 0.25 dyn/cm^2, which corresponds to a column height of helium of only 18 μm. Thus it was essential to keep a fairly constant helium level in the sample chamber during the thermal-expansion measurement. This was achieved by having low-temperature valves on the fill and vent lines (see Fig. 1) and closing them during the experiment. The valves were similar to those described by Roach *et al.*,[13] but used a mechanical instead of a hydraulic drive. With the valves closed the helium level still changes by a small amount due to evaporation of the liquid as the temperature is increased. In addition, the density of the liquid changes slightly because of compression by the vapor pressure. However, below 0.6 K both of these effects give density changes which are at least three orders of magnitude less than those due to thermal expansion.

The ^{4}He liquid used in these experiments was condensed from gas from natural gas wells, and had not been especially purified to reduce the ^{3}He content. Other impurities were condensed out by passing the gas through two traps at 77 K, one of which was filled with molecular sieve material.

C. Electronics

A simplified block diagram of the electronics that was used to measure the cavity resonance is shown in Fig. 2. The problem of generating very stable frequency signals at X band was handled as follows. A rubidium frequency standard[14] produced a 5-MHz signal with short-term stability better than 5×10^{-12} rms averaged over 1 sec. A coherent microwave synchronizer[15] was then used to lock an X-band klystron to a high harmonic of the frequency standard. In this way it was possible to lock the klystron at discrete frequencies 10 MHz apart in the X-band region. To obtain a stable frequency near the resonant frequency ω_c of the cavity the klystron output (frequency ω_k) was mixed with an intermediate frequency ($\omega_{\text{i.f.}}$) from a continuously tunable frequency synthesizer[16] using a single-sideband modulator.[17] The i.f. signal was in the range 35–70 MHz.

For convenience in taking data it was decided to lock electronically the frequency $\omega_0(\equiv\omega_k - \omega_{\text{i.f.}})$ to the center of the cavity resonance. The method used is similar to that developed by Pound and modified by Trela and Fairbank.[18] Stein and Turneaure[19] have locked Gunn oscillators to superconducting cavities in this way. In our scheme the synthesizer frequency $\omega_{\text{i.f.}}$ was frequency modulated at frequency ω_m (about 4.7 kHz) with a modulation excursion of 2 kHz. The modulating voltage was taken from the reference output of a lock-in amplifier. The output from the single-sideband modulator then becomes a carrier at frequency ω_0 and two principal side bands at $\omega_0 \pm \omega_m$. The entire signal is fed to the cavity through a circulator as shown in Fig. 2. When ω_0 is within the bandwidth of the resonant cavity the carrier is phase shifted upon reflection by an amount which depends upon $\omega_c - \omega_0$. The sidebands have frequencies outside the range of the cavity resonance, and so the phase of the reflected sidebands does not depend in a sensitive way upon $\omega_c - \omega_0$. The returning signal can thus be approximated by

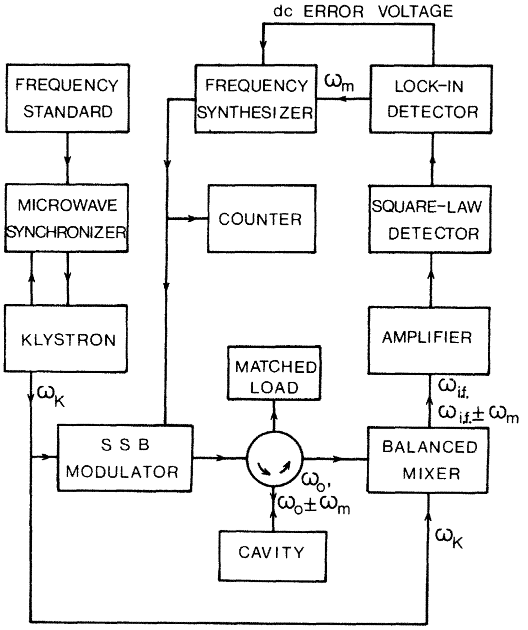

FIG. 2. Block diagram of the electronics.

$$V_r = V_0 \Gamma e^{i(\omega_0 t+\psi)} + V_1 e^{i(\omega_0+\omega_m)t} - V_1 e^{i(\omega_0-\omega_m)t}, \tag{3}$$

where Γ and ψ are the magnitude of the reflection coefficient and the phase shift for the carrier, respectively, both of which depend on $\omega_c - \omega_0$. The signal given by Eq. (3) is then fed into one input of a balanced mixer. A suitable amount of the klystron output of amplitude V_k and frequency ω_k is fed into the other arm, i.e., a voltage

$$V' = V_k e^{i(\omega_k t+\phi)}, \tag{4}$$

where ϕ is a possible phase shift. The output of the mixer is the product of these inputs and is passed through an amplifier which has a bandwidth greater than $2\omega_m$. The output of this amplifier is square-law detected, and then fed into the lock-in amplifier from which the modulating voltage at frequency ω_m is taken. It can be shown using Eqs. (3) and (4) that the signal detected by the lock-in is proportional to $\Gamma \sin\psi$. The detected dc output of the lock-in is then used to electronically tune the frequency synthesizer, and thus to shift ω_0. For small $\omega_c - \omega_0$, Γ is constant and

$$\psi \propto \omega_c - \omega_0 .$$

Thus by an appropriate choice of the amplification levels in the i.f. amplifier and the lock-in it is possible to lock the frequency ω_0 to the center of the cavity resonance. Small changes of ω_c can then be measured by monitoring the output frequency $\omega_{\text{i.f.}}$ of the synthesizer with a counter. We have used a counter whose time base is controlled by the rubidium standard (see Fig. 2).

The stability of the locking system was such that under normal conditions with the cavity at constant temperature the frequency was constant to ±1 Hz when measured with a 10-sec counting time. There was some evidence that the stability may have been limited by mechanical vibrations. For example, at night when the laboratory was very quiet the fluctuations were reduced to ±0.1 Hz.

In addition to these fairly short term fluctuations, a drift in the resonant frequency of about 1 Hz/h was observed. This drift was definitely correlated with the level of liquid nitrogen in the outer Dewar.

One effect of the nitrogen level is to change the electrical length of the coaxial cable[20] from the circulator to the waveguide section above the cavity. However, this adds the *same* phase shift to each of the three components in the returning signal from the cavity [see Eq. (3)] and it is easy to show that this leads to no change in the correcting voltage coming out of the lock-in. Hence a simple change in the length of the cryostat cannot explain the drift that was observed. In fact this insensitivity to electrical length is a major advantage of the Pound-Trela-Fairbank locking scheme. However, in a more realistic analysis of the experiment one should include in V_r contributions from other reflected waves in addition to those reflected at the cavity. Some of these waves (e.g., those coming from reflections at the coaxial connector at the top of the cryostat) will not have passed along the section of coaxial line in which the temperature variation due to the nitrogen level occurs. It can be shown[21] that interference between the waves which suffer no temperature-dependent phase shift and the three waves included in Eq. (3) can give rise to a spurious correcting voltage from the lock-in, and hence to a frequency shift depending on the nitrogen level. We believe that it is this effect which produces the observed drift.

D. Thermometry

The temperature was measured by a germanium resistance thermometer. The resistance was monitored using an ac bridge. A calibration of the thermometer was performed using the superconducting cavity to measure the the density of ^{3}He vapor in equilibrium with a small amount of bulk liquid condensed in the sample chamber below the resonant cavity. The dielectric constant ϵ of a low-density gas is given by

$$\epsilon - 1 = 4\pi\alpha/V_m\,, \qquad (5)$$

where α is the molar polarizability and V_m is the molar volume. Thus the fractional shift in frequency of the cavity when ^{3}He gas with molar volume V_m is introduced should be

$$\Delta\nu/\nu = -\,2\pi\alpha/V_m\,. \qquad (6)$$

For a nearly ideal gas the molar volume can be related to the pressure through the virial expansion

$$PV_m/RT = 1 + B(T)/V_m + \cdots\,. \qquad (7)$$

Thus

$$\Delta\nu/\nu = -\,(2\pi\alpha P/RT)\,[1 - B(T)P/RT + \cdots]\,. \qquad (8)$$

The molar polarizability α has been measured with good accuracy by Kerr and Sherman.[22] Inserting their value gives

$$\Delta\nu = -1.177\times10^5\frac{P}{T}\left(1 - \frac{B(T)P}{RT} + \cdots\right)\ \mathrm{Hz}\,, \qquad (9)$$

where P is in Torr and T is in K. The ^{3}He temperature scale provides a relation between the saturated vapor pressure of ^{3}He and the absolute temperature. Thus, if the virial coefficients are known with sufficient accuracy a measurement of the cavity frequency shift when it contains saturated ^{3}He vapor can be used to determine the temperature.

Figure 3 shows the expected $\Delta\nu$ as a function of T on the basis of the ideal-gas term alone, together with the temperature dependence of the uncertainty in $\Delta T/T$ arising from a ±1-Hz uncertainty in a measurement of $\Delta\nu$. The uncertainty in ΔT decreases very rapidly as the temperature increases. However, above 0.6 K the virial correction becomes significant. The coefficient $B(T)$ has been measured by Grimsrud and Werntz[23] between 1.2 and 3.6 K and by Keller[24] between 1.5 and 3.0 K. We have used Keller's data as reanalyzed by Roberts, Sherman, and Sydoriak,[25] and have extrapolated it to our temperature range. The value of B is

$$B(T) = 5 - (271/T)\ \mathrm{cm^3/mole}\,. \qquad (10)$$

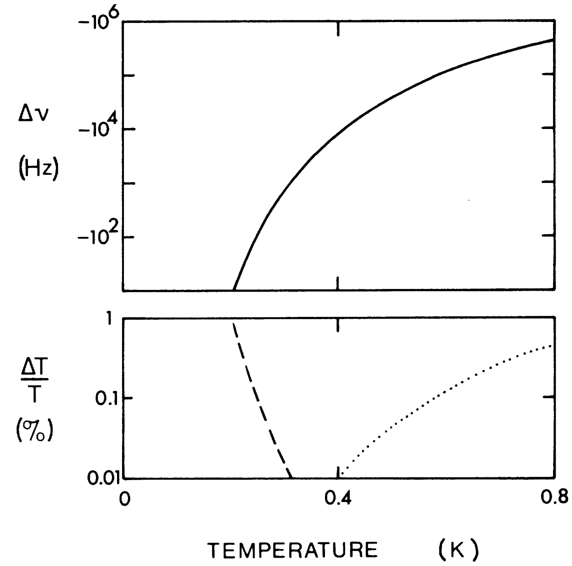

FIG. 3. Behavior of the cavity as a vapor-density thermometer. Solid curve show expected frequency shift when the sample chamber contains saturated ^{3}He vapor. The vapor is assumed to behave as an ideal gas. Dashed curve shows percent uncertainty in temperature resulting from a ±1-Hz uncertainty in the cavity resonant frequency. Dotted curve shows correction to temperature from the second virial coefficient.

In our temperature determination this leads to a correction of the magnitude shown in Fig. 3. As a check on the validity of the extrapolation of the virial coefficients we performed simultaneous measurements of $\Delta\nu$ and vapor pressure P (measured at room temperature) at temperatures in the range 0.6–1.0 K. Corrections for the thermomolecular pressure effect were applied using the data of Roberts and Sydoriak.[26] In this temperature range the discrepancy between the temperature obtained through the use of $\Delta\nu$ [i.e., through Eq. (9)] and through the vapor pressure was no more than 0.3%.

A correction was also applied to allow for the 0.8% ^{4}He contamination of the ^{3}He gas used.[25] This correction was 0.15% at 1 K, but only 0.04% at 0.4 K. The final accuracy of the calibration in the temperature range 0.25–0.6 K was in fact limited mainly by the sensitivity of the germanium resistance thermometer and by the curve-fitting procedures. We estimate the accuracy as ±0.2%. In addition, there is an uncertainty associated with the ^{3}He vapor-pressure scale itself.[27]

Below 0.25 K the accuracy becomes increasingly limited by the ±1-Hz resolution in $\Delta\nu$ (see Fig. 3). In this range of temperature the germanium thermometer was calibrated against cerium magnesium nitrate (CMN). The uncertainty was no worse than ±1% down to 0.1 K.

III. RESULTS AND DISCUSSION

A. Dielectric constant measurements

The dielectric constant ϵ_0 of liquid helium at zero temperature ($T<0.1$ K) was measured by finding the resonant frequency of the cavity empty and filled with liquid. Then

$$\epsilon_0 = (\nu_{\text{empty}}/\nu_{\text{full}})^2 . \tag{11}$$

Five separate measurements were made and the result was

$$\epsilon_0 = 1.057\,278\,4 \pm 0.000\,000\,5 . \tag{12}$$

The accuracy is limited by two effects. Opening or closing the low-temperature valves causes a mechanical shock which can shift the resonant frequency of the cavity by as much as 1 kHz. The second problem is that it was necessary to fill the sample chamber until the cavity was completely immersed. The height of the liquid above the cavity was not known, and thus the hydrostatic pressure inside the cavity varied from run to run and was somewhat uncertain.

The dielectric constant has been measured previously by Harris-Lowe and Smee[28] at temperatures of 1.072 and 1.018 K. If their results are corrected to zero temperature[29] one finds values of 1.057 258±0.000 020 and 1.057 242±0.000 010. These results are thus in reasonable agreement with our value.

The dielectric constant was also measured at the freezing pressure. A complication that occurs is the compression of the cavity. This changes all linear dimensions by a fraction

$$\tfrac{1}{3}P\kappa_{\text{Nb}} ,$$

where P is the pressure and κ_{Nb} is the compressibility of niobium. The compressibility of polycrystalline niobium depends to some extent on metallurgical details,[30] but is probably in the range $(6\pm0.2)\times10^{-13}$ cm^2/dyn. Thus a pressure of 25 atm causes an increase in the resonant frequency of the cavity by 5.1±0.2 ppm, which must be subtracted before calculating the dielectric constant. In addition to this problem it was found that after pressurizing to 25 atm and returning to zero pressure the resonant frequency was reduced by 20 kHz from its original value. This was presumably caused by some form of mechanical shock. If a ±10-kHz uncertainty is included in the frequency shift, the result for the dielectric constant at freezing is

$$\epsilon_f = 1.068\,385 \pm 0.000\,002 . \tag{13}$$

The measurement of the thermal expansion relies on the use of the Clausius-Mossotti (CM) relation

$$(\epsilon - 1)/(\epsilon + 2) = \tfrac{4}{3}\pi(\alpha\rho/M) , \tag{14}$$

where α is the molar polarizability and M is the molecular weight. We can check the CM relation as follows. Kerr and Taylor[31] have measured the molar volume at zero pressure. Their results give a density at 1.2 K of 0.145 094 g/cm^3 with an absolute accuracy of ±0.02%. This gives a value for the density ρ_0 at $T=0$ K of 0.145 119 g/cm^3. Combining this with our result (12) for the dielectric constant leads to a molar polarizability

$$\alpha = 0.123\,363 \pm 0.000\,025 \text{ cm}^3/\text{mole} . \tag{15}$$

This can now be used with ϵ_f from (13) to calculate the density at freezing via (14). The result is

$$\rho_f = 0.172\,61 \pm 0.000\,03 \text{ g/cm}^3 . \tag{16}$$

This result lies between the values 0.172 93 and 0.172 46 g-cm^3 obtained by Grilly[32] and by Abraham *et al.*,[33] respectively, using methods which do not rely on the CM relation.

In our experiment it is also important that there are no temperature-dependent corrections to the Claussius-Mossotti relation. To look for these we filled the sample chamber with liquid under pressure and closed both of the low-temperature valves. It was then possible to vary the temperature while keeping the volume and density of the liquid nearly constant. The results are shown in Fig. 4 for 0.4 and ~25 atm. It is possible to explain most of the observed shift in terms of the expansion of the sample chamber caused by the increase of the helium pressure as the temperature is raised. A calculation of the elasticity of the sample chamber gave for the fractional change in volume caused by an internal pressure ΔP

$$\Delta V/V = 2.8\times 10^{-11}\Delta P\,, \tag{17}$$

where ΔP is in dyn/cm^2. This result may be uncertain by as much as ±50%, because of the complicated geometry of the can. Using (17) we can calculate the change in helium density as a function of temperature, and can then find the expected frequency shift from the CM relation. The result is the dashed curves in Fig. 4. The shifts have a similar temperature dependence to the experimental result but are smaller by ~30% at 0.4 atm and nearly 50% at 25 atm. If it is assumed that the volume change of the can is actually

$$\Delta V/V = 4.2\times 10^{-11}\Delta P\,, \tag{18}$$

the 0.4-atm data are fit well and the 25-atm data all lie within 1 Hz of the expected shift (solid line in Fig. 4). Thus our data provide no *positive* indication of a temperature-dependent correction to the CM relation and show that any effect must give a contribution to the dielectric constant which is less than 10^{-9} for $T<0.45$ K.

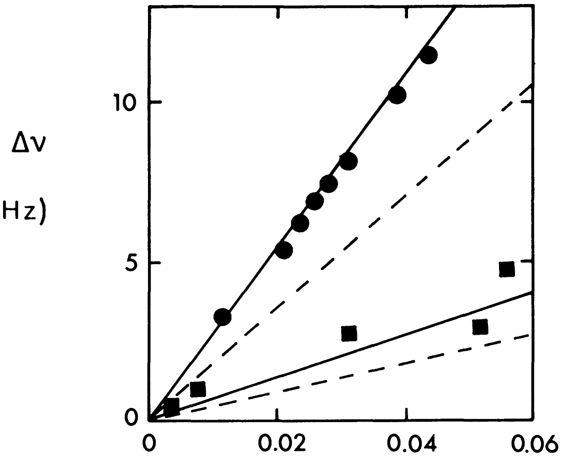

FIG. 4. Results of a search for temperature-dependent corrections to the Clausius-Mossotti relation. Solid circles and solid squares are data at pressures of 0.4 and ~25 atm. The dashed and the solid lines are calculations of the frequency shifts expected due to the extension of the sample chamber caused by the increase in the helium pressure with increasing temperature (see text for details).

B. Thermal-expansion measurements

The liquid density was measured in the temperature range 0.091–0.620 K. At 0.091 K (the lowest temperature attainable) there was still a measurable thermal expansion, and so it was necessary to make an extrapolation to find the density ρ_0 at $T=0$. In doing this it was adequate to assume that the thermal expansion α was proportional to T^3. We estimate that the uncertainty in ρ_0 due to the extrapolation is no more than 5 parts in 10^9. Results for $(\rho_0-\rho_T)/\rho_0 T^4$ are shown in Fig. 5. The error bars represent the combined effect of uncertainties in ρ_T $(\pm 4\times 10^{-9})$, in ρ_0, and in the temperature (see Sec. II). The data at the lowest temperatures, which are primarily of value in determining ρ_0, have not been included in Fig. 4. In the temperature range above 0.3 K our results are in reasonable agreement with the unpublished data of Van Degrift.[34]

1. Roton contribution

At temperatures close to 0.6 K there is a significant contribution to the thermal expansion α_r from rotons. We have estimated this using the expression for α_r given by Atkins and Edwards.[35] Roton parameters were taken from Deitrich *et al.*[36] The density change due to rotons is only 1.2% of the total density change at 0.62 K and is completely negligible below 0.55 K.

2. Analysis of the low-temperature data

In the limit of low temperature only very-low-energy phonons are excited in the liquid. For these phonons one can assume linear dispersion

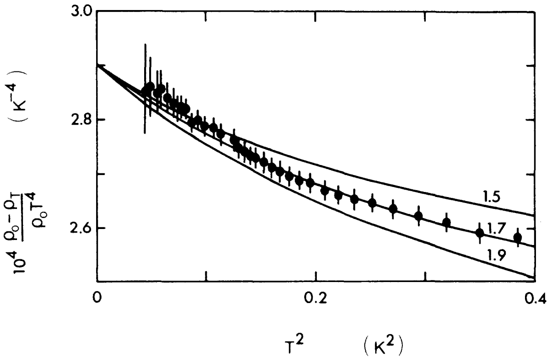

FIG. 5. Experimental results for the thermal expansion compared with calculated values based upon dispersion curves with $\alpha_2 = 1.5$, 1.7, and 1.9 Å^2. The remaining parameters of the dispersion curves are given in the text.

$$\epsilon = c_0 p . \tag{19}$$

Then

$$(\rho_0 - \rho_T)/\rho_0 = A_4 T^4 , \tag{20}$$

with

$$A_4 = (2\pi^2 k_B^4 / 15\hbar^3 \rho_0 c_0^5)(u_0 + \tfrac{1}{3}) , \tag{21}$$

where u_0 is the Grüneisen constant defined as

$$u_0 \equiv \frac{\partial \ln c_0}{\partial \ln \rho_0} . \tag{22}$$

Using the values[37] $c_0 = (2.383 \pm 0.001) \times 10^4$ cm/sec and[38] $u_0 = 2.84 \pm 0.01$ we find

$$A_4 = (2.90 \pm 0.01) \times 10^{-4}\ \mathrm{K}^{-4} . \tag{23}$$

The data give a slightly higher value of A_4 as can be seen from Fig. 5. A weighted least-squares fit of the form

$$(\rho_0 - \rho_T)/\rho_0 = A_4 T^4 + A_6 T^6 + A_8 T^8 \tag{24}$$

using all of the data gave $A_4 = 2.932 \times 10^{-4}\ \mathrm{K}^{-4}$. A similar fit but with a T^7 term instead of T^8 gave $A_4 = 2.955 \times 10^{-4}\ \mathrm{K}^{-4}$. Possible origins of the discrepancy are (i) the uncertainty in the data, (ii) the choice of polynomials like Eq. (24) is inappropriate, (iii) the uncertainty in u_0 is slightly larger than stated above, or (iv) our temperature calibration is too low by approximately 0.2%. We mention possibility (iv) because there have been previous proposals[27] that the ^{3}He scale itself may be too low by about 0.2%.

3. Relation of density change to phonon dispersion

In the case of a general dispersion relation $\epsilon(p)$ it can be shown that

$$\frac{\rho_0 - \rho_T}{\rho_0} = \frac{4\pi}{\rho_0 c_0^2 h^3} \int \left(\frac{\partial \ln \epsilon}{\partial \ln \rho_0} + \frac{1}{3} \frac{\partial \ln \epsilon}{\partial \ln p} \right) \times \epsilon n(\epsilon) p^2 dp . \tag{25}$$

For fairly small momentum we can write ϵ as

$$\epsilon = c_0 p (1 + \alpha_1 p/\hbar + \alpha_2 p^2/\hbar^2 + \cdots) . \tag{26}$$

Then for a given set of parameters $\{\alpha_i\}$ and $\{\partial \alpha_i / \partial \rho_0\}$ the thermal expansion can be calculated numerically and compared with the data. As a means of determining the $\{\alpha_i\}$ this procedure is limited because of the large number of variables involved. Fortunately other experiments and theory provide the following constraints:

(i) It is known from ultrasonic measurements[39] that $\alpha_1 = 0$, and this is presumably true regardless of density. Thus $\partial \alpha_1 / \partial \rho_0 = 0$.

(ii) Kemoklidze and Pitaevskii and Feenberg[40] have independently derived the following result for α_3:

$$\alpha_3 = -\pi^2 U_0 / 24 \rho_0 c_0^2 , \tag{27}$$

where U_0 is related to the long-range part of the van der Waals potential $V(R)$ between two helium atoms by

$$V(R) = -U_0 (R_0/R)^6 . \tag{28}$$

R_0 is the mean spacing of atoms in the liquid and is defined by $\rho_0 R_0^3 = M$ (M is the mass of helium atom). Using the value of U_0 calculated by Davison[41] one obtains

$$\alpha_3 = -3.34\ \text{Å}^3 , \tag{29}$$

$$\frac{\partial \ln \alpha_3}{\partial \ln \rho_0} = -4.68 . \tag{30}$$

(iii) There is much evidence from thermodynamic data and from transport measurements[3-8] that α_2 is negative. It is also known that when the liquid is compressed α_2 decreases and changes sign around 20 atm.[42] This imposes some constraints on "reasonable" values of $\partial \ln \alpha_2 / \partial \ln \rho_0$. For example, if it is assumed that α_2 varies linearly with ρ_0,

$$\frac{\partial \ln \alpha_2}{\partial \ln \rho_0} = -6.2 . \tag{31}$$

On the other hand, if it is assumed that α_2 varies linearly with P,

$$\frac{\partial \ln \alpha_2}{\partial \ln \rho_0} = -4.1 . \tag{32}$$

Using these results we are left with α_2, and the higher coefficients α_4, α_5, etc., and their density derivatives as free parameters. For u_0, and c_0 we have assumed the values measured in Refs. 37 and 38. In Fig. 5 we show the density change calculated from Eq. (25) with $\alpha_2 = 1.5$, 1.7, and 1.9 Å^2, and α_4 and all higher coefficients zero. $\partial \ln \alpha_2 / \partial \ln \rho_0$ has been set equal to -5. The best fit to the experimental results is with $\alpha_2 = 1.7$, but $\alpha_2 = 1.6$ or 1.8 Å^2 are only marginally inconsistent with the data. If $\partial \ln \alpha_2 / \partial \ln \rho_0$ is changed from -5 to -8 the best fit is obtained with $\alpha_2 = 1.5$ Å^2. If a nonzero value of α_4 is assumed it is still possible to obtain a reasonable fit, provided that α_4 is not too large. Reasonable fits are obtained for combinations from $\alpha_2 = 1.4$ Å^2 and $\alpha_4 = 6$ Å^4 to $\alpha_2 = 1.9$ Å^2 and $\alpha_4 = -6$ Å^4. These are shown in Fig. 6. Similarly it is also possible to obtain agreement for a range of values of $\partial \ln \alpha_4 / \partial \ln \rho_0$ if small adjustments are made in α_2.

We may summarize the results of the attempts at fitting the data as follows. There exist uncertainties in α_2 of ± 0.1, ± 0.2, and ± 0.25 Å^2 arising from scatter in the data, uncertainty in $\partial \ln \alpha_2 / \partial \ln \rho_0$, and uncertainty in α_4, respectively. By combining these errors we obtain a final value for

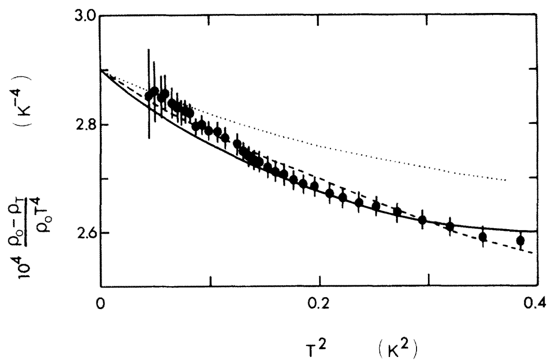

FIG. 6. Experimental results for the thermal expansion compared with calculated values. Solid curve: $\alpha_2=1.9$ Å^2, $\alpha_4=-6$ Å^4. Dashed curve: $\alpha_2=1.5$ Å^2, $\alpha_4=6$ Å^4. Dotted curve is calculated from the theory of Aldrich (Ref. 43).

α_2 of 1.7 ± 0.34 Å^2.

It is very difficult to compare this value of α_2 with earlier determinations because different workers have used different parametrizations of the dispersion curve. In particular, the Kemoklidze-Pitaevskii-Feenberg term has always been omitted. We feel that it is important to include this in the analysis because it is the only term in the power series (26) whose coefficient can be evaluated with confidence from first principles. Dynes and Narayanamurti[8] have used tunnel junctions to determine the energy ϵ_c above which phonons are stable against unharmonic decay. They have also measured the group velocity v_c of these phonons. To relate their results to phonon dispersion they assumed that the dispersion relation was of the form of Eq. (1), and that corrections beyond $\alpha_4 p^4/\hbar^4$ were negligible. At the saturated vapor pressure they obtained $\alpha_2=0.12$ Å^2 and $\alpha_4=0.36$ Å^4. Their value of α_2 is thus an order of magnitude less than our result, and is definitely not consistent with our experimental results. However, the reason for this discrepancy is almost certainly that the two experiments measure different parts of the dispersion curve. Our experiment is mostly sensitive to phonons of energy below 2 or 3 K, and we believe that in this energy range it is not too bad an approximation to use a power-series approximation to the dispersion relation. However the ultrasonic measurements of Dynes and Narayanamurti determine properties of the phonon spectrum at much higher energy, approximately 10 K, corresponding to a momentum of about 0.5 Å^{-1}. It is very unlikely that a power series of the form of Eq. (1) or (26) is a reasonable approximation to the dispersion curve up to such a high momentum.

From an analysis of the viscosity measurements of Whitworth, Maris[5] found the "most likely value" of α_2 to be in the range 0.9–1.3 Å^2. The difference between this value and the present result of 1.7 Å^2 is again presumable because higher-energy phonons were involved in the viscosity experiment and because a different parametrization of the dispersion was made. Similar comments apply to attempts to determine α_2 from analyses of ultrasonic attenuation and velocity data (see Ref. 6).

The results can be compared with two recent microscopic calcuations of the phonon dispersion relation. Aldrich[43] used polarization potential theory to calculate the dispersion curve and its pressure dependence. The results of a calculation of the thermal expansion based on Aldrich's dispersion curve are included in Fig. 6. It is clear that Aldrich's theory underestimates the dispersion. However, the agreement is perhaps suprisingly good considering that this is a microscopic theory. The other microscopic theory is by Bhatt and McMillan,[44] who obtain the results $\alpha_2=0.055$ Å^2, which is much too small a dispersion to be consistent with our experimental results.

IV. CONCLUSIONS

The dielectric constant ϵ of liquid ^{4}He has been measured in the temperature range 0.1–0.6 K. An accurate value for ϵ in the limit of low temperature was obtained. No evidence was found for deviations from the Clausius-Mossotti relation. The thermal expansion of the liquid was calculated from the temperature dependence of the dielectric constant and the expansion was related to the dispersion curve for thermal phonons. The data indicate that the dispersion of phonons is anomalous and quantitative information about the dispersion was obtained.

Measurements of the low-temperature expansion of dilute solutions of ^{3}He in ^{4}He are in progress and should give useful information about the dispersion relation for ^{3}He quasiparticles.

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