state by studying the rate of entropy production in this state. It is clear that entropy is produced since the energy absorbed from the external source is converted into thermal energy of the heat bath.

We can write,[5] for the total rate of entropy production,

$$\frac{dS}{dt}=-k\left[\sum_i \frac{dp_i}{dt}\log p_i+\sum_i \frac{\epsilon_i}{kT}\sum_j{}'(a_{ji}p_j-a_{ij}p_i)\right]. \quad (7)$$

The first term comes from the redistribution over the states i and vanishes in the steady state. The second term arises from the energy delivered to the heat bath at temperature T. Making use of Eq. (4) and of the fact that all dp_i/dt vanish in the steady state, we can write the rate of entropy production in the form

$$dS/dt=b(p_1^{(s)}-p_2^{(s)})(\epsilon_2-\epsilon_1)(1/T). \quad (8)$$

[5] The expression for the rate of entropy production can be derived by the method used in M. J. Klein and P. H. E. Meijer, Phys. Rev. **96**, 250 (1954), Sec. III. It can be proved that the steady state solution of Eq. (4) is the state of minimum entropy production as in the above reference. This point will be discussed further in a paper now in preparation.

The first factor, $b(p_1^{(s)}-p_2^{(s)})(\epsilon_2-\epsilon_1)$, is the rate at which energy is absorbed from the external source, and the factor $(1/T)$ arises from the transfer of this energy to the heat bath at temperature T.

Now the rate of entropy production can also be expressed in terms of the energy delivered to the heat bath per unit time as

$$\begin{aligned}dS/dt=&(\epsilon_2-\epsilon_1)(1/T)[(a_{21}p_2^{(s)}-a_{12}p_1^{(s)})\\&+(a_{31}p_3^{(s)}-a_{13}p_1^{(s)})]+(\epsilon_3-\epsilon_2)(1/T)\\&\cdot[(a_{32}p_3^{(s)}-a_{23}p_2^{(s)})+(a_{31}p_3^{(s)}-a_{13}p_1^{(s)})], \quad (9)\end{aligned}$$

where we have written $(\epsilon_3-\epsilon_1)=(\epsilon_3-\epsilon_2)+(\epsilon_2-\epsilon_1)$. It is seen from the foregoing analysis, particularly Eqs. (5) and (6), that these two expressions, Eqs. (8) and (9), agree precisely because of the failure of the principle of detailed balance and because of the existence of a cycle of transitions.

In conclusion, we may state that the argument carried out above for a special example should apply quite generally: the principle of detailed balance cannot hold in nonequilibrium steady states.

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Ultrasonic Propagation in Magnetically Cooled Helium*†

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Measurements have been made of the velocity and attenuation of ordinary sound in liquid helium at a frequency of 12.1 Mc/sec, over the temperature range from 1°K down to 0.1°K. The velocity is essentially independent of temperature and has the value 240±5 m/sec. The attenuation passes through two closely spaced maxima near 0.9°K and then falls smoothly to zero as the absolute zero is approached. These results agree qualitatively with the theoretical predictions of Khalatnikov.

I. INTRODUCTION

THE present paper describes a series of experiments on the propagation of ordinary ("first") sound in liquid helium below 1°K. Interest in these measurements was first stimulated by the experiments performed by one of the authors[1–2] in the range of temperatures obtainable by pumping on the helium bath and by the theoretical calculations of Khalatnikov.[3–5] These measurements showed that the velocity of propagation u_1 levels off to an almost constant value at temperatures somewhat above 1°K, but that the attenuation passes through a maximum just below 1°K and exhibits the frequency dependence, over the range from 2 Mc/sec to 12 Mc/sec, characteristic of a relaxation process. This behavior was predicted by Khalatnikov. It was therefore decided to extend the measurements into the temperature range obtainable by the adiabatic demagnetization of a paramagnetic salt, focusing attention chiefly upon the attenuation. These experiments were performed under the saturated vapor pressure at a frequency of 12.1 Mc/sec, and extend from about 1°K to 0.1°K.

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† Preliminary results have been reported in a recent Letter by the authors [Phys. Rev. **95**, 565 (1954)].

‡ Now at Lincoln Laboratory, Massachusetts Institute of Technology.

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[2] C. E. Chase, Proc. Roy. Soc. (London) **A220**, 116 (1953).

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II. EXPERIMENTAL METHOD

1. Demagnetization Cryostat

The cryostat in which the experiments were performed is shown schematically in Fig. 1. This apparatus is essentially the same as one originally designed by Dr.

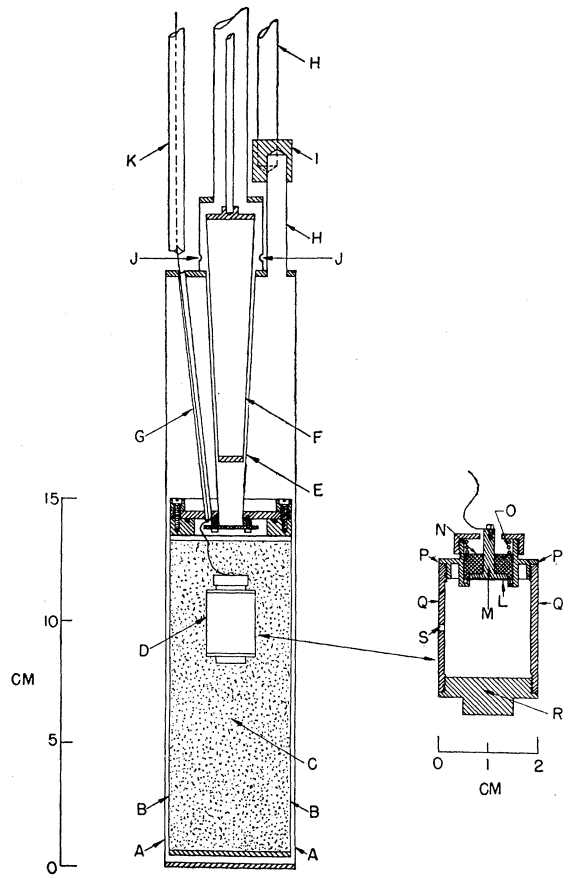

FIG. 1. The demagnetization cryostat: *A*, *B*, concentric cans; *C*, paramagnetic salt; *D*, etalon; *E*, *F*, thin-walled cones; *G*, vaseline-filled tube for leads; *H*, pumping tube; *I*, radiation trap; *J*, holes to admit helium; *K*, coaxial cable. Inset: *L*, quartz crystal; *M*, electrode; *N*, Bakelite insulator; *O*, beryllium-copper spiral spring; *P*, brass case; *Q*, brass tube; *R*, reflector; *S*, hole to admit helium.

J. Ashmead, at present in use in the Royal Society Mond Laboratory, Cambridge. It differs from conventional apparatus for the magnetic cooling of liquid helium chiefly in the method of introducing the liquid from the bath and subsequent isolation during demagnetization. The two thin-walled stainless-steel cones, *E* and *F*, are machined and lapped to fit together as closely as possible. The inner cone is sealed off full of air, which freezes at helium temperatures to produce a thermal vacuum inside the cone. This cone may be raised and lowered by means of a thin-walled stainless-steel tube extending to the cryostat cap. When the cone is raised, helium from the surrounding bath flows in to fill the experimental chamber, and the liquid filling the space between the cones provides almost perfect thermal contact for the rapid removal of the heat of magnetization. When the cone is seated, the heat flow is reduced to a very low value, and the thermal isolation is sufficient to keep the experimental chamber colder than the bath for more than an hour.

Since it is not necessary to use exchange gas with this apparatus, tedious pumping out of the vacuum space is eliminated and magnetizing times can be very short (of the order of two or three minutes). It is therefore possible to obtain many more demagnetizations during a run. In actual practice, precooling was facilitated by introducing approximately 1 cm of air into the vacuum space; this freezes out, however, and presents no problem. Occasionally a few bursts of helium gas were introduced during the transfer of liquid helium to assist in the further cooling of the large amount of paramagnetic salt present. This procedure was probably not strictly necessary, but it somewhat increased the efficiency of transfer. This exchange gas was readily pumped out before the bath temperature reached 1°K, and pressures better than 10^{-6} mm Hg were then maintained for the duration of the run.

The inner can *B* was filled with approximately 200 grams of ferric ammonium alum, and the ultrasonic etalon *D* was buried in the middle of this salt to make certain that the two were in thermal equilibrium. Most of the salt was in the form of large crystals, with the interstices loosely filled with fine powder. This arrangement allows the helium to penetrate freely through the salt and makes possible the rapid establishment of thermal equilibrium. The leads for the ultrasonic apparatus and for a resistance thermometer (not shown in the figure) were brought out through three stainless-steel tubes *G*, one-eighth inch in diameter, which were then filled with Vaseline. The Vaseline freezes and prevents the flow of helium, and has a low thermal conductivity in the solid state. The inner can was held in place by means of a gold O-ring seal.

2. The Etalon

The ultrasonic etalon is shown in the inset in Fig. 1. An X-cut quartz crystal *L*, 1.0 cm in diameter, was held against the face of the outer case *P* by the spring-loaded electrode *M* and insulator *N*. The curious shape of the outer case was designed to minimize direct propagation of the ultrasonic pulses through the supporting tube *Q*, which would give rise to severe clutter. This tube serves to align the crystal with the reflector *R*, and its faces are machined parallel to within 0.0002 inch at room temperature. Subsequent misalignment upon cooling was found to be negligible. By using different supporting tubes, it was easy to change the path length between runs. Each path was measured at room temperature with micrometers and a micrometer depth gauge, and a correction of 0.5 percent was made to allow for contraction upon cooling. This was only an approximate estimate, but introduced errors of negligible importance compared with the outer sources of experimental error.

3. Electronics

The electronic apparatus is essentially the same as that described earlier.[1,2] A pulsed oscillator was used

to drive the quartz crystal at its resonant frequency. The resulting sound pulses, after transmission through the helium and reflection at the reflector, were detected by the same crystal, amplified, demodulated, and displayed on an oscilloscope. A DuMont type 256D *A/R* oscilloscope was used for this purpose, and provided the necessary synchronizing pulses as well as a calibrated time delay, which was used in the measurement of the velocity.

For measuring the attenuation, a comparison pulse from a pulsed signal generator at the operating frequency was fed through a calibrated attenuator and into the input of the amplifier along with the received signal. By setting the attenuator so that both pulses appeared the same height on the oscilloscope, it was possible to determine the signal size directly, and all nonlinearities in the amplifiers were cancelled out.

4. Demagnetization Technique

Demagnetizations were performed from a starting temperature of about 0.9°K, obtained by means of a large diffusion pump, and with an initial field of 6200 gauss. Under these conditions the lowest temperature reached was 0.10°K. During demagnetization the following procedure was adopted. With the bath at 0.9°K and the inner cone raised, the magnet was turned on. The heat of magnetization was rapidly conducted away to the bath; within two or three minutes, the temperature of the inner chamber had fallen once more to its original value. The progress of this cooling was followed by means of the resistance thermometer. After equilibrium was reached, the cone was firmly seated, the magnet switched off and moved away, and the experimental observations were begun.

During the initial runs with this apparatus, the warming rate was very high, and the cryostat warmed up from the lowest temperature to bath temperature in about ten minutes. It was later found that a large part of this heat leak was caused by eddy currents induced in the brass can by the field of the measuring coil which was used to determine the temperature of the salt. When this field was reduced, the warming time was more than one hour.

5. Temperature Measurement

The temperature of the helium bath was determined from the vapor pressure of the liquid by using the vapor pressure tables of van Dijk and Shoenberg.[6] A mercury manometer read with a cathetometer was used for pressures above 4 cm Hg, a McLeod gauge for lower pressures. A correction for the hydrostatic pressure head was applied at all temperatures above the λ point.

Temperatures below 1°K were measured by means of an ac mutual inductance bridge operated from a stable $33\frac{1}{3}$ cps source. The output of the bridge was amplified by a three-stage "twin-T" amplifier with a band width of about one cycle, and displayed on an oscilloscope. By synchronizing the oscilloscope to the bridge input, it was possible to tell whether the unbalance of the bridge was resistive or reactive and in which direction the resistance or mutual inductance must be varied, thus making it possible to balance the bridge very rapidly. Measurements could be made to within about 2 μh, corresponding to a temperature error of 0.002°K at 1°K. The errors in the absolute temperature are probably greater than this, however, because of errors in the calibration region above 1°K where the sensitivity was much smaller and because of a tendency for the calibration to drift slowly during the course of a run. During one run this drift amounted to 0.05°K at 1°K, over a period of several hours.

The mutual inductance readings in the region above 1°K were fitted to a Curie law by the method of least squares. At the lowest temperatures the departures from Curie's law become significant; these were determined from the measurements of Kurti, Laine, and Simon[7] on the susceptibility of iron ammonium alum. The correction amounted to 0.015°K at 0.10°K. In order to determine this correction, the demagnetizing factor of the sample had to be calculated. For this purpose, the salt was approximated by an ellipsoid of major and minor axes 6.5 cm and 2.5 cm, respectively, and the demagnetizing factor calculated from formulas given by Garrett.[8] It turned out, however, that both the present sample and that used by Kurti, *et al.*, were sufficiently close to a long cylinder so that this was of small importance at temperatures down to 0.1°K.

III. SOURCES OF ERROR

1. Temperature Errors

The chief sources of temperature errors were mentioned in the preceding section: errors in the mutual inductance calibration and drift of this calibration, presumably the result of the effect of drift of room temperature on the balancing coils. The first of these errors would be most important at low temperatures where the extrapolation of the calibration curve is greatest; however, from the good agreement between points obtained in separate runs with independent calibrations it seems unlikely that this error is greater than perhaps 0.01°K. The second error appeared to be a constant rate of drift of a few microhenries per hour, and changed the readings near 1°K by a few hundredths of a degree, while affecting the lowest temperatures very little. It was found that if the first and last of a series of demagnetizations were adjusted so that the attenuation agreed with its known value above 1°K and a linear rate of drift was assumed, the points for intermediate demagnetizations agreed well with the rest. This pro-

[6] H. van Dijk and D. Shoenberg, Nature (London) **164**, 151 (1949).

[7] Kurti, Laine, and Simon, Compt. rend. **204**, 675 (1937).

[8] C. G. B. Garrett, *Magnetic Cooling* (Harvard University Press, Cambridge, 1954).

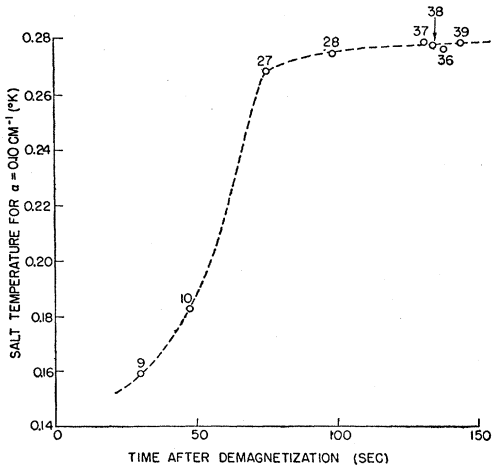

FIG. 2. Effect of warming rate on thermal equilibrium between salt and helium. The numbers are serial numbers of demagnetizations.

cedure was therefore adopted for determining the temperatures in those runs shown as black dots in Fig. 3. In the earlier runs (shown as open circles) this effect was unimportant, since the runs were of shorter duration and the measurements did not extend to sufficiently high temperatures for the error to be appreciable.

2. Thermal Equilibrium Between Salt and Helium

It has often been observed that in demagnetization experiments where the warmup time is very short, thermal equilibrium between the salt and helium is not attained. This question must therefore be carefully considered if any weight is to be attached to measurements made within the first few minutes after demagnetization. In the present work a number of the measurements were made with total warmup times of the order of ten minutes, and it was observed that these results were completely self-consistent and agreed well with later measurements made with warmup times of about an hour as long as the warming rate was less than a certain critical amount. For faster warming rates, however, a considerable shift in the attenuation curves in the direction of lower magnetic temperature was observed. This can be explained by the assumption that the helium is not actually as cold as the salt, which is serving both as coolant and thermometer. This effect is illustrated in Fig. 2. In this diagram the magnetic temperature corresponding to an attenuation of 0.10 cm^{-1} is plotted against the time after demagnetization at which this value was reached. The attenuation of sound thus serves as a thermometer to determine the temperature of the helium, while the magnetic measurements are used to determine the temperature of the salt. It will be seen that for times less than about 70 sec after demagnetization the helium is definitely warmer than the salt, but that equilibrium is rapidly reached thereafter. All those points obtained with insufficiently long warmup times have been omitted from the curves in the following section.

3. Velocity Errors

Velocity measurements made with a fixed path length are necessarily subject to certain errors because of the necessity of measuring the absolute magnitude of the path length and estimating the amount of its contraction upon cooling. An even larger error may result from the assumption that this figure corresponds to the acoustical path length, for it has been found that the processes of reflection and detection by quartz crystals lead to spurious extra delays which may be of the order of a few microseconds. This effect is not understood at present but presumably results from the relatively high Q of the crystals, which require several cycles to start oscillating. A third source of error lies in the difficulty of estimating the positions of the feet of the input pulse and received signal as the attenuation varies. This may have introduced errors of 2 μsec or 3 μsec in the present measurements. As a consequence of all these sources of error, the velocity measurements are considered to be accurate only to within ± 2 percent.

4. Attenuation Errors

The largest single source of error in the attenuation may come from the fact that the absolute value cannot be found from measurements with a single path length, and the results had therefore to be fitted to the known attenuation above 1°. However, the accuracy of this fitting procedure is confirmed by the fact that results obtained with two different path lengths agree well with one another, and indirectly by the fact that the resulting attenuation curve falls smoothly to zero at the absolute zero. A further source of error arose from possible drift of the size of the input pulse during a run; this was eliminated as far as possible by checking the signal size against the known attenuation at bath temperature at the end of every demagnetization. With one exception this drift was negligible; in that instance it amounted to 0.04 cm^{-1}, and the results were consistent with the other runs when a correction of this amount was applied. The largest remaining source of error was the calibration of the attenuator on the signal generator, which was accurate only to a few percent. As a result of all these sources of error, it is estimated that the attenuation measurements are accurate to within ± 0.05 cm^{-1}.

IV. RESULTS

1. The Velocity

Because of the inherent difficulties in velocity measurements discussed in the preceding section and the fact that no appreciable changes occurred below 1°K, only a few observations were made. From the results of five different demagnetizations, with path lengths of 3.94 cm and 1.96 cm, it was concluded that the velocity at 0.1°K is equal to 240 ± 5 m/sec. This is in agreement

with the earlier estimate[2] of 239±2 m/sec obtained from extrapolation of the results at higher temperatures. Although the measured value is less accurate than the extrapolated one, it excludes the possibility of any gross changes in the velocity in this range and confirms the relationship between the velocities of first and second sound given by Landau[9] and discussed earlier by one of the authors.[1,2] Measurements in the region between 0.7°K and 1.1°K were even more difficult because of the high attenuation, and were only possible with the shorter of the path lengths given above. It was found, however, that no appreciable variations occur in the neighborhood of the maximum attenuation.

2. The Attenuation

The results of the attenuation measurements are shown in Fig. 3. The open circles represent those points taken with a path length of 3.94 cm. With this path length the signal became comparable with noise near 0.7°K, and measurements in the neighborhood of the maximum attenuation were impossible. A shorter path length of 1.96 cm was accordingly used to investigate this region, and the results are shown as solid circles. The full curve reproduces the earlier results of one of the authors.[2] The points shown are only representative of all those taken in a total of 35 demagnetizations with the longer path and 10 with the shorter. Since only the relative attenuation was determined, the results have been adjusted to agree with the earlier results above 1.2°K. In all of those runs with the longer path length, the warmup time was only about ten minutes to bath temperature, and observations were possible for about four minutes before the attenuation became unmeasurably high. In the later runs the warmup time was over an hour during three demagnetizations from a bath temperature of 0.9°K, and only about 10 minutes during several other runs starting from 1.2°K.

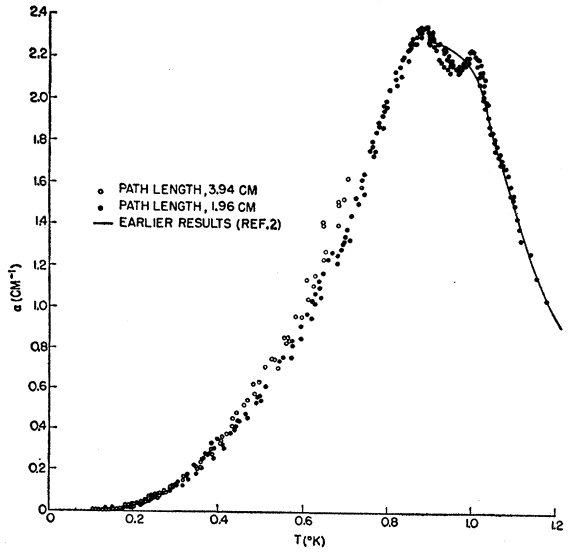

FIG. 3. Attenuation of sound in liquid helium at 12.1 Mc/sec.

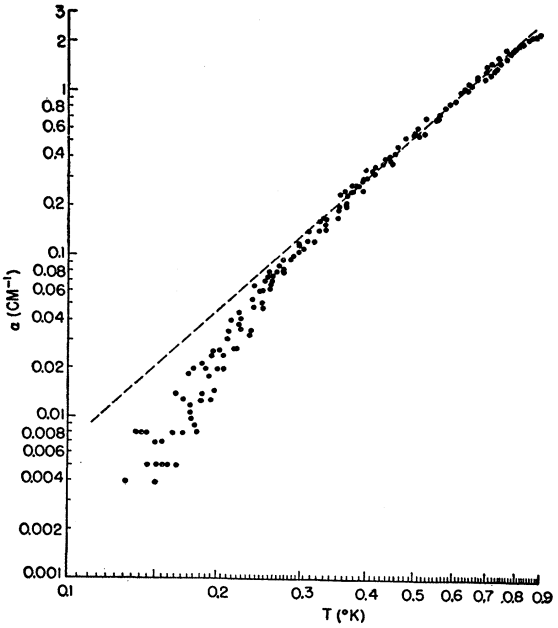

FIG. 4. Attenuation of sound in liquid helium at 12.1 Mc/sec. The slope of the dotted line is 2.8.

The most significant feature of Fig. 3 is the presence of a double maximum in the attenuation around 0.9°K. This double maximum was not observed during the earlier measurements, apparently because the relatively few points taken at these temperatures happened to fall on the smooth curve shown in the diagram and the minimum was completely missed. The earlier results at 2.0 Mc/sec and 6.0 Mc/sec are presumably similarly in error. The existence of these twin maxima provides direct confirmation for the prediction of Khalatnikov[3–5] that the attenuation of sound in helium II is determined by two relaxation times. The relation of these measurements to the theory will be discussed in more detail in the following section.

Another important fact is that the attenuation falls smoothly to zero as the absolute zero is approached. Such behavior is to be expected on the basis of any theory that explains the attenuation of sound in terms of the elementary excitations of the liquid, since the density of these excitations falls to zero as the absolute zero is approached.

In Fig. 4 the results below 0.8°K are reproduced on a logarithmic plot. From this diagram another effect may be seen: above 0.3°K the attenuation is quite accurately proportional to $T^{2.8}$; at lower temperatures the variation is more rapid. It does not appear to be possible to fit the entire curve to any sort of exponential or other simple relation, and therefore it seems probable that this abrupt change near 0.3°K represents some sort of change in the fundamental processes giving rise to attenuation of sound. It is perhaps significant that this

[9] L. Landau, J. Phys. (U.S.S.R.) 5, 71 (1941).

occurs just in the region where the phonon mean free path is becoming comparable with the dimensions of the apparatus, and where the velocity of second sound starts to rise from approximately $u_1/\sqrt{3}$ to u_1.[10,11]

V. KHALATNIKOV THEORY

In his derivation of the temperature variation of the attenuation of first sound in liquid helium,[3–5] Khalatnikov assumes that the mechanism responsible for the attenuation is inelastic collisions of the elementary excitations, that is, processes in which the numbers of phonons and rotons are changed. From his calculations it turns out that there are two relaxation times governing the attenuation, corresponding to the two dominant processes: phonon-phonon and phonon-roton interactions. The presence of two maxima in the attenuation *vs* temperature curve suggests that this is indeed the case. However, an exact comparison of theory with experiment is complicated by the fact that elastic collision processes become important at temperatures of the order of 0.9°K. These processes are the ones which, according to Landau and Khalatnikov,[12,13] are responsible for the viscosity of the liquid. At temperatures above 1°K the viscous relaxation times are so short that the viscosity at 12 Mc/sec is the same as the viscosity at zero frequency, and the viscous attenuation can be found from the usual Stokes formula. This part of the attenuation is independent of that calculated by the Khalatnikov formulas, and the two contributions must be added to obtain the experimentally observed value. Below 1°K, however, the viscous relaxation times become comparable with the time of one cycle of the sound wave, and the Stokes formula may no longer be used to calculate the viscosity attenuation. In this region the concepts of first and second viscosity no longer apply, and all the relaxation times must be considered explicitly as contributing to the absorption. Since the maximum viscous attenuation is believed to occur just in the region where the attenuation from inelastic scattering processes is greatest, the situation is extremely complicated, and a quantitative calculation does not appear to be feasible.

Fortunately, it is possible to reach certain qualitative conclusions without the aid of a complete theory. At any temperature, the viscous attenuation will certainly be no greater than that calculated from the low-frequency viscosity by the Stokes formula, since the effect of the finite viscous relaxation time is to make the viscous attenuation pass through a maximum and eventually fall to zero. At 0.9°K and 12.1 Mc/sec this is less than 20 percent of the total attenuation, so that the errors involved in neglecting it altogether are not large. The constants of the Khalatnikov theory may thus be determined approximately. If the two maxima in the attenuation are associated with the phonon-phonon and phonon-roton relaxation times, the constants a and b occurring in Eqs. (2) and (3) of reference 6 may be evaluated from the positions of the maxima independently of the behavior above 1°K. An estimate of their values gives $1/a \approx 2\times10^{-44}$, $1/b \approx 0.8\times10^{-50}$. This value of $1/a$ is approximately twice as large as the value found earlier[2] from the data at higher temperatures, and would make the agreement in that range considerably worse. The exact shape of the curve is, however, extremely sensitive to the value of Δ, one of the parameters in Landau's energy-momentum relation for rotons[9,14] and this parameter is not known with any degree of accuracy. In fact, it now seems likely that sound attenuation measurements provide the most accurate method of determining its value, if the effects of relaxation in the viscous processes can be calculated. An approximate estimate suggests that a value of $\Delta = 9.2$°K would provide good agreement between theory and experiment both in the relaxation regions and at higher temperatures.

ACKNOWLEDGMENT

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[13] L. Landau and I. M. Khalatnikov, J. Exptl. Theoret. Phys. (U.S.S.R.) **19**, 709 (1949).
[14] L. Landau, J. Phys. (U.S.S.R.) **11**, 91 (1947).