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Third Sound in Liquid Helium Films*

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The existence of "third sound," a surface wave on thin films of superfluid helium, has been demonstrated experimentally. The velocity and attenuation of the waves have been measured as functions of excitation frequency, film thickness, and temperature. In the main, the results agree with earlier predictions and yield new information about the forces which cause the formation of helium films. Preliminary measurements on third sound in moving films and on surface-tension ripples on bulk liquid helium are also described.

1. INTRODUCTION

IT is well known that bulk liquid helium II is capable of transmitting two distinct types of wave motion: first sound, in which the normal and superfluid components oscillate in phase, giving a pressure wave; and second sound, in which the two components oscillate out of phase, giving a temperature wave. In 1959, one of us[1] predicted that two further types of wave motion, which were termed third and fourth sound, might be expected to occur when the helium is subjected to certain boundary conditions. Actually, the existence of fourth sound had already been anticipated on rather different grounds by Pellam.[2] Both third and fourth sound have now been detected experimentally.[3,4] The present paper contains a more detailed description of our experiments on third sound, of which a preliminary report has already appeared elsewhere.[3]

Third sound is a surface wave on a liquid helium film, in which the superfluid component oscillates while the normal component remains locked to the wall. Coupled with the liquid motion is a distillation process through the vapor which tends to reduce the temperature differences between peaks and troughs of the wave. The restoring force arises from the combined effect of the classical pressure gradient and a term due to the thermomechanical effect. The result is an oscillation in the thickness of the film with second order variations in pressure and temperature. Third sound is therefore somewhat similar to classical shallow-water waves, but the superfluidity of liquid helium is essential to its existence since such a wave would be rapidly attenuated in a thin film of an ordinary viscous liquid.

In the elementary theory of third sound, the velocity of the superfluid is assumed to be independent of distance from the wall, and motions of the normal fluid are neglected. The velocity u_3 is then given by

$$u_3^2=(\rho_s/\rho)fd[1+TS/L], \qquad (1)$$

where ρ is density, ρ_s density of superfluid, f restoring force per unit mass at surface of film due to attraction of the wall, d film thickness, T temperature, S entropy, and L latent heat of vaporization. The term TS/L is about 0.01 at 1°K and 0.15 at the λ temperature. Hence, u_3 is approximately proportional to $(\rho_s/\rho)^{1/2}$, vanishing as expected at the λ point. According to Eq. (1) the velocity is independent of frequency. More detailed analysis suggests that at frequencies above about 10^5 cps, a phase difference may develop between the superfluid flow and the distillation process, possibly giving rise to dispersion and attenuation. However, the experiments to be described below indicate that other sources of attenuation may be important besides those discussed in the original theory.

The restoring force f is equal to the derivative of the chemical potential μ. According to the theory of van der Waals' forces due to Dzyaloshinskii *et al.*,[5] μ is equal to α_1/d^4 for thick films and $\alpha_2/\lambda d^3$ for thin films, where α_1 and α_2 are constants determined by the nature of the wall, and λ is a characteristic length marking the transition between the two formulas. The measurements of Hemming[6] suggest that this theory is in satisfactory agreement with experiment for helium films. Except in the transition region between the two cases, it is convenient to write μ equal to α/d^n. Then f is given by $n\alpha/d^{(n+1)}$, and the film thickness at height h above the bulk liquid is found by equating α/d^n to the gravitational potential gh. Substituting into Eq. (1) and neglecting the term TS/L, we obtain

$$u_3^2=n(\rho_s/\rho)gh. \qquad (2)$$

Taking n equal to 3, this gives a velocity for third sound of about 50 cm/sec for a height of 1 cm. A surprising feature of Eq. (2) is that u_3 is independent of α; in other words the velocity should be identical for different types of wall. Since the value of n may be determined from the experimental results, measurements of third sound give an alternative test of the theory of Dzyaloshinskii *et al.*

In the following experiments, the third sound was excited in a horizontal film of helium II by pulses of infrared radiation. The variations in thickness were

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[1] K. R. Atkins, Phys. Rev. **113**, 962 (1959).
[2] J. R. Pellam, Phys. Rev. **73**, 608 (1948).
[3] C. W. F. Everitt, K. R. Atkins, and A. Denenstein, Phys. Rev. Letters **8**, 161 (1962).
[4] I. Rudnick and K. A. Shapiro, Phys. Rev. Letters **9**, 191 (1962).
[5] I. E. Dzyaloshinskii, E. M. Lifshitz, and L. P. Pitaevskii, Advan. Phys. **10**, 165 (1961).
[6] D. Hemming, Bull. Am. Phys. Soc. **8**, 91 (1963).

measured by an optical method developed originally by Drude and applied to static helium films by Jackson *et al.*[7] Broadly speaking, the results confirm the theory besides suggesting a number of new features concerning the behavior of helium films.

2. EXPERIMENTAL PROCEDURE

The main part of the apparatus is shown in Fig. 1. The helium film is formed on a horizontal stainless steel mirror, H, 4 cm long and 0.9 cm wide, contained in an enclosed experimental chamber. Mirrors of other materials are employed in certain subsidiary experiments. The mirror is mounted on a U-shaped bracket 12 cm high which is supported within a copper framework F by means of four levelling screws G. The use of a horizontal mirror provides a film of uniform thickness which can be varied by altering the level of the liquid in the chamber. The framework of heavy annealed copper serves to maintain thermal equilibrium. For the same purpose, there are two horizontal sheets of copper in the vapor above the mirror, and the whole system is surrounded by a sheath of high-conductivity copper extending to the bottom of the experimental chamber.

The infrared radiation used to excite the third sound is obtained from a 54-W strip-filament "sound-recording" lamp I, mounted on an optical table to one side of the cryostat. With the aid of an external lens M and a 45° mirror E, inside the experimental chamber, an image of the filament 0.1 cm broad is formed over the whole width of the horizontal mirror H. The intensity of the radiation is controlled by an iris diaphragm, K, so arranged that at maximum aperture the emitter supplies about twice the energy required to boil away the film completely. The radiation is pulsed by a chopping wheel W driven by a variable speed motor. Visible light is eliminated by means of an infrared filter L. All these parts are mounted on an optical table, which stands in a vertical hydraulic press in front of the apparatus. By raising or lowering the press, the image of the filament may be displaced along the mirror in increments of 0.01 cm. Also attached to the press is a cathetometer Q to measure the level of the helium inside the experimental chamber.

The detection system is mounted on top of the cryostat. A narrow beam of mercury green or blue light A, polarized at 45° to the axis of the stainless steel mirror H, passes down into the experimental chamber by the left-hand vertical tube shown in Fig. 1. It is reflected on to the mirror at an angle of $67\frac{1}{2}°$ by the left-hand prism D, and passes thence via the second prism up the right-hand tube to a quarter-wave plate, analyzing prism, and photomultiplier. Upon reflection, the beam becomes elliptically polarized to an extent that depends on the angle of incidence, and the thickness and refractive index of the helium film. The quarter-wave plate

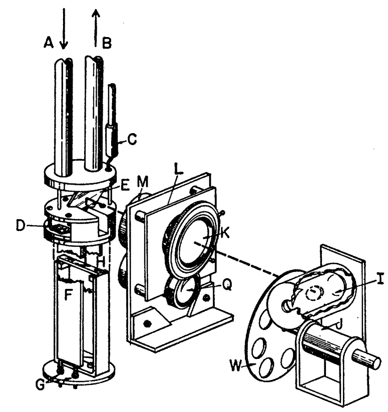


FIG. 1. General view of apparatus. A, incoming plane polarized light beam; B, outgoing light beam which is elliptically polarized after reflection from the mirror H; C, needle valve for filling closed experimental chamber with liquid helium; D, $67\frac{1}{2}°$ prism; E, 45° mirror; F, copper framework; G, leveling screws; H, horizontal mirror holding the helium film on which third sound is propagated; I, light source; J, rotary switch for phase sensitive detector; K, iris diaphragm; L, infrared filter; M, condensing lens; W, chopping wheel; Q, cathetometer.

converts the emerging beam back into plane-polarized light, but the plane of polarization is tilted through a small angle proportional to the thickness of the film. With the angle of incidence of $67\frac{1}{2}°$ (the optimum value for a stainless-steel mirror) a helium film 200 Å thick produces a change in the plane of polarization of about 1°.

It follows that, when the film thickness oscillates, there is an oscillation in space of the plane of polarization, which is converted by passage through the analyzing prism into an oscillation in intensity. The output from the photomultiplier is amplified and fed into a phase-sensitive detector made from a twelve pole rotary switch J, driven on the same shaft as the chopping wheel. After passing through an integrating circuit, the signal emerges finally as a progressive displacement of a pen-recorder, the magnitude of which depends on the amplitude and phase of the third sound wave.

The most critical problem of design was to devise a satisfactory detection system. In the original form of the apparatus the detection beam entered through the side of the Dewar vessel, but this arrangement gave rise to wild fluctuations in the plane of polarization, owing to the bubbles and convection currents in the liquid nitrogen. Accordingly, the apparatus was arranged with the detection beam entering from above. Very careful design was then required in order to align the beam accurately enough to get a really strong signal. The arrangement finally adopted is indicated in Fig. 2. The two $67\frac{1}{2}°$ prisms D rest in copper cradles N supported from the copper prism table by inverted kinematic mounts. The cradles are retained in position from above by pairs of compression springs P. The main alignment is made before assembling the apparatus by means of the three leveling screws O attached to each

[7] A. C. Ham and L. C. Jackson, Proc. Roy. Soc. (London) **A240**, 243 (1957).

prism mount. Further small adjustments are possible even at helium temperatures by displacing the whole experimental chamber sideways with the aid of a chain and screw device (not shown). By these means an intense signal is obtained and the proportion of scattered light is reduced to about 0.3% of the total. The signal-to-noise ratio also depends on the setting of the analyzing prism. Trial and error indicated that the optimum position is about 10° away from the minimum. With this setting and a time constant of 10 sec in the integrating circuit, the detection system is capable of measuring oscillations in film thickness of 2 or 3%. The radiation from the detector, being much weaker than that from the emitter, does not disturb the film thickness in any way.

A collimating slit immediately above the first $67\frac{1}{2}°$ prism delimits the detection beam to a narrow line, 0.01 cm wide, parallel to the emitter. Hence, the shortest measurable wavelength of third sound is about 0.02 cm. For convenience the detector remains fixed about 1 cm from the end of the mirror, the propagation distance being varied by displacing the emitter in the manner already described. The object of using an infrared exciting source is to avoid a spurious signal at the detector due to scattered light from the emitter. The radiation reaching the mirror comprises wavelengths between 8500 and 20 000 Å, a region to which the photomultiplier in the detection system is not sensitive. The use of a strip source extending over the whole width of the mirror ensures that the third sound is propagated in nearly plane waves. Unless otherwise stated, all measurements were made at 1.2°K.

3. RESULTS

The two curves in Fig. 3 illustrate a typical experiment in which the output was measured as a function of the position of the emitter for two different settings of the phase-sensitive detector. For curve 1, the detector was adjusted to accept signals exactly in phase with the exciting radiation, so that a large signal was observed when emitter and detector were superimposed at point A. For curve 2, the detector was approximately 90° out of phase with the emitter and so a small signal was observed at A. We consider the existence of a deflection to imply that there were oscillations in thickness along the film. The approximately sinusoidal variation of amplitude with distance indicates a variation in phase of the oscillations relative to the detector.

The two curves may be regarded as delineating the profile of the film at two instants separated by a quarter of a cycle. The pronounced minimum in curve 1 at A represents the deep pit dug in the film by the infrared beam. The displacement between the nodes of the two curves indicates that in this case a travelling wave had been excited, rather than a standing wave. However, in some experiments with thinner films standing waves were excited. The wavelength on Fig. 3 is 0.92 cm. Since the excitation frequency was 100 cps, this means that the wave velocity was 92 cm/sec. The amplitude was $\pm 25\%$ of the mean film thickness, and the attenuation coefficient was about 0.3 cm^{-1}.

Proof that there was a genuine oscillation in film thickness is supplied by the fact that the signal disappeared: (1) when the emitter was switched off, (2) when the detecting light was switched off (this eliminates any possibility of a spurious signal due to scattered radiation from the emitter), and (3) when the whole film was evaporated by means of an auxiliary radiant heater. The only likely explanation of the oscillations apart from third sound is that the film was undergoing evaporation from behind by thermal waves propagating in the mirror. Three pieces of evidence indicate that the signal was not due to thermal waves: (1) The observed attenuation was small, whereas thermal waves are always strongly attenuated; (2) the velocity for a given thickness was independent of frequency, whereas the velocity of thermal waves is proportional to the square root of the frequency; (3) the velocity depended strongly on film thickness, whereas thermal waves would depend solely on the nature of the mirror.

It appears, therefore, that the existence of third sound is established. Its variation with excitation frequency,

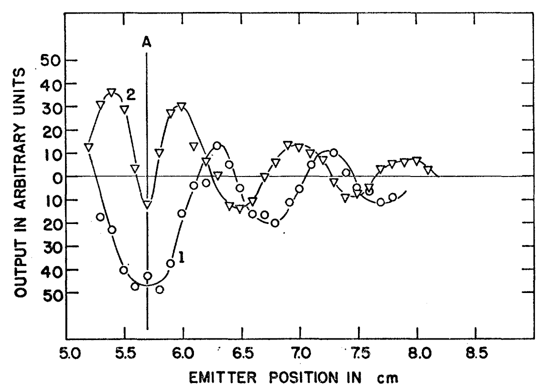


FIG. 3. Output of the phase-sensitive detector as a function of the position of the emitter relative to the receiver. ○, curve 1, the detector adjusted to accept signals at the instant the infrared beam has maximum intensity. ▽, curve 2, the detector adjusted to accept signals approximately one quarter of a period before curve 1. A is the position at which the emitter and detector are exactly superimposed.

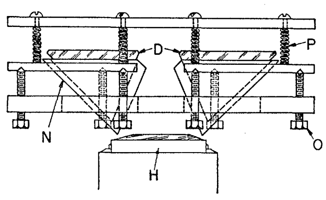


FIG. 2. Details of the prism system. D, $67\frac{1}{2}°$ prisms; H, horizontal mirror holding the helium film on which third sound is propagated; N, prism cradle; O, leveling screws; P, retaining springs.

film thickness, and temperature has been investigated systematically for several different mirrors. In order to reduce distortion the intensity of the exciting radiation was usually set at about 25% of that required to boil away the film at point A. The main results are summarized in Figs. 4 to 8, certain other subsidiary measurements being carried out as described below.

Figure 4 shows typical measurements of velocity as a function of frequency for two different experiments at a film height of 9.0 cm. The results are accurate to about 2% except at frequencies lower than 30 cycles where the wavelength becomes comparable with the total length of the mirror. At frequencies above 1300 cycles the wavelength is too short to be resolved with the present detection system. Over the range examined the velocity of third sound proved to be independent of frequency in accordance with Eqs. (1) and (2).

The variation of velocity with film height for three different mirrors is plotted logarithmically in Fig. 5.

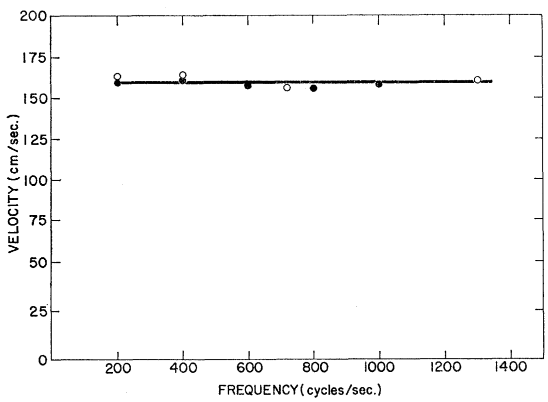


FIG. 4. Velocity versus frequency for a film height of 9.0 cm. The open and filled circles represent two runs on different days.

Curves A and B give the experimental results for two stainless steel mirrors with different degrees of polishing. Examination with a phase-contrast microscope indicated that any polishing marks on the first mirror were shallower than the film depth (i.e., less than 300 Å). By contrast, the polishing marks on the second mirror were substantially deeper (above 1000 Å). Since the two curves were identical, it appears that third sound is not sensitive to the detailed shape of the substrate. The surfaces were carefully cleaned with detergent and low conductivity distilled water before each experiment, but it was found that the velocity of third sound was not greatly affected even by the presence of grease on the mirror. For film heights above 0.8 cm the points lie on a straight line of slope approximately 0.5 in accordance with Eq. (2). The actual magnitude of the velocities is substantially higher than that reported in our previous paper.[3] The discrepancy may be attributed to the fact that the original measurements were obtained with an earlier version of the apparatus in which fewer precautions had been taken to maintain the liquid and vapor in thermal equilibrium. The present results must therefore be regarded as superseding the original ones.

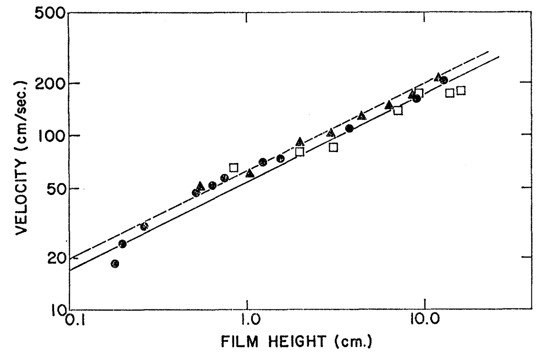
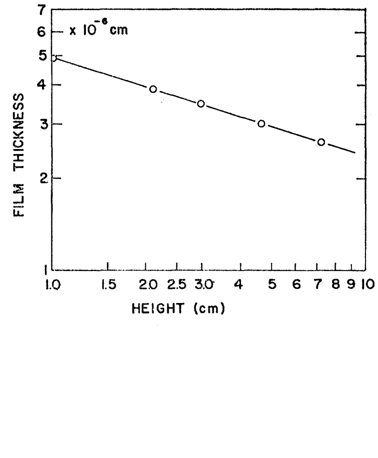


FIG. 5. Velocity of third sound versus film height. ▲, A, highly polished stainless steel; ●, B, roughly polished stainless steel; □, C, nickel. ——, theoretical curve with $n=3$. – – – – – –, theoretical curve with $n=4$.

The two lines on Fig. 5 show the calculated velocities, obtained by assuming Eq. (2) and values for n of 3 and 4, corresponding respectively to the thin-film and thick-film approximations of Dzyaloshinskii *et al.*[5] Measurements of film thickness versus height, reproduced in Fig. 6, indicate that, for heights between 1.0 cm and 8.0 cm, n is equal to 3.1 ± 0.1. These results were obtained by measuring the change in the minimum position of the analyzing prism when the film was boiled away. No corresponding results are available for film heights less than 1.0 cm; but the experiments of Hemming[6] suggest that the thick-film approximation ($n=4$) may be applicable in this region. Hence, the velocity of third sound should coincide with the $n=3$ line for film heights above 1.0 cm, bending gently upwards towards the $n=4$ line for film heights below 1.0 cm. Curve C of Fig. 5, which gives preliminary measurements made by K. Pickar, on a nickel mirror appears to lie fairly near the $n=3$ line for heights above 1.0 cm, but curves A and B lie much closer to the $n=4$ line, and curve *downwards* in the region below 1.0 cm. Hence, although the theory is in broad agreement with experiment, Eqs. (1) and (2) appear to be only approximately correct. Further experiments are required to elucidate the details of the discrepancy.

FIG. 6. Film thickness versus film height.



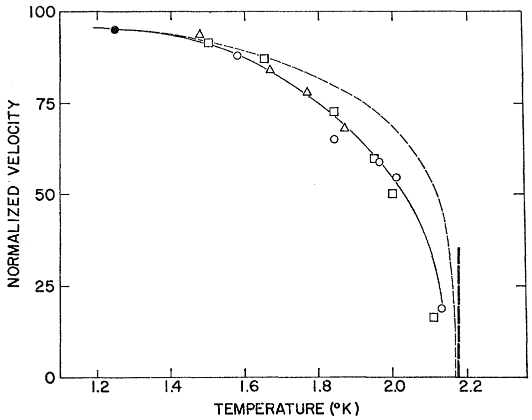


FIG. 7. Normalized third sound velocity versus temperature for different film heights. Film heights: ○, 0.44 cm; □, 12.3 cm; △, 13 cm ------, proportional to $(\rho_s/\rho)^{1/2}(1+TS/L)^{1/2}$.

Figure 7 shows normalized curves of the variation of velocity with temperature; together with the theoretical values obtained from Eq. (2), assuming that ρ_s/ρ for the film has the same value as that found in standard experiments on the bulk liquid. Identical results were obtained at all film thicknesses, and the velocity of third sound vanished at the λ point as predicted. However, once again a significant discrepancy exists between the theoretical and experimental curves. The discrepancy could conceivably be attributed to the film not being at equilibrium thickness, perhaps as a result of the steady heat input from the exciting source. However, effects of this type would probably make the film thickness decrease with increasing temperature, giving a higher velocity than the theory, whereas the opposite effect was observed. Moreover, the same curve was obtained in both heating and cooling experiments, which suggests that temperature differences between mirror and bath were negligible. Provisionally, therefore, we conclude that the discrepancy between experiment and theory is genuine.

Three possible explanations deserve consideration: (1) The ratio ρ_s/ρ in the film might differ from that in bulk liquid helium; (2) the restoring force f and film thickness d may vary with temperature; (3) there may be temperature-dependent terms in an exact theory of third sound, not included in Eq. (2). None of these explanations can be completely ruled out at present. The first seems somewhat unlikely, since Rudnick and Shapiro's latest experiments on fourth sound[8] show that in narrow channels ρ_s/ρ agrees to within 1% with the value found in bulk liquid. On the other hand, the film thickness in our experiments was an order of magnitude lower than the pore-diameter used by Rudnick and Shapiro. As for the second explanation, f and d certainly are affected by temperature, but the form of the dependence is not yet worked out, and Dzyaloshinskii *et al.*[5] content themselves with stating that the effects are "relatively small." Finally, the third explanation is quite possible in view of the deviations from the simple theory already discovered. It should be noticed that the small temperature dependent term in Eq. (1) above makes the velocity higher and so cannot account for the observed discrepancy.

Measurements were also made of attenuation coefficients. Contrary to what might be expected, the waves were much less strongly attenuated in thin films than thick ones. In fact, at film heights greater than about 10 cm the attenuation was so small that a standing wave was set up, probably owing to reflection from the end of the mirror. The existence of standing waves was established by the fact that the position of the nodes was independent of the phase-setting, in contrast to the traveling wave illustrated in Fig. 3 above.

Spreading of the third-sound beam may have produced a spurious contribution to the attenuation. Even though a strip source extending right across the mirror was used, the waves may have spread over the edge on to the underside of the mirror. However, in a subsidiary experiment in which third sound was excited on a mirror with a step 0.3 cm high, it was found that the wave did not propagate beyond this step. This suggests that the waves were not able to spread over the edges or ends of the ordinary mirror. In any case, diffraction effects probably produced a highly directional beam at the shorter wavelengths. Even for the longest wavelengths (about 0.5 cm) it is unlikely that spreading made a contribution greater than 0.3 cm^{-1} to the attenuation. Since attenuations several times this value were observed, it seems that we were definitely observing attenuations due to mechanisms other than spreading.

Figure 8 shows some of the attenuation data. It suggests that the attenuation increases with increasing frequency, but it also demonstrates a lack of reproducibility which precludes decisive conclusions at this stage in the investigation. Other experiments showed that the attenuation increased with temperature, becoming very

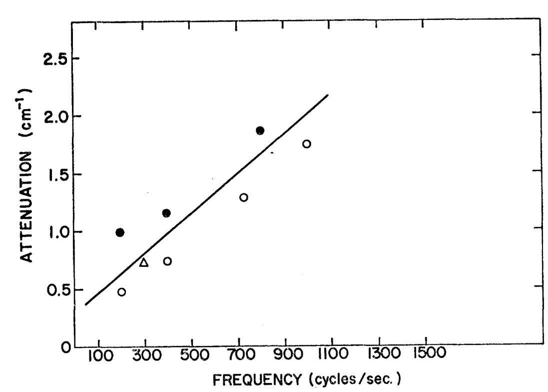


FIG. 8. Attenuation versus frequency at a constant film height of 9 cm. ○, △, and ● represent three different experiments performed on three different days.

[8] I. Rudnick and K. A. Shapiro (private communication).

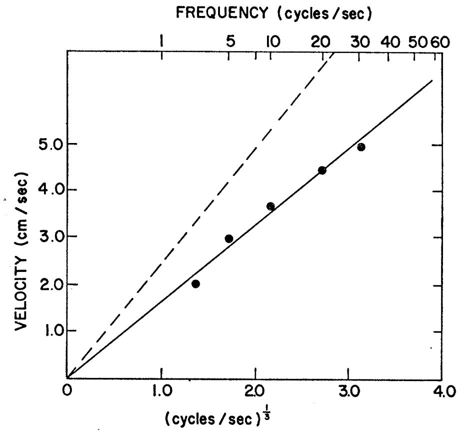


FIG. 9. Velocity of surface ripples on bulk liquid as a function of the cube root of the frequency. ——, $(2\pi\sigma\nu/\rho)^{1/3}$, the velocity of a capillary wave on a normal liquid.

large in the neighborhood of the λ point. All of these results involve considerable errors, but they do appear to indicate the existence of a real attenuation much larger than anything predicted in the original theory. One possible explanation is that the attenuation is due to residual motions of the viscous normal fluid, arising perhaps from pressure differences between the peaks and troughs of the waves. Since the motion of the normal component is probably less prominent in thinner films, this hypothesis would account for the observed decrease of attenuation with decreasing film thickness. It would also explain the rapid increase in attenuation near the λ point. However, preliminary calculations by Grobman raise some doubt whether the motions of the normal component are large enough to account for the observed attenuations.

The propagation of third sound in moving films has also been investigated, the film being set in motion by small heaters at either end of the mirror. The velocity of the waves in the laboratory frame of reference was shown to increase when the flow was in the direction of propagation and to decrease when the flow was in the opposite direction. It should be possible to obtain direct estimates of the critical velocity of flow in this way. Preliminary measurements indicate that the critical velocity is approximately half the third sound velocity, but the results must be viewed with reserve, since the film thickness was affected by the heaters.

Measurements were also made on the propagation of ripples across a puddle of liquid helium 0.1 cm deep. Presumably the ripples are surface-tension waves.[9] Figure 9 shows that the velocity was proportional to the cube-root of the frequency as expected. The attenuation was negligible. The slope of the experimental curve in Fig. 9 is 1.7 cm/(sec)$^{2/3}$. According to theory, the slope should be equal to $(2\pi\sigma/\rho)^{1/3}$, where σ is the surface tension, and this quantity has the value 2.5 cm/(sec)$^{2/3}$. Thus the theory is only approximately confirmed. Perhaps an exact theory of surface ripples on liquid helium II must take into account the two-fluid theory and the thermomechanical effect.

4. CONCLUSIONS

Broadly speaking, the experiments establish the existence of third sound in agreement with the theory. However, there are a number of significant deviations from the simple theory, indicating that Eqs. (1) and (2) are only approximately correct. In particular the velocity of third sound in thick films deviates considerably from the predicted value, and so does the variation of velocity with temperature. In addition, the attenuation coefficients, although not accurately determined in our experiments, are much larger than expected. It is conceivable that the assumptions made in deriving the simple theory (i.e., that the normal component remains fixed and the superfluid velocity is independent of depth) are both oversimplifications. The variation of velocity with temperature may indicate changes in film thickness and restoring force of the type suggested by Dzyaloshinskii *et al.*

The measurements on surface tension waves are in fair agreement with theory, but indicate that the classical theory may require some modification when applied to liquid helium II.

ACKNOWLEDGMENTS

We are indebted to Dr. L. Grimes for some valuable preliminary work on this experiment. We also thank K. Pickar, W. Grobman, and S. Coren for assistance with measurements and calculations.

[9] John William Strutt, Baron Rayleigh, *The Theory of Sound* (Macmillan and Company, Ltd., London, 1929), Vol. II, p. 343.