

Above the λ temperature no discontinuities in the mobility have been found until now for drift velocity ranging between 20 and 100 cm/sec. For sake of comparison we report in Fig. 2 typical results of the measurements of the mobility taken below and above the λ point.

The behavior of the critical velocity, $\langle v_c \rangle$, above 1.4°K, together with the apparent lack of discontinuities above the λ point, leads us to believe that the phenomenon in question is peculiar to liquid helium II only.

The authors are deeply grateful to Professor Giorgio Careri for his encouragement.

*Work supported by Consiglio Nazionale delle Ricerche.

¹G. Careri, S. Cunsolo, and P. Mazzoldi, Phys. Rev.

Letters **7**, 151 (1961); Phys. Rev. **136**, A303 (1964).

²G. Careri, S. Cunsolo, and M. Vicentini-Missoni, Phys. Rev. **136**, A311 (1964).

³J. A. Cope and P. W. F. Gribbon, in Proceedings of the Ninth International Conference on Low-Temperature Physics, Columbus, Ohio, 1964 (Plenum Press, New York, 1965), p. 153.

⁴J. A. Cope and P. W. F. Gribbon, Phys. Letters **16**, 128 (1965).

⁵K. Huang and A. C. Olinto, Phys. Rev. **139**, A1441 (1965).

⁶C. di Castro, Nuovo Cimento **42**, 251 (1966).

⁷J. T. Tough, W. D. McCormick and J. G. Dash, Phys. Rev. **140**, A1524 (1965).

⁸S. Cunsolo and P. Mazzoldi, Nuovo Cimento **20**, 949 (1961).

⁹R. P. Feynman, Phys. Rev. **94**, 262 (1954); J. L. Yarnell, G. P. Arnold, P. J. Bendt, and E. C. Kerr, Phys. Rev. **113**, 1379 (1959); D. G. Henshaw and A. D. B. Woods, Phys. Rev. **121**, 1266 (1961).

BRILLOUIN SCATTERING IN LIQUID HELIUM II

Michael A. Woolf,* P. M. Platzman, and M. G. Cohen

Bell Telephone Laboratories, Murray Hill, New Jersey

(Received 6 July 1966)

Using the technique of Brillouin scattering, we have measured the attenuation and velocity of high-frequency (556 and 723 Mc/sec) acoustic phonons (first sound) in liquid helium below the lambda point. Previous workers,^{1,2} using conventional ultrasonic pulse techniques, have measured the temperature and frequency dependence of the attenuation $[\alpha(T, \Omega)]$ to temperatures as low as 0.1°K and recently³ to frequencies as high as 150 Mc/sec. A peak in α near 1°K broadly defines two regimes: the hydrodynamic regime on the high-temperature side of the peak up to $T \approx 1.9^\circ\text{K}$, and the collisionless regime on the low-temperature side. In the hydrodynamic regime, α is proportional to Ω^2 .⁴ The temperature dependence of α is determined by certain collision processes⁵ which can be roughly characterized by a temperature-dependent relaxation time, $\tau(T)$. The peak in $\alpha(T)$ is predicted⁵ to occur at a temperature, $T_p(\Omega)$, where $\Omega\tau(T_p) \approx 1$. In the collisionless regime $[\Omega\tau(T) > 1]$, α is postulated to arise from three-phonon collisions which depend indirectly on $\tau(T)$. The theories of this process⁶⁻⁹ give $\alpha \sim \Omega T^4$ from temperatures near $T_p(\Omega)$ to $T \approx 0.1^\circ\text{K}$. In the region $0.2^\circ\text{K} < T < 0.5^\circ\text{K}$, Abraham et al.³ find $\alpha \sim T^4$, while Jeffers and Whitney² find $\alpha \sim \Omega^{3/2} T^3$. In addition, the data

at 30 Mc/sec³ seem to show a less rapid variation than T^4 near $T_p(\Omega)$. The situation in this regime is therefore somewhat uncertain.

The Brillouin scattering technique allows one to measure, at high frequencies, a much larger attenuation than is possible with conventional ultrasonic methods. We therefore felt that, aside from the intrinsic interest in a light-scattering experiment, such a measurement would provide additional information on the behavior of sound in liquid helium.

In Brillouin scattering, the incident photon (frequency ω_i , wave vector \vec{k}_i) interacts with a longitudinal phonon (Ω, \vec{K}_0) and is scattered through some angle 2Θ . Since the total energy and momentum are conserved in the collision process, the scattered photon (ω_s, \vec{k}_s) has $\omega_s = \omega_i + \Omega$ and $\vec{k}_s = \vec{k}_i + \vec{K}_0$. Since $\Omega \ll \omega_i$, $k_i \cong k_s \equiv k$ and therefore,

$$\Omega/v \equiv K_0 = 2nk \sin\Theta, \quad (1)$$

where Θ is the Bragg angle, v the velocity of sound, and n the index of refraction of the medium. In a simple wave picture, the incident light is diffracted and Doppler shifted by the moving grating set up by the periodic density fluctuations of the sound wave. In the classic Brillouin-scattering experiment,¹⁰ the incident

light is scattered by thermally generated phonons. Analysis of the frequency shift and the width of the lines in the Brillouin doublet requires extremely high-resolution spectroscopy, especially in the case of liquid helium where v is small (238 m/sec).

We have used a new technique, developed by Gordon and Cohen,¹¹ for observing Brillouin-scattered light. High resolution and the use of a multimode laser light source is made possible by beating the light scattered from injected acoustic phonons with the unshifted laser beam.¹² Phonons of a well-defined frequency, Ω , are generated by a thin CdS film evaporated onto the back face of a quartz rod. The sound enters the liquid helium in a direction normal to the optically polished front face of the rod. A laser beam is incident on this face at an angle of $\frac{1}{2}\pi - \Theta$ to the normal, where Θ is the Bragg angle appropriate to Ω . Light scattered by the sound is collinear with that part of the incident beam specularly reflected from the transducer face. The frequency-shifted scattered light and the unshifted reflected light beams¹³ are then focused onto a square-law photodetector, where the beat note between them gives rise to a microwave photocurrent, $i(\Omega)$, at the sound frequency.¹⁴

Finite attenuation in the medium introduces a Lorentzian spread in the amplitude of the wave vectors, \vec{K} , for sound of a fixed frequency. We trace out this distribution by rotating the transducer through a small angle $\Delta\theta \equiv \theta - \Theta$,

while keeping the direction of \vec{k}_i fixed. Light is scattered by phonons of wave vector $\vec{K} = \vec{K}_0 + \Delta\vec{K}$, where $\Delta K = 2nk\Delta\theta \cos\Theta$. The reflected and the scattered beams remain collinear as the transducer is rotated. Since the magnitude of the beat note is proportional to the number of phonons of wave vector \vec{K} ,

$$i(\Omega) \sim \frac{\alpha + i\Delta}{\alpha^2 + \Delta^2}, \quad (2)$$

where $\Delta \equiv 2nk\Delta\theta$.¹¹ If $(\Delta\theta)_h$ corresponds to the half width of the distribution, the attenuation $\alpha = 2nk(\Delta\theta)_h \cos\Theta$.

The signal from the photodetector (a planar silicon diode) is homodyne detected as shown in Fig. 1. The incident laser beam is chopped at 1000 cps. A portion of the cw microwave driving power to the transducer is injected with adjustable phase, φ_* , into the signal line as the homodyne reference.¹⁵ The chopped output voltage of the superheterodyne receiver is¹¹

$$V \sim \text{Re} \left(\frac{\alpha + i\Delta}{\alpha^2 + \Delta^2} e^{i\varphi} \right) = \frac{\alpha \cos\varphi - \Delta \sin\varphi}{\alpha^2 + \Delta^2}. \quad (3)$$

V is plotted as a function of Δ , and the resulting curve is computer fitted to a function of the form given by (3) in order to find the value of α . The velocity of the sound is calculated from (1).

Data shown in Fig. 2 were taken at 556 and 723 Mc/sec, corresponding to incident light wavelengths of 6328 Å (helium-neon laser) and 4880 Å (argon-ion laser), respectively ($\Theta \approx \frac{1}{4}\pi$). With an incident light power at either wavelength of about 25 mW and with a microwave input power¹⁶ of 50 mW to the transducer, the signal-to-noise ratio was about ten. The helium used in these experiments was transferred directly from a storage Dewar. In a given run, data were taken with a single helium fill and showed reasonable consistency at different temperatures.¹⁷ The remaining scatter (indicated by the bars in Fig. 2) is due, at least in part, to some inevitable coupling between the mechanical rotation and both the microwave transducer drive and the optics. This scatter is generally greater than the estimated errors in the measurement of Θ ($\pm 1\%$) and $\Delta\theta$ ($\pm 2\%$). For some runs, the points were consistently higher or lower than those shown in Fig. 2 (by as much as 10%). The amount of impurities (N_2 and O_2 particles) in the helium could have varied widely from run to run, possibly affecting phonon relaxation times and contributing to this variation in the

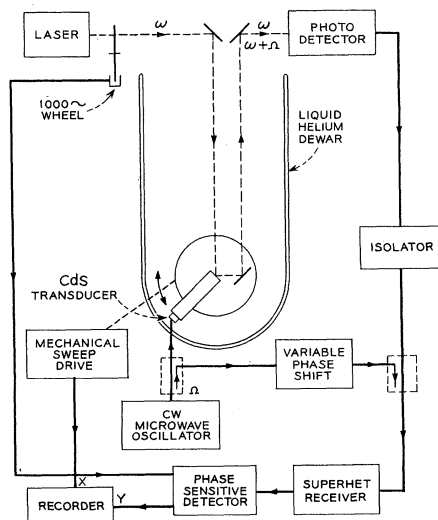


FIG. 1. Experimental arrangement. The mechanical and electrical connections to the transducer were brought in from the top of the Dewar.

data. Our measurements of the velocity agree with the earlier low-frequency results⁴ to within experimental error (about $\pm 1\%$).

The solid curves in Fig. 2 are the high-temperature 30-Mc/sec data of Abraham *et al.*,³ scaled up by Ω^2 to our frequencies. As predicted by the hydrodynamic theory,⁵ the attenuation scales quite well with frequency in the range 1.6 to 1.9°K. More surprising, perhaps, is the fact that the minimum at $T \approx 2^\circ\text{K}$ and the rise near the lambda point seem also to scale as Ω^2 . The frequency dependence of α below the peak cannot be determined by comparison with earlier results since the low-frequency data are in the peak region near 1°K.

The dashed curves were calculated¹⁸ from

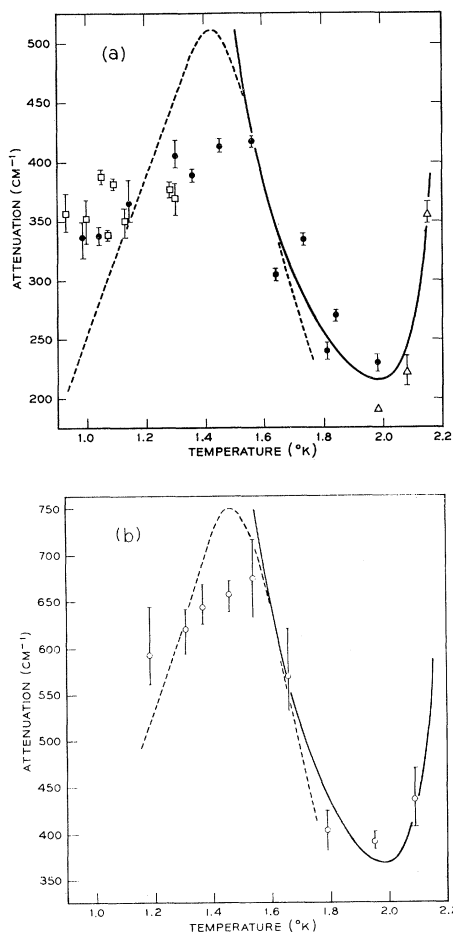


FIG. 2. Attenuation of first sound in liquid helium. (a) $\Omega = 556$ Mc/sec. Triangle represents run 1; solid circles represent run 2; open squares represent run 3. The bars represent the scatter at each point for the run shown. (b) $\Omega = 723$ Mc/sec. Solid curves: 30-Mc/sec data of Abraham *et al.*,³ scaled up by Ω^2 ; dashed curves: attenuation computed from the theory of Khalatnikov and Chernikova.¹⁹

the recent theory of Khalatnikov and Chernikova.¹⁹ This theory gives a single analytic expression for $\alpha(T, \Omega)$ (with one adjustable parameter²⁰) for temperatures including the region of the peak. Since the theory does not include the effect of the lambda point on the attenuation, divergence from the data above $T \approx 1.9^\circ\text{K}$ is expected. More significant is the disagreement below $T \approx 1.4^\circ\text{K}$. Our results indicate that α decreases less rapidly than T^4 from 1.2 to 0.95°K . The quantitative analysis of Khalatnikov and Chernikova¹⁹ would predict that at our frequencies $\alpha \sim T^4$ in this temperature range. (It is certainly possible that the attenuation becomes proportional to T^4 at still lower temperatures.) Also evident in our data is a marked rounding off of the peak. There is similar disagreement in the data at 30 Mc/sec,^{3,18} although at 6 Mc/sec agreement with the theory seems quite good.⁷ This trend may indicate that some modification of the theory is necessary at high frequencies.

Further experiments in pure helium and at lower temperatures should help clarify some of these details.

The authors wish to thank P. C. Hohenberg for his constant interest and stimulation. Special thanks are due to E. I. Gordon who made this experiment possible by sharing his original ideas with us. The technical assistance of P. W. Gundy was much appreciated.

*Present address: Department of Physics, University of California, Los Angeles, California.

¹C. E. Chase, Proc. Roy. Soc. (London) **A220**, 116 (1953); C. E. Chase and M. A. Herlin, Phys. Rev. **97**, 1447 (1955); K. Dransfeld, J. A. Newell and J. Wilks, Proc. Roy. Soc. (London) **A243**, 500 (1958).

²W. A. Jeffers and W. M. Whitney, Phys. Rev. **139**, A1082 (1965).

³B. M. Abraham, Y. Eckstein, J. B. Ketterson, and J. H. Vignos, Phys. Rev. Letters **16**, 1039 (1966).

⁴K. R. Atkins, *Liquid Helium* (Cambridge University Press, Cambridge, England, 1959).

⁵I. M. Khalatnikov, Zh. Eksperim. i Teor. Fiz. **20**, 243 (1950).

⁶K. Dransfeld, Phys. Rev. **127**, 17 (1962).

⁷I. M. Khalatnikov and D. M. Chernikova, Zh. Eksperim. i Teor. Fiz. **2**, 566 (1965) [translation: Soviet Phys.-JETP **2**, 351 (1965)].

⁸P. C. Kwok, P. C. Martin, and P. B. Miller, Solid State Commun. **3**, 181 (1965).

⁹C. J. Pethick and D. ter Haar, to be published.

¹⁰G. Benedek and T. Greytak, Proc. IEEE **53**, 1623 (1965).

¹¹E. I. Gordon and M. G. Cohen, to be published; M. G. Cohen and E. I. Gordon, IEEE J. Quantum Electron.

QE-2, 8A-8 (1966); Bell System Tech. J. **44**, 693 (1965).

¹²Each narrow mode of the laser beats with its own scattered light to give a net signal.

¹³Phase modulation of the reflected beam by the moving face of the transducer introduces sidebands at $\omega_i \pm \Omega$, whose beat notes with the carrier at ω_i exactly cancel. The reflected light is therefore effectively unshifted.

¹⁴Light scattered out of the reflected beam is downshifted and gives rise to a beat note out of phase with $i(\Omega)$. Total cancellation of the signal is avoided by making the reflectivity of the transducer face less than 1.0.

¹⁵M. G. Cohen and E. I. Gordon, Bell System Tech. J. **42**, 3068 (1964).

¹⁶The input power was varied from 20 to 100 mW with no observable change in the attenuation at 1.2°K.

¹⁷The temperature of the bath was measured by its vapor pressure and with a carbon resistor placed about 2 cm from the transducer. These two measurements always agreed to within a few mdeg independently of the power in the transducer.

¹⁸The theoretical analysis is based on recent unpublished work of P. C. Hohenberg and P. M. Platzman. We are indebted to P. C. Hohenberg for bringing to our attention the possibility of applying the theory of Khalatnikov and Chernikova (Ref. 19) to our data.

¹⁹I. M. Khalatnikov and D. M. Chernikova, Zh. Eksperim. i Teor. Fiz. **49**, 1957 (1965); **50**, 411 (1966).

²⁰The effect of this parameter Λ on α is significant only at high temperatures. We find that $\Lambda = (2.0 \pm 0.2) \times 10^{43}$ gives the best fit to our data in the hydrodynamic region. This value is more accurate than Khalatnikov's estimate of $\Lambda = 3.4 \times 10^{43}$ which was based on earlier low-frequency attenuation data.

IDENTIFICATION OF A NEW SPECTRAL COMPONENT IN BRILLOUIN SCATTERING OF LIQUIDS*

W. S. Gornall, G. I. A. Stegeman, B. P. Stoicheff, R. H. Stolen,[†] and V. Volterra[‡]

Department of Physics, University of Toronto, Toronto, Canada

(Received 11 July 1966)

An intense broad background has been observed in the Brillouin spectrum of liquid CCl₄ and is shown to be in agreement with a theory by Mountain which includes thermal relaxation.

Recent refinements in experimental techniques have led to renewed interest in Brillouin spectroscopy and to suggestions by Griffin,¹ Mountain,² and Gillis and Puff³ that various new sound-wave modes might be detectable by such light-scattering experiments. We wish to report the observation of a new feature in the Brillouin spectrum of liquid CCl₄ which originates from density fluctuations caused by a nonpropagating sound-wave mode predicted by Mountain.²

In Fig. 1(a) are shown the Brillouin spectra of liquid CCl₄ observed at scattering angles of 44, 97, and 155°. We draw attention to the prominent continuous background between the central unshifted component and the Doppler-shifted Brillouin component, in all of these spectra. This background extends symmetrically on either side of the central component to the Brillouin components, producing an asymmetry in their intensity profiles, and then decreases in intensity on the high-frequency sides of the Brillouin components. It is polarized in the same plane as the exciting laser light, and its intensity and breadth are independent of temperature in the range 15 to 75°C. From Fig. 1 it is evident that this background accounts for an appreciable part (approximately 20%)

of the total intensity in these spectra. Thus, it represents a significant new component in the Brillouin spectrum of CCl₄.

The experimental arrangement is essentially that used by Chiao and Stoicheff.⁴ A He-Ne laser of 2 m length and 5-mW output power at $\lambda 6328$ was used as the exciting source. Provisions were made for keeping the liquid sample at a constant temperature of $20.0 \pm 0.2^\circ\text{C}$ and for varying the scattering angle from 44 to 155°. The light scattered at a fixed angle was analyzed with a pressure-scanned Fabry-Perot interferometer having a free spectral range of 15 000 Mc/sec (or 0.5 cm^{-1}). The combined laser and instrumental width was approximately 530 Mc/sec.

With this apparatus, we have made extensive measurements on the velocity of longitudinal hypersonic waves in CCl₄ (at 20.0°C) in the frequency range 1.6 to 4.7 Gc/sec, and a preliminary report has already been presented.⁵ Our measurements reveal a characteristic thermal relaxation process in this frequency range. Moreover, the observed velocity dispersion agrees with calculations based on a single relaxation theory for the total vibrational specific heat. Consequently, it was possible to eval-