MODES OF SOUND PROPAGATION IN DILUTE SOLUTIONS OF He³ IN LIQUID He⁴ †

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The attenuation of sound in a dilute solution of He³ in He⁴ has recently been measured.¹ At temperatures above 0.3° K, the form of the attenuation is very similar to that of pure He⁴, except that the phonon relaxation time is changed by the presence of the He³. At temperatures below 0.3° K the phonon contribution to the attenuation (which varies as T^4) becomes negligible, but an added peak is observed in the attenuation. This peak is apparently due to the absorption of the sound by He³. It is the purpose of this Letter to investigate the modes of sound propagation in dilute solutions of He³ in He⁴ at such low temperatures that the presence of phonons may be neglected.

The propagation of sound will be studied by using the kinetic equation method.² The temperature is assumed to be so low that the only excitations present are the He³ quasiparticles which form a degenerate Fermi liquid.

The excitation energies $\epsilon(p)$ are assumed to have the form

$$\epsilon(p) = \epsilon_0(p) + \frac{p^2}{2m^*} + \vec{p} \cdot \vec{v}_s$$
$$+ \frac{2}{3} \frac{\epsilon_F}{N_0} \int \left(F_0 + \frac{F_1}{2m^* \epsilon_F} \vec{p} \cdot \vec{p}' \right) \delta n(\vec{p}') d^3 p', \qquad (1)$$

where \vec{p} is the He³ momentum relative to the medium at rest. The first term $\epsilon_0(\rho)$ is the binding energy of the He³ in He⁴, which is, of course, dependent upon the density of fluid ρ . The second term is the kinetic energy, where m^* is the effective mass. The third term gives the energy shift required by Galilean invariance when the fluid is moving with velocity \vec{v}_s . The last term is due to the interactions between He³ atoms: F_0 and F_1 are the parameters defined by Landau,³ δn the change in the distribution function from equilibrium, $\epsilon_{\rm F}$ the Fermi energy, and N_0 the equilibrium density of He³ atoms. If the local density of He³ is given by $N(\mathbf{\vec{r}}, t) = N_0 + \delta N(\mathbf{\vec{r}}, t)$, then the last term in (1) can be written as

$$\frac{2}{3} \frac{\epsilon_{\rm F}}{N_0} \left(F_0 \delta N + \frac{F_1}{2m^* \epsilon_{\rm F}} \vec{\rm p} \cdot \vec{\rm J} \right), \qquad (2)$$

where $\mathbf{J} = \int \mathbf{p} n d^3 p$ is the total excitation momentum. The distribution function $n(\mathbf{p}, \mathbf{r}, t)$ is found by solving the linearized Boltzmann equation of Khalatnikov and Abrikosov,⁴ where ϵ is given by (1). Let $\delta \rho = \rho - \rho_0$, where ρ_0 is the equilibrium density. Then, if it is assumed that $\delta \rho$, v_s , and δn vary, like the sound wave, as $\exp[i(\mathbf{q}\cdot\mathbf{r}-\omega t)]$, one may find δn as a function of ρ and \mathbf{v}_s . Two additional equations are then required to find these quantities. One is the equation of continuity of mass:

$$\partial \rho / \partial t + \nabla \cdot \left[\rho \vec{\mathbf{v}}_{o} + \vec{\mathbf{J}} \right] = 0.$$
 (3)

This has the same form as for phonons in $\text{He}^{4,2}$ The other equation which is necessary may be derived in a manner analogous to the derivation for phonons in He^4 by de Boer.⁵ The linearized equation is given by

$$\rho \frac{\partial \vec{\mathbf{v}}_{S}}{\partial t} = -\nabla P_{0} - \rho \nabla \int \frac{\partial \epsilon}{\partial \rho} n d^{3} \rho - N \nabla \int \frac{\partial \epsilon}{\partial N} n d^{3} \rho, \quad (4)$$

where P_0 is the partial pressure of the He⁴, and $\partial \epsilon / \partial N$ is found by using expression (2) in Eq. (1). Equations (3) and (4) may also be solved by using the assumption that $\delta \rho$, \vec{v}_S , δN , and \vec{J} vary as $\exp[i(\vec{q} \cdot \vec{r} - \omega t)]$. The result is

$$\frac{\delta \rho_s}{\rho_s} = \frac{\vec{q} \cdot \vec{v}_s}{\omega} = \frac{m_0^* \delta N}{\rho_s} \frac{\overline{s}^2 (1 + \Lambda')}{s^2 - \overline{s}^2}, \quad (5)$$

where $\rho_s = \rho - N_0 m_0^*$ is the superfluid density; $m_0^* = m^*/(1 + \frac{1}{3}F_1)$; $s = \omega/qv_F$ where v_F is the Fermi velocity; $\overline{s} = (c_0/v_F)(1 - N_0 m_0^*/\rho)^{1/2}$; and c_0 is the velocity of sound in He⁴, $c_0^2 = \partial P_0/\partial \rho$; also

$$\Lambda' = \left[\frac{\rho_0}{m * c_0^2} \frac{\partial \epsilon}{\partial \rho} + \frac{1}{3} \left(\frac{v_F}{c_0}\right)^2 F_0\right] \left[1 + \frac{1}{3}F_1\right]$$
$$= \Lambda + \frac{1}{3} \left(\frac{v_F}{c_0}\right)^2 F_0 \left(1 + \frac{1}{3}F_1\right).$$

In the derivation of (5) the equation of continuity of number of He³ quasiparticles was used in order to relate \mathbf{J} to δN . When (5) is inserted into the expression for δN derived from the Boltzman equation, the following compatibility equation is derived:

$$\frac{-i\omega\tau}{1-i\omega\tau} = w\left(s\left[1+\frac{i}{\omega\tau}\right]\right)\left\{\frac{1}{1-i\omega\tau}+\frac{3is^2}{\omega\tau}+F_0+\frac{F_1s^2}{1+\frac{1}{3}F_1}+\frac{3N_0m_0^*}{\rho_0(1+\frac{1}{3}F_1)}\left(\frac{c_0}{v_F}\right)^2\left[\frac{(1+\Lambda)(1+\Lambda')\overline{s}^2}{s^2-\overline{s}^2}+(\Lambda+\Lambda'+1)\right]\right\}, \quad (6)$$

where τ is the He³ relaxation time. The function $w(\xi)^4$ is given by

$$w(\xi) = \frac{\xi}{2} \ln \frac{\xi + 1}{\xi - 1} - 1.$$
 (7)

Equation (6) is a transcendental equation whose solutions for complex s give the velocity and attenuation of sound. When the He³-He⁴ interaction vanishes, the term in $N_0m_0^*/\rho_0$ drops out and the result is identical with that of Ref. 4 for pure He³.

Equation (6) is generally very difficult to solve. However, for $|\xi| \gg 1$, it is possible to expand $w(\xi)$ in a power series as in Ref. 4, and then Eq. (6) becomes a quadratic equation in s^2 . One solution of this equation gives the following results for the sound velocity c and the attenuation α :

$$c^{2} = c_{0}^{2} [1 + (N_{0}m_{0}^{*}/\rho_{0})(\Lambda + \Lambda' + \Lambda\Lambda')], \qquad (8)$$

$$\alpha = \frac{2}{15} \left(\frac{v_{\rm F}}{c_{\rm o}} \right)^2 \frac{N_0 m^*}{\rho_0 c} (1 + \Lambda) (1 + \Lambda') \frac{\omega^2 \tau}{1 + (\omega \tau)^2}.$$
 (9)

For this solution $s^2 \approx 80$, and, therefore, the expansion for large ξ and $w(\xi)$ was valid; an error of about 2% is made by dropping terms

of order $1/s^2$ with respect to 1. In (8) and (9) only terms linear in the concentration have been retained.

The velocity of sound for this mode is very close to that in pure He⁴, so that this must be the mode observed by Abraham et al.¹ To the order of approximation used, the velocity is independent of temperature, in agreement with their observation of a velocity change of less than 1% as $\omega \tau$ varied from $\ll 1$ to ~ 10 . The attenuation is given by a relaxation-type formula, in agreement with the experimental results, as discussed in Ref. 1. The value of α/ω at peak attenuation is independent of frequency. The calculated value of $(\alpha/\omega)_{\text{peak}}$ from (9) is $\approx 0.37(1 + \Lambda)(1 + \Lambda') \times 10^{-8} \text{ cm}^{-1}$ sec, whereas the experimental results for frequencies 20, 60, and 100 MHz are 0.65, 0.70, and 0.64 $\times 10^{-8}$ cm⁻¹ sec, respectively. If the average of these values is compared with the theoretical values, and F_0 is neglected in Λ , one obtains $\Lambda = \Lambda' = 0.34$.

The second solution of the quadratic equation gives $\operatorname{Res}^2 \approx \frac{1}{3}$. Therefore, for this solution the expansion of $w(\xi)$ for large $|\xi|$ is not valid unless $\omega \tau \ll 1$. For this case the second solution is

$$c^{2} = v_{\mathbf{F}}^{2} \left[\frac{1}{3} (1 + \frac{1}{3}F_{1})(1 + F_{0}) - \frac{N_{0}m^{*}}{\rho_{0}} \left\{ \frac{1}{3} (1 + F_{0})(1 + \Lambda)(1 + \Lambda') + \frac{\Lambda\Lambda'}{1 + \frac{1}{3}F_{1}} \left(\frac{c_{0}}{v_{\mathbf{F}}} \right)^{2} \right\} \right], \tag{10}$$

$$\alpha = \frac{2}{15c} \omega^2 \tau \left(\frac{v_{\rm F}}{c}\right)^2 \left[1 + \frac{1}{3}F_1 - \frac{N_0 m^*}{\rho_0} (1 + \Lambda)(1 + \Lambda')\right].$$
(11)

If the He³-He⁴ interaction vanishes, then the terms in $N_0 m^*/\rho_0$ do not appear, and the results (10) and (11) become identical with the results for the hydrodynamic mode of pure He^{3.4} The condition for the existence of a He³-like zero-sound mode may be investigated by examining (6) for 0°K ($\omega \tau = \infty$) as done by Landau³ for pure He³. The necessary condition for a zero-sound mode to exist is that

$$A = F_{0} + \frac{F_{1}}{1 + \frac{1}{3}F_{1}} + \frac{3N_{0}m_{0}^{*}}{\rho_{0}(1 + \frac{1}{3}F_{1})} \left(\frac{c_{0}}{v_{F}}\right)^{2} \left[\frac{(1 + \Lambda)(1 + \Lambda')c_{0}^{2}}{v_{F}^{2} - c_{0}^{2}} + (\Lambda + \Lambda' + 1)\right] > 0.$$

Since the third term is negative, the He³-He⁴ interaction makes the condition upon F_0 and F_1 even more stringent to fulfill than in the pure He³ case. If this mode exists, it is easy to show that the attenuation will vary as $1/\tau$, and at 0°K, $c \approx v_F \{1 + 2 \exp[-2(1 + 1/A)]\}$.

We conclude with a physical description of the modes of oscillation. If the velocity of sound (8) is inserted in (5), we find that $\delta \rho_S / \rho_S \approx (1$ $(\delta N/N_0)$. Thus the relative bunching of the He³ is of the same order of magnitude as the relative density changes in the He⁴: The superfluid and the normal fluid respond in roughly the same way to the sound. On the other hand, if the velocity of the He³-like modes is inserted into (5), s^2 may be dropped in comparison with \overline{s}^2 , and we find that $\delta \rho_S / \rho_S \approx -(1 + \Lambda') (N_0 m_0 * / N_0 * /$ $\rho_{\rm s}$)($\delta N/N_0$). That is, the relative density changes in the superfluid are quite negligible in comparison with the He³ bunching. This is reasonable, for we would expect that the He³-like modes would be propagated mainly by density changes in the He³ and not by the superfluid. However, because of the much larger density of He³, the absolute value of the density change $\delta \rho_{\rm S}$ is the same order of magnitude as $m_{\rm 0} * \delta N$ = $\delta \rho_n$,⁶ the normal density change. Actually, the He³-like modes are rather similar to second sound in pure He⁴. This is because the density changes in the normal fluid are of the same order of magnitude, and of opposite phase to the density changes in the superfluid: $\delta \rho_s$ $+ \delta \rho_n \approx -\Lambda' \delta \rho_n.$

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¹B. M. Abraham, Y. Eckstein, J. Ketterson, and J. Vignos, Phys. Rev. Letters, preceding paper [Phys. Rev. Letters 17, 1254 (1966)].

²I. M. Khalatnikov, <u>Introduction to the Theory of Su-</u> <u>perfluidity</u> (W. A. Benjamin, Inc., New York, 1965), Chap. 18. Many other references are listed in this book.

³L. D. Landau, Zh. Eksperim. i Teor. Fiz. <u>32</u>, 59 (1957) [translation: Soviet Phys.-JETP <u>5</u>, 101 (1957)].

⁴I. M. Khalatnikov and A. A. Abrikosov, Zh. Eksperim. i Teor. Fiz. <u>33</u>, 110 (1957) [translation: Soviet Phys.-JETP <u>6</u>, <u>84</u> (1958)], Eqs. (1) and (2).

⁵J. de Boer, in <u>Liquid Helium</u>, edited by G. Careri (Academic Press, Inc., New York, 1963). See especially Sec. 3.

⁶The terms "normal fluid" and "superfluid" have been used very imprecisely here. In fact, the definitions for normal fluid and superfluid density given here agree with the usual definition (see Ref. 2, for example) only if interactions between the He³ quasiparticles are neglected. However, these are useful concepts for a <u>qualitative</u> understanding of the modes of propagation.

PRESSURE-INDUCED PHONON FREQUENCY SHIFTS IN LEAD MEASURED BY INELASTIC NEUTRON SCATTERING*

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Phonon frequency shifts due to the change of temperature have already been measured for several materials, in particular for lead.¹ These shifts reflect the deviation of the crystal potential from that assumed in the harmonic approximation. This deviation can also be explored by artificially changing the relative positions of the atoms in the crystals by application of external forces.

We have measured by the method of neutron spectrometry the frequency shifts of six selected phonons in lead due to the application of a hydrostatic pressure of 3000 atm. The measurements were done on a conventional triple-axis spectrometer in a constant-Q mode of operation.² The scattering chamber for 3000 atm and for a sample of 30 mm diameter and 65 mm length is described in detail elsewhere.³

The positions of the six selected phonons are shown in Fig. 1 as circles in the dispersion curves of lead⁴ for the directions $[\zeta 00]$ and $[\zeta \zeta \zeta]$. Figure 2 shows the results of a typical shift measurement. Because of the low phonon energy, the phonon peak is superimposed on a falling background due mainly to elastic scattering in the sample and in the wall of the pressure chamber. For the evaluation of the shift the center of a peak was defined by the intersection of two straight lines fitted to the flanks of the peak. The assumption was made that the shape of the phonon peak is not altered by the pressure. This assumption is not contradicted by any of our measurements. The influence of a falling background on the shift is eliminated by this evaluation method as long as the background is approximately linear.

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