$\mathcal{H}_a{}^{0}$[10]: $\exp[i(\mathcal{A}+\mathcal{B})t]\rightarrow\exp[i(\mathcal{A}-\mathcal{B})t]$, which is not equivalent to a time reversal since $\mathcal{A}$ is large. In the present experiments, the corresponding correction term $\bar{\mathcal{H}}_{\mathrm{DTR}}{}^{(2)}$ can be made as small as desired by a well-defined procedure. In this respect our experiments are closer to the inhomogeneous spin echo,[11] but succeed in reversing the dynamics of a system of interacting particles.

Our experiments show that the concepts of semiequilibrium and spin temperature in solids, while clearly of great value, must not be employed indiscriminately. We are of course only pointing out a special case of a general problem concerning criteria for irreversibility in isolated dynamical systems. We intend to discuss this matter more fully elsewhere, as well as extensions of the NMR experiment to repeated bursts and the problem of line narrowing in solids.

We thank J. D. Ellett, M. Gibby, and M. Mehring for their help.

*Supported in part by the National Institutes of Health.

†Dalton Predoctoral Fellow.

[1]For recent reviews see M. Goldman, *Spin-Temperature and Nuclear Magnetic Resonance in Solids* (Oxford Univ., New York, 1970); J. Jeener, *Advances in Magnetic Resonance* (Academic, New York, 1968), Vol. III; A. G. Redfield, Science 164, 1015 (1969).

[2]I. J. Lowe and R. E. Norberg, Phys. Rev. 107, 46 (1957).

[3]S. Hartmann and E. L. Hahn, Phys. Rev. 128, 2042 (1962).

[4]R. C. Tolman, *Principles of Statistical Mechanics* (Clarendon, Oxford, 1938), p. 152ff.

[5]A. G. Redfield, Phys. Rev. 98, 1787 (1955).

[6]U. Haeberlen and J. S. Waugh, Phys. Rev. 175, 453 (1968).

[7]W.-K. Rhim, thesis, University of North Carolina, 1969 (unpublished); H. Schneider and H. Schmiedel Phys. Lett. 30A, 298 (1969); W.-K. Rhim and H. Kessemeier, to be published.

[8]J. Jeener and P. Broekaert, Phys. Rev. 157, 232 (1967).

[9]J. G. Powles and P. Mansfield, Phys. Lett. 2, 58 (1962); J. G. Powles and J. H. Strange, Proc. Phys. Soc., London 82, 6 (1963).

[10]J. S. Waugh and L. M. Huber, J. Chem. Phys. 47, 1862 (1967).

[11]E. L. Hahn, Phys. Rev. 80, 580 (1950); H. Y. Carr and E. M. Purcell, Phys. Rev. 94, 630 (1954).

PHONON DISPERSION AND THE PROPAGATION OF SOUND IN LIQUID HELIUM-4 BELOW 0.6°K†

Humphrey J. Maris and Walter E. Massey
Department of Physics, Brown University, Providence, Rhode Island 02912
(Received 12 June 1970)

Recent experiments on the attenuation and velocity of sound in liquid He^4 at low temperatures are discussed in terms of the excitation model of liquid He^4. By assuming for the long-wavelength excitations the dispersion relation $\epsilon(p)=cp[1-\gamma p^2-\delta p^4\cdots]$ with γ *negative*, we are able to reconcile previous disagreements between theory and experiment.

In this Letter we make some general comments on the attenuation and velocity of sound in liquid helium-4 in the temperature range below 0.6°K where the only thermal excitations of importance are phonons. Despite considerable theoretical effort, the attenuation and velocity in this temperature range are not well understood. The theories of Pethick and ter Haar,[1] Kwok, Martin, and Miller,[2] Khalatnikov,[3] and Disatnik[4] give for the attenuation

$$\alpha=\frac{\pi^2}{30}\frac{(u+1)^2}{\rho\hbar^3}\frac{(kT)^4}{c^6}\omega\times[\arctan\omega\tau-\arctan(\tfrac{3}{2}\gamma\bar{p}^2\omega\tau)], \quad (1)$$

and for the change in velocity Δc,

$$\Delta c=\frac{\pi^2}{60}\frac{(u+1)^2}{\rho\hbar^3}\left(\frac{kT}{c}\right)^4\ln\frac{1+(\omega\tau)^2}{1+(\tfrac{3}{2}\gamma\bar{p}^2\omega\tau)^2}, \quad (2)$$

where u is the Grüneisen constant $(\rho/c)\partial c/\partial\rho$, ρ the density, k Boltzmann's constant, c the velocity of sound, ω the frequency of the sound wave, τ the thermal phonon lifetime, $\bar{p}=3kT/c$, and γ is defined by the energy-momentum relation for low-momentum phonons,

$$\epsilon(p)=cp[1-\gamma p^2-\delta p^4\cdots]. \quad (3)$$

In the derivation of Eqs. (1) and (2) it is assumed that the γp^2 term dominates over the δp^4 term for most of the thermal phonons with which the sound wave interacts. We note the following:

(a) The experimental attenuation[5-7] is larger than predicted by Eq. (1) when the known value of u is used.[8] There is uncertainty regarding the correct values of γ and τ. γ has generally been assumed to be positive and of the order of 10^{35}

to 10^{37} cgs units. The attenuation is a maximum when τ is such that $\omega\tau \gg 1 \gg \frac{3}{2}\gamma\bar{p}^2\omega\tau$. When τ satisfies these conditions, it follows that

$$\alpha = \frac{\pi^3}{60}\frac{(u+1)^2}{\rho\hbar^3}\frac{(kT)^4}{c^6}\omega. \quad (4)$$

Since all quantities in this expression are known, this constitutes an upper bound on the theory. However, this bound lies below the experimentally determined attenuation for much of the frequency-temperature range investigated by Abraham et al.[5,7] and Waters, Watmough, and Wilks.[6] The bound lies as much as a factor of 2 below the experimental results at 12 MHz and 0.25°K, and also at 36 MHz and 0.4°K.

(b) The data of Abraham et al.[7,9] indicate that the velocity of sound decreases with increasing frequency in the frequency range 12 to 84 MHz. This is in disagreement with Eq. (2).

We propose here that the generally assumed form of the energy-momentum relationship [i.e., Eq. (3) with $\gamma > 0$] is incorrect. A number of attempts[10] have been made to determine γ from neutron-scattering data. Recently, detailed measurements[11] at low p have given $\gamma = (0 \pm 2)\times 10^{36}$ g^{-2} cm^{-2} sec^2, and $\delta = (2.4 \pm 0.2)\times 10^{75}$ g^{-4} cm^{-4} sec^4. Our suggestion is that γ is negative and within the range of values implied by the neutron experiments.[12]

The assumption of a negative γ is not contradicted by any direct experimental evidence and does not appear to violate any fundamental theoretical concepts. On the contrary, the available evidence, if anything, tends to support this hypothesis. In addition to the neutron-scattering data, this hypothesis is consistent with the recent x-ray–scattering measurements of the static liquid structure function,[13] which is directly related to the long-wavelength excitations in liquid He^4.[14] As we show in this Letter, it offers a possible explanation of the velocity of sound and attenuation measurements in liquid He^4.

If γ is negative and the δp^4 term in Eq. (3) is ignored, the difficulty referred to above regarding the attenuation is removed because from Eq. (1) the upper bound of the attenuation is increased by a factor of 2. One can understand the experimental attenuation falling below the bound for certain ranges of frequencies and temperatures because (1) there will be some phonons for which the δp^4 term dominates over the γp^2 term. For these, the extra factor of 2 will not occur. (2) Even if the δp^4 term is ignored, there will be come phonons for which $\frac{3}{2}|\gamma|p^2\omega\tau$ is not $\gg 1$. If $\frac{3}{2}|\gamma|p^2\omega\tau \ll 1$ the factor of 2 is absent.

One, therefore, expects that if γ is negative, the attenuation will lie between that predicted by Eq. (4) and twice this value. This is in agreement with the results of Abraham et al.[5,7] and Waters, Watmough, and Wilks.[6]

It is not obvious how the assumption of negative γ will affect the temperature and frequency dependence of the velocity of sound. Here we wish to mention only some of the effects that must be taken into account in attempting such a calculation. The assumption that γ is negative changes generally accepted[15] ideas about phonon-phonon interactions in helium because momentum- and energy-conserving collisions between thermal phonons may now occur in first order in the cubic anharmonicity. Previously the dominant processes were assumed to be four-phonon transitions arising in second order in perturbation theory.[15,16] Since the γp^2 term is assumed to be much less than 1, the new allowed processes are very small-angle collisions, and the wide-angle scattering time τ will still be governed by the second-order processes previously considered. Consequently, the small-angle time $\tau_\parallel$ will be much less than $\tau_\perp$. One therefore expects that three different sound velocities will occur corresponding to the conditions $\omega\tau_\parallel \gg 1$, $\omega\tau_\parallel \ll 1 \ll \omega\tau_\perp$, $\omega\tau_\perp \ll 1$. The first of these corresponds to true collisionless or zero sound, and in this case, we may express the velocity correction in the form

$$\Delta c_0 = -\frac{\pi^2}{30}\frac{(u+1)^2}{\rho\hbar^3}\left(\frac{kT}{c}\right)^4 \ln\left|\tfrac{3}{2}\gamma\bar{p}^2\right|$$

$$= \frac{\pi^2}{30}\frac{(u+1)^2}{\rho\hbar^3}\left(\frac{kT}{c}\right)^4 \langle \ln|(c-v_p)/2c|\rangle, \quad (5)$$

where $v_p = \partial\epsilon/\partial p$ is the phonon group velocity and the angular brackets denote an average over the phonons with which the sound wave interacts. The second expression is more general in that the dominant phonon approximation has not been used, and the assumption that the γp^2 term dominates over the δp^4 term is not necessary. At lower frequencies where $\omega\tau_\parallel \ll 1$ but $\omega\tau_\perp \gg 1$, thermal phonons with parallel momenta will be coupled together by small-angle scattering. In the case where this coupling is assumed strong, a reasonable modification of Eq. (5) is[17]

$$\Delta c_\parallel = -\frac{\pi^2}{30}\frac{(u+1)^2}{\rho\hbar^3}\left(\frac{kT}{c}\right)^4 \ln|(c-\langle v_p\rangle)/2c|.$$

We refer to this as "parallel sound." At even

lower frequencies when $\omega\tau_\perp \ll 1$, sound travels at the adiabatic velocity corrected by the coupling to second sound.[18] In the frequency and temperature range investigated by Abraham et al.,[7,9] $\omega\tau_\perp \gg 1$. A possible interpretation of these velocity measurements is that the decrease in velocity observed as the frequency increases arises from a transition from parallel sound to zero sound. This would require $\Delta c_\parallel > \Delta c_0$. This is possible because $\Delta c_\parallel$ may be large and positive if γ and δ are such that the average group velocity $\langle v_p \rangle$ of the thermal phonons is nearly equal to the velocity c of the sound wave.

A more detailed discussion of these points must await measurements of the helium dispersion curve to lower momenta and more exact solutions of the transport equations. We note that even the recent measurements of Woods and Cowley still do not reach that part of the spectrum that is thermally excited in the temperature range discussed in this paper.

We thank Professor Leo Kadanoff, Professor Charles Elbaum, and Professor George Waters for useful discussions.

---

†Work supported in part by the Advanced Research Projects Agency and the National Science Foundation.

[1]C. J. Pethick and D. ter Haar, Physica 32, 1905 (1966).

[2]P. C. Kwok, P. C. Martin, and P. B. Miller, Solid State Commun. 3, 181 (1965).

[3]A. Andreev and I. M. Khalatnikov, Zh. Eksp. Teor. Fiz. 44, 2058 (1963) [Sov. Phys. JETP 17, 1384 (1963)]; I. M. Khalantnikov and D. M. Chernikova, Zh. Eksp. Teor. Fiz. 49, 1957 (1965) [Sov. Phys. JETP 22, 1336 (1966)], and Zh. Eksp. Teor. Fiz. 50, 411 (1966) [Sov. Phys. JETP 23, 274 (1966)].

[4]Y. Disatnik, Phys. Rev. 158, 162 (1967).

[5]B. M. Abraham, Y. Eckstein, J. B. Ketterson, and J. M. Vignos, Phys. Rev. Lett. 16, 1039 (1966).

[6]G. W. Waters, D. J. Watmough, and J. Wilks, Phys. Lett. 26A, 12 (1967).

[7]B. M. Abraham, Y. Eckstein, J. B. Ketterson, M. Kuchnir, and J. P. Vignos, Phys. Rev. 181, 347 (1969).

[8]B. M. Abraham, Y. Eckstein, J. B. Ketterson, M. Kuchnir, and P. R. Roach, Phys. Rev. A 1, 250 (1970).

[9]B. M. Abraham, Y. Eckstein, J. B. Ketterson, and M. Kuchnir, Phys. Rev. Lett. 19, 690 (1967).

[10]See, for example, the discussion in Ref. 7.

[11]A. D. B. Woods and R. A. Cowley, Phys. Rev. Lett. 24, 646 (1970).

[12]The possibility of a positive dispersion (negative γ) was suggested some time ago by H. W. Jackson, thesis, Washington University, 1962 (unpublished). See also, W. E. Massey, Phys. Rev. Lett. 12, 719 (1964).

[13]R. B. Hallock, Phys. Rev. Lett. 23, 830 (1969), and private communications.

[14]For an analysis of the consistency of the neutron and x-ray data, see D. Pines and C.-W. Woo, Phys. Rev. Lett. 24, 1044 (1970).

[15]I. M. Khalatnikov, *Introduction to the Theory of Superfluidity* (Benjamin, New York, 1965).

[16]J. Wilks, *The Properties of Liquid and Solid Helium* (Oxford Univ., London, England, 1967).

[17]This result can be obtained by considering the extreme case when the thermal phonons attain local equilibrium along a line in K space (zero-angle scattering). The solution of the Boltzmann equation for the more complicated problem of small (but nonzero) angle scattering is being pursued but has not yet been attained. For a discussion of the relevant transport equations involved in the calculation, see Ref. 3.

[18]See Wilks, Ref. 16, Chap. 8.

RAMAN-SCATTERING SELECTION-RULE BREAKING AND THE DENSITY OF STATES IN AMORPHOUS MATERIALS*

Reuben Shuker and Robert W. Gammon

Vitreous State Laboratories, Physics Department, The Catholic University of America, Washington, D. C. 20017

(Received 25 May 1970)

We present a calculation of the dielectric correlation function in glasses showing how the assumption of short correlation length for normal modes breaks the momentum selection rules and leads to expressions for the first-order Raman-scattering intensity in terms of the density-of-states functions and known frequency-dependent amplitudes.

Because of the current interest in Raman scattering and the properties of amorphous solids, we have been attempting to understand the shapes of the bands observed in Raman scattering of glasses. In the most frequently studied substance, vitreous silica, good data are available at room temperature[1] and low temperatures[2,3] but an adequate interpretation of the scattering in glasses giving the observed bands has been lacking.

In this Letter we present the outline of a calculation leading to an equation for the spectral