THREE-PHONON PROCESS AND THE PROPAGATION OF SOUND IN LIQUID HELIUM-4†

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The current theoretical formulations of the three-phonon mechanism for the velocity dispersion and for the attenuation of sound in liquid He-4 are shown to be incompatible with new experimental data presented here.

An acoustic wave propagated through liquid He^4 is a powerful probe for studying the interactions between elementary excitations. Recently published measurements[1] of the attenuation of sound in liquid He^4 showed that, in the temperature range 200-500 mdeg, the attenuation varied nearly as T^4, as required for a three-phonon mechanism. The agreement with theory was only qualitative, however, since the calculated attenuation was approximately a factor of 3 too low.[2]

According to the theories of Pethick and ter Haar[3] and of Kwok, Martin, and Miller,[4] the attenuation, α, due to the three-phonon process is given by

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

and the change in velocity, Δc, by

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

where the symbols are defined as follows: ρ is the density; c, the velocity of sound; k, the Boltzmann constant; u, the Grüneisen constant $(\rho/c)(\partial c/\partial\rho)$; ω, the angular frequency; τ, the thermal phonon lifetime; γ defined through the energy-momentum relation $\epsilon = cp[1-\gamma p^2]$; and $\bar{p} = 3kT/c$.[5] Similar expressions for the attenuation and velocity change were derived by Khalatnikov and Andreev,[6] Khalatnikov and Chernikova,[7] Ron,[8] Eckstein,[9] and Disatnik.[10]

The earlier measurements of attenuation at 30, 90, and 150 MHz have now been extended to include an additional seven frequencies, 12, 36, 60, 84, 108, 132, and 204 MHz. A least-squares fit to the data as a function of frequency was made at several temperatures from which the best value for u was found to be 5.30 in lieu of the accepted value 2.65.[7] Furthermore, $\tfrac{3}{2}\gamma\bar{p}^2\tau$ had the value $(2.7\times10^{-10})/T$, which implies that τ varies as T^3 and not as T^{-9} as is expected from theory.[7] Thus Eq. (1) provides only a qualitative description of the data.

Since the velocity and the attenuation are, respectively, the real and the imaginary parts of the same self-energy diagrams, velocity changes as a function of temperature and frequency were made along with the attenuation measurements at 12, 36, 60, and 84 MHz in order to provide a more thorough test of the theories. It was hoped that internal consistency could be achieved and that values for τ and γ could be extracted from the measurements. However, the velocity changes are in complete disagreement with the theoretical predictions for the dispersion and temperature dependence and cast serious doubt on the validity of current theoretical formulations for the three-phonon process.

The sonic cell, refrigeration technique, and the electronics were the same as those previously described[1,2,11] with the exception that a calibrated variable delay line was inserted into the reference channel. Velocity measurements were made by matching the amplitude of a reference signal to the signal from the helium and adjusting the phase of the reference with the delay line to null the signal from the helium; both pulses were obtained from the same oscillator operated in cw mode. Measurements of attenuation and of velocity were made at increments of $0.1T$ starting from the lowest temperature. Once the frequency was chosen, care was exercised to stay within 10 Hz of the initial frequency with the aid of an electronic counter. The measured phase shift of the attenuator was less than $0.1°/10$ dB which corresponds to 0.02 nsec/10 dB, thereby introducing a negligible error.

This technique yields only a value for the change in transit time, Δt, through the helium, i.e.,

$$\frac{1}{c(\omega, T_2)} - \frac{1}{c(\omega, T_1)} = \frac{\Delta t}{D}, \quad (3)$$

where $c(\omega, T_i)$ is the velocity at angular frequency ω and temperature T_i, and D is the acoustic path length. Equation (3) can be re-
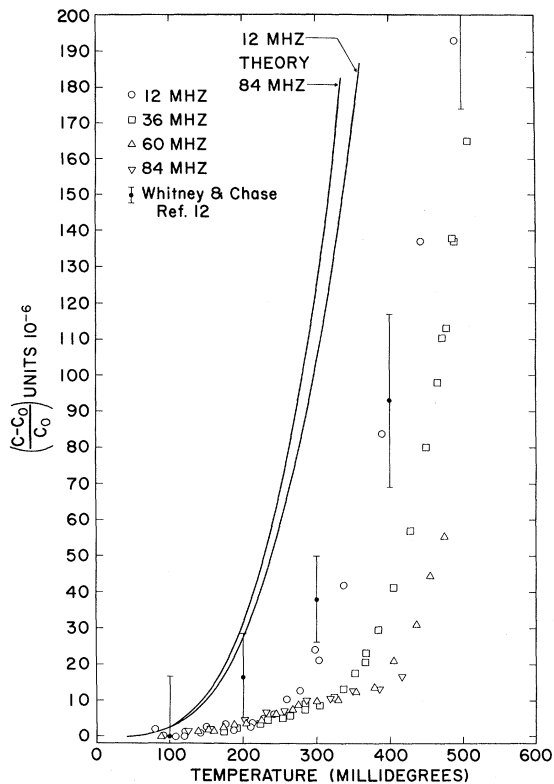
FIG. 1. Velocity changes in liquid helium-4 as a function of temperature and frequency. (Points are experimental; solid curves are theoretical.)

written as

$$\frac{\Delta c}{c(\omega,0)} = \frac{c(\omega,T)-c(\omega,0)}{c(\omega,0)} \approx -\frac{c(\omega,0)\Delta t}{D}, \qquad (4)$$

where $c(\omega,0)$ is the velocity of sound at the absolute zero of temperature. If Δt is zero, then Δc is also zero. In the temperature interval 35–175 mdeg, no change in transit time was observed for any of the four frequencies within the precision of 0.1 nsec; therefore, Δc in this temperature interval was zero within a precision of 4.8×10^{-2} cm/sec. Theoretically, one finds that the velocity changes go to zero as T^4; experimentally, one finds that the velocity changes go to zero as T^3, in the low-temperature limit. Therefore, the experimental values for $\Delta c/c(\omega,0)$, calculated from Eq. (4), represent actual changes from the zero-temperature velocity within the precision of two parts in 10^6. The value for $c(\omega,0)$ in Eq. (4) was taken from Whitney and Chase,[12] 2.3827×10^4 cm/sec. It should be emphasized at this point that any change in the value for $c(\omega,0)$ will in no way alter the conclusions in

this communication.

The results are presented in Fig. 1, where the size of the points represents the experimental error. The solid curves were calculated from the theory of Pethick and ter Haar,[3] Eq. (2). The calculations according to the theory of Khalatnikov and Chernikova[7] contain extra contributions which would result in lowering the theoretical curves by about 10%. In order to avoid confusion the theoretical curves only at the frequency extremes (12 and 84 MHz) are plotted. The points with the vertical error bars are recent measurements of Whitney and Chase[13] at 11.9 MHz which agree with these data within the error limits. Since we made no measurements below 12 MHz, a comparison with the Whitney and Chase data at 1 and 4 MHz cannot be made; however, their data show an inverse frequency dependence from those presented here. Because of the inadequacies of the theories no further comment about their data can be made. It is immediately apparent from Fig. 1 that experiment does not agree with theory: First, changes in the velocity are smaller than expected theoretically; second, for small changes the temperature dependence starts as T^3 and then goes over to T^6 where the theory predicts T^4; third, the frequency dependence is much greater than expected from theory and is in the opposite direction; i.e., experimentally, the higher the frequency, the smaller the velocity change. Whereas it was possible to bring the attenuation measurements into accord with theory by changing u and the temperature dependence of τ, this is not possible with the velocity changes. If one employs the value for u found from the attenuation, then the disagreement in magnitude between theory and experiment becomes greater; for u has to be increased because the theoretical value for the attenuation was too low, but the theoretical value for the velocity change is too high. The situation with respect to frequency dependence is worse; there is no way to invert the frequency dependence of the equations. It is concluded, therefore, that the current formulations for the three-phonon process in liquid He[4] are incorrect.

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SHALLOW IMPURITY SURFACE STATES IN SILICON

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Levine[1] has proposed an effective-mass model for shallow donors on semiconductor surfaces. His potential is $V = -e^2/\kappa r$ in the interior of the crystal, where κ is the dielectric constant, and $V = +\infty$ exterior to the crystal. The exterior potential is taken to be at $+\infty$ due to the high electron affinity of the crystal. However, two very important contributions to the potential have been omitted in his model. Because of the dielectric discontinuity of the surface, the potential[2] due to the impurity center is e^2/r reduced by the <u>average</u> of the exterior and interior dielectric constants rather than by just the interior κ, and there is also a potential[2] caused by the image charge of the excess electron. In a recent paper,[3] Petukhov, Pokrovskii, and Chaplik have used the same Hamiltonian that we use. They have made an error in that their image potential is too small by a factor of 2. They do not solve the exact Hamiltonian, but rather obtain solutions to an approximate hydrogenic Hamiltonian and then use first-order perturbation theory to account for the image potential, which has cylindrical symmetry.

Using our potential in the case of a high-dielectric-constant material with a clean surface, the effective dielectric constant for the impurity center is halved, and the image charge produces a repulsive potential in the interior which reduces the total potential up to 25% and reduces the symmetry from hemispherical to cylindrical. The resulting impurity states are somewhat deeper than those calculated from Levine's results. We have calculated the ground state and the four lowest optically excited states for a shallow donor in Si using an isotropic effective mass.

Our potential is

$$V = \frac{-2e^2}{(\kappa+1)r} + \frac{e^2(\kappa-1)}{2\kappa z(\kappa+1)} \quad z \geq 0, \tag{1a}$$

$$V = +\infty \quad z < 0, \tag{1b}$$

with $\kappa = 12.0$, $m^* = 0.309 m_0$.[4] The surface is taken to be at $z = 0$, where z is distance perpendicular to the surface. Since $V = +\infty$ outside the crystal, the boundary condition on the wave function is $\psi(0) = 0$. This requires that only those spherical harmonics may appear which satisfy

$$(l + m) = \text{odd}. \tag{2}$$

Since our potential (1) has cylindrical symmetry, the azimuthal quantum number m is a constant of the motion. We used the following variational functions for a given m:

$$\Psi = N r^{m+1} e^{-r/a} \sum_{l=m+1}^{L} a_{lm} Y_{lm}, \tag{3}$$

where all the l's are either even or odd in order to satisfy (2). The ground state is the lowest $m = 0$ state, since this state contains the lowest order Y_{lm} permitted by (2), namely,