Temperature and frequency dependence of longitudinal sound in hcp ^{4}He

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Data are presented for the longitudinal sound velocity of single crystals of hcp ^{4}He at a molar volume of 17.4 cm^3/mol from 0.25 to 3.0 K. The velocity was measured at 5, 15, and 25 MHz on each crystal. Most, but not all, crystals show a strongly anomalous velocity and attenuation below about 1.8 K. The anomaly moves to lower temperatures with increasing frequency. Very large dispersion is found in all crystals, the dispersion being normal in most crystals, but anomalous in two crystals in the 15 to 25 MHz range. Various possible explanations are discussed.

Several investigations in the last few years have shown that the propagation of ultrasound in the megahertz region in hcp ^{4}He shows anomalous behavior at temperatures below about 1.5 K.[1-4] The anomaly takes the form of a deviation from adiabatic propagation and of a steeply rising attenuation with falling temperature. Two different explanations for this effect have been advanced: Franck and Hewko[1] proposed that the effect is due to the coupling between first and second sound, whereas Wanner *et al.*[3] propose a coupling between first sound and vibrating dislocation lines.[5] We present here data on the temperature and frequency dependence of the longitudinal sound velocity of single crystals of hcp ^{4}He. The data show the following: the longitudinal sound velocity has very large dispersion at all temperatures from the melting point on down; the anomalous behavior is not always present; in crystals that show an anomaly (the majority of those grown), the anomaly moves to lower temperatures with increasing frequency. An analysis of the data shows that both explanations advanced so far for the effect have difficulty in explaining the observed results.

The experimental apparatus consists essentially of a pressure cell which contains the ultrasonic etalon. We used $\frac{1}{2}$-in. quartz tranducers of 5-MHz fundamental frequency, gold plated and overtone polished.[6] The same transducer was used for transmitting and receiving. The reflecting surface was made from beryllium-copper bronze, polished flat to $\frac{1}{2}$ Na wavelength. Its diameter was large enough to accommodate the full ultrasonic beam at all crystal orientations.[7]

All crystals were grown from purified helium gas at a constant pressure of 124.4 bar corresponding to a molar volume of 17.4 cm^3/mol, over a period of 6–8 h. We frequently obtained superior crystals as judged by the number of sound echoes received and the appearance of the interference pattern. Crystals with fewer than 100 echoes near melting were rejected for measurement.

Measurements of the absolute sound velocity were made using the pulse-echo-overlap method.[8-11] In this method, an audio-frequency pulse generator triggers the oscilloscope at a rate equal to the inverse travel time between echoes. The ultrasonic transducer, on the other hand, is triggered at a slower rate, typically 1/1000, obtained by frequency division. In this way, individual pulses are allowed to die out in the crystal before a second pulse is triggered. The overlap of the rf waveform near the center of successive echoes was visually observed on the viewing screen of the oscilloscope. The inverse of the triggering frequency at which perfect overlap occurs gives then the travel time between two rf oscillations in two adjoining echoes, i.e., it measures the phase velocity. The method can produce gross errors if noncorresponding rf oscillations are overlapped in successive echoes. We used McScimin's method[12] for the choice of correct oscillations; the results obtained using this criterion always agreed well with the velocity obtained from the average time between echoes along the entire echo train. For the measurement of the temperature dependence the same set of rf oscillations was followed over the entire temperature range. Sensitivities for detecting changes in the velocity were about 1×10^{-5} at the fundamental (5 MHz), 2×10^{-5} at the third harmonic (15 MHz), and 1×10^{-4} at the fifth harmonic (25 MHz). For the measurement of dispersion McScimin's method had to be used separately at each frequency. We found that this criterion always led to the smallest possible shift of velocity with frequency. Measurements at different frequency also introduce errors due to finite pulse width[13] and to diffraction.[14] Both effects can be estimated to be below 5×10^{-5} when going from 5 to 15 MHz, or from 15 to 25 MHz. These estimates were borne out by observations on the liquid above the melting point. Observed changes in the measured velocity of the liquid were -6×10^{-5} between 5 and 15 MHz and -1×10^{-5} between 15 and

25 MHz.

Estimates of the angle γ between crystal axis (c axis) and wave normal were obtained by comparing the absolute velocity with the anisotropic longitudinal velocity. Crystals with velocities over almost the whole anisotropic range were observed. In the range $42° \leq \gamma \leq 90°$ the longitudinal velocity is double-valued. In this range we used the observed interference pattern as a guide to choose a "most likely" angle γ. This possibility exists because of the angular dependence of the beam deviation and the consequent change in the interference pattern.

The experiment was cooled by a SHE minifridge.[15] Temperatures were determined from the resistance of a calibrated 220-Ω $\frac{1}{2}$-W Speer resistor mounted on the pressure cell.

In Figs. 1 and 2 we show results for two crystals that exhibit the anomaly. At high temperature, the sound velocity is well described by an expression of the form

$$v(T) = v_0 - aT^4 + bT^6 \ . \qquad (1)$$

This behavior is expected in the adiabatic range. At some temperature near 1.8 K the attenuation starts to rise sharply with falling temperature, as judged by the decrease in the amplitude of the echoes. A crystal that near melting showed in excess of 100 echoes may go down to about three to five visible echoes at the lowest temperature for constant excitation levels. We are refraining from quoting absolute values of attenuation because of the difficulty of separating out purely geometric effects. The attenuation in the adiabatic range is, however, certainly very low.

At or below the temperature at which the attenuation rises steeply, the phase velocity also shows strongly anomalous behavior. In most crystals the phase velocity rises steeply above the adiabatic velocity, Fig. 1, reaching a plateau at temperatures about 0.2 to 0.5 K lower. The relative change from the extrapolated adiabatic velocity can be as large as 3×10^{-3}. The onset of the velocity anomaly was observed to occur in the range 0.9 to 1.6 K for different crystals. In two of the crystals we observed a decrease in phase velocity, of the order 2×10^{-3}; such a crystal is shown in Fig. 2. This decrease in velocity started at about 1.8 K.

Measurements of the frequency dependence of the sound velocity were done on each crystal by going to the third and fifth harmonics (15 and 25 MHz). The following was observed: all crystals show a remarkably large dispersion, at all temperatures from the melting point on down. The velocity decreases in most crystals with rising frequency (normal dispersion) by between 2×10^{-4} and 8×10^{-4} for a frequency change of 10 MHz. This dispersion is exceptionally large; it can obviously not persist over too large a frequency interval. The large normal dispersion in the 5- to 25-MHz region will have to be accompanied almost necessarily by an equally large anomalous dispersion region, probably at higher frequency. In fact, the crystal in Fig. 2 shows anomalous dispersion already in the 15- to 25-MHz range; anomalous dispersion was observed in another crystal as well. We find further that the velocity anomaly

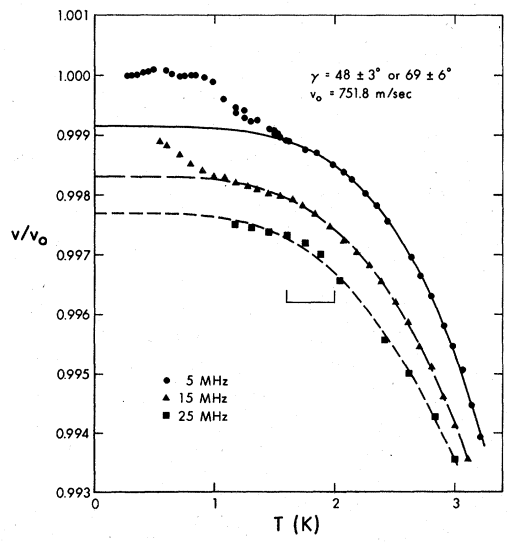

FIG. 1. Reduced velocity vs temperature for crystal $A6$-E at a most probable orientation of 69° from the c axis. The curves are fits to Eq. (1). The horizontal bracket indicates the range within which the attenuation began to rise sharply.

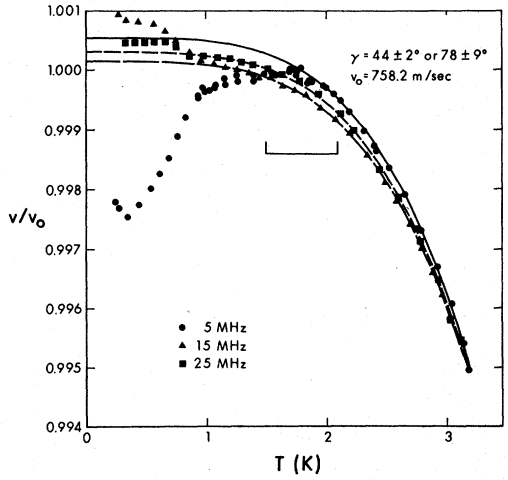

FIG. 2. Reduced velocity vs temperature for crystal $A8$-A at a most probable orientation of 78° from the c axis. The curves are fits to Eq. (1). Horizontal bracket as in Fig. 1.

becomes generally smaller and moves to lower temperature as the frequency increases. The negative-going velocity anomaly at 5 MHz in the crystal of Fig. 2 becomes positive going and also moves to lower temperature.

We have also, surprisingly, observed two crystals in which both anomalies are absent, Fig. 3. In those crystals we estimate that sound propagated at 43° and 72°, respectively, from the c axis. The sound velocity follows Eq. (1) down to the lowest temperatures. These crystals show a slight decrease in echo height down to about 1.8 K. Below this temperature there is little or no further change. At 0.25 K we observed about 50 echoes in these crystals, as compared to three to five in the other crystals.

All crystals measured have always shown reproducible behavior, both for temperature cycling and for changes in frequency. We have so far not found any discernible orientation dependence of the effect. We feel that the effect must be connected with some type of lattice imperfection, whose concentration differs from crystal to crystal. Any orientation dependence that might exist is, at this stage, overshadowed by the influence of imperfections.

We will briefly discuss the various explanations put forward for the observed effects. Explanations based on interactions with vibrating dislocation lines use the result of Granato and Lücke[5] for the relative velocity change $\Delta v/v$ induced by dislocation lines of length l:

$$\frac{\Delta v}{v} = R^2 \Lambda(l) \frac{4v_0^2}{\pi^3} \frac{\omega^2 - \omega(l)^2}{[\omega^2 - \omega(l)^2]^2 + (\omega d)^2} \cdot \qquad (2)$$

Here, $\omega/2\pi$ is the sound frequency, $\omega(l)$ is the eigenfrequency of the dislocation line, $\Lambda(l)$ is the total length of dislocations of length l per volume, $R^2 = \sin\gamma\cos\gamma$ is a geometric factor (γ is the angle between sound wave vector and c axis), and d is a damping constant. Generally a distribution of dislocations of various lengths must be assumed, an exponential distribution being the most likely case. The damping arises via scattering of phonons,[16] and therefore is of the form $d = \text{const} T^n$, where $n = 5$ in the low-temperature limit ($T \ll \theta_D$). Wanner *et al.*[3] propose that the observed anomaly represents the transition from large damping, $d/\omega \gg 1$, to small damping, $d/\omega \ll 1$. At a given frequency it is always possible to fit the velocity anomaly to such an expression, although not necessarily for an exponential distribution. We have made such fits and find in crystals that show an anomaly a total dislocation density Λ of about $10^4/\text{cm}^2$. This is an order of magnitude smaller than quoted by Wanner *et al.*[3] In the crystals that show no anomaly, only an upper limit of about $10^2/\text{cm}^2$ can be given for Λ. These values of Λ are surprisingly small. The exponent n was found in the range 2.7–5.0 for different crystals; it is different even for the same crystal at different frequencies. This apparent frequency dependence of n is difficult to reconcile with the idea of a damping constant dependent only on the external phonon bath. The most stringent requirements, however, have to be met when the results for different frequencies on the same crystal are compared. As follows from Eq. (2), the temperature at which the anomaly occurs (e.g., the maximum slope of $\Delta v/v$ vs T) should be given by $d/\omega = \text{const}$. Since d should be frequency independent,[16] this leads to $T_2/T_1 = (\omega_2/\omega_1)^{1/n}$. This result holds also very nearly when a distribution of dislocation line lengths is assumed. We examined this numerically for the exponential distribution. In going to the third harmonic, we would then expect the anomaly to occur at a temperature 20% to 50% larger. Instead, we find a decrease of from 15% to 35% in all crystals. The frequency dependence of the observed effect is therefore in disagreement with the requirement of Eq. (2).

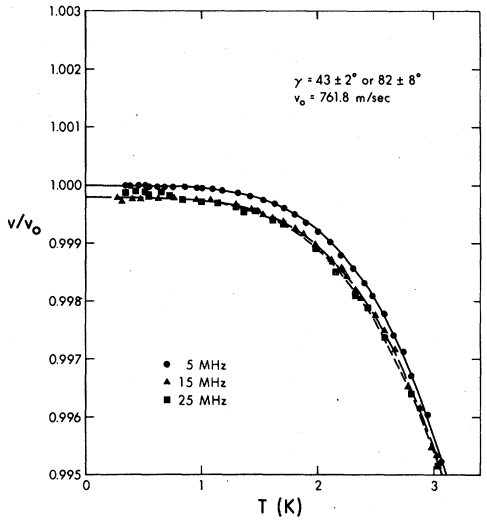

FIG. 3. Reduced velocity vs temperature for crystal A7-A at a most probable orientation of 43° from the c axis. The attenuation in this crystal did not experience a sharp rise.

This result is decisive since there are no further adjustable parameters involved in a frequency comparison. Irreversible changes in the crystal also appear to be ruled out since the results at different frequencies are completely reproducible. We believe that the observed frequency dependence of the effect rules out interactions with dislocations as the underlying cause. Moreover, any theory based on an interaction with a system of independent resonators will have great difficulty in giving the observed frequency behavior.

Coupling between first and second sound[17-19] predicts anomalous behavior in the "second-sound window" $\omega\tau_N \ll 1 \ll \omega\tau_U$, where τ_N and τ_U are the normal and umklapp relaxation times. In the ballistic range, $\omega\tau_N > 1$, the velocity should approach the zero-sound limit, which at 0 K is identical with the extrapolated adiabatic velocity.[19] Arguments against such an explanation were first given by Wanner *et al.*[3] and are essentially: the observed anomalous $\Delta v/v$ is orders of magnitude larger than any reasonable prediction for the difference between first and zero sound velocity; the velocity at low temperatures fails to approach the extrapolated adiabatic velocity. One might add to this that a coupling between first and second sound is essentially a bulk effect, whereas the observed effects are obviously connected with lattice imperfections. Estimates of τ_N and τ_U from thermal conductivity measurements,[20] moreover, indicate that no second-sound window exists for the present measurements at the higher frequencies.

The existence of anomalous dispersion, observed so far in two crystals and strongly hinted at in all others, has changed this analysis somewhat, due to allowed three-phonon processes. Calculations for liquid helium[21-25] which also exhibits anomalous dispersion, have shown that the approach to the zero-sound regime is resonancelike and extends over an extremely large temperature interval. This is due to an increase of the second-sound velocity towards the first-sound velocity in the megahertz region and the existence of collective second sound well into the region $\omega\tau_N > 1$. Sound velocity measurement on liquid helium[26] have shown this behavior. The magnitude of the velocity anomaly expected to occur during the transition from first to zero sound is also critically dependent upon the exact form of the dispersion relation.[19] In the solid we believe that the anomalous dispersion region is dependent on lattice imperfections (vacancies?).[27] Calculations would require an exact knowledge of the dispersion relation in each crystal, and are not available at present.

We are at present not in a position to propose any other explanation for the observed effects. There have been a number of proposals for a superfluid phase of hcp ^{4}He.[28-32] These theories are not developed enough to allow detailed comparison with experiment.

It is interesting to note that recently Andronikashvili *et al.*[33] have observed an attenuation anomaly in hcp ^{4}He at a frequency of 500 Hz in the temperature range 1.4–2.0 K. Whether this anomaly is the same as the one seen in the megahertz region is not clear at present.

ACKNOWLEDGMENT

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