VOLUME DEPENDENCE OF THE LONGITUDINAL SOUND VELOCITY IN hcp He⁴ SINGLE CRYSTALS

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Measurements of the longitudinal sound velocity v_I in htp He⁴ as a function of direction relative to the hexagonal axis are reported for molar volumes between 20.3 and 17.3 cm³/mole. The variation of v_I with molar volume is much stronger than predicted by theory. Agreement is found with theoretical predictions for the anisotropy of v_I .

The sound velocity in solid helium, calculated according to classical lattice dynamics, turns out to be imaginary.¹ A large amount of theoretical work has therefore been done to establish a quantum theory of lattice dynamics² which has been applied to hcp He⁴.^{3, 4} From the experimental side, the only measurements of longitudinal velocities⁵ done on polycrystalline samples gave values with a spread of about 12%, indicating the presence of large single crystallites and an anisotropy of that order. It has therefore been difficult so far to judge the success of these theories in accurately predicting sound velocities.

We present here measurements of the longitudinal sound velocity in single crystals of hcp He⁴ of known orientation at three different densities. We used the pulse-echo method at a frequency of 10 MHz to measure the velocity with an accuracy of about 0.5%. The flight time of the sound pulses was measured between the first and second echo using the calibrated delayed sweep of an oscilloscope. The path length is calculated from the flight time in liquid helium at 4 K and the known sound velocity in the liquid.⁵ Crystals were grown at constant pressure in a cylindrical copper-beryllium container with a copper bottom by applying a time-dependent temperature gradient that produced a constant growth rate of 2 cm/h.⁶ After about 1 h of growth the sound echos from the liquid disappeared and a new echo with a shorter flight time, corresponding to propagation in solid He, appeared, grew bigger, and usually reached a constant amplitude about 3 h after start of growth. Some samples also showed a drift in velocity of up to 2% during this time, usually to higher values, which we ascribe to recrystallization effects.

The crystal orientation was found by using the fact that the hexagonal phase of helium is birefringent. The difference of the refractive indices has been measured by Vos et al.^{7,8} and Heybey and Lee.⁹ We shine plane-polarized light from a He-Ne laser through a 12-mm-diam fused-quartz window into the sample container where it is reflected from the surface of the aluminized ultrasonic transducer and leaves the container through the same window. The window is also the sound reflector so that light and sound paths coincide. After leaving the cell, the light is in general elliptically polarized and its state is analyzed with a quarter-wave plate and an analyzer for several polarization directions of the entering light. From the change of state of polarization, the birefringence $n_e - n_o$, and the thickness of the sample (2.08 cm), the angle between the c axis and the light path can be calculated.⁸ Corrections for birefringent windows and other phase-shifting optical elements in the light path have to be made. They are found when the container is filled with liquid helium. At this point one can also see whether the sample is a single crystal or not, because a polycrystalline sample with a few large crystallites would give ambiguous results in the orientation determination. Approximately half of all the crystals grown were single crystals. The birefringence method does not give the direction of the *a* axis, but fortunately this information is not required since the velocity surface of hexagonal materials has cylindrical symmetry around the c axis.¹⁰ The sound velocity depends therefore only on the angle α between the c axis and the wave normal of the sound wave.

In Fig. 1 we plot sound velocities versus α for a sample grown at a melting pressure of 36.0 bar, corresponding to a molar volume of 20.32 cm³.¹¹ The spread of the data is due to uncertainty in the orientation measurement ($\pm 5^{\circ}$ at α = 45°, $\pm 10^{\circ}$ at α = 10° and at 80°) and possibly to small undetected inclusions of liquid or of crystals of different orientation in some of the samples. Less extensive data have been taken at 58.0 bar ($V = 19.28 \text{ cm}^3/\text{mole}$) and two points at 128.0 bar (V = 17.33 cm³/mole). The agreement between theory and experiment is seen to be good as far as the anisotropy is concerned. Similar results have been obtained at a density of 20.97 $cm^3/mole$ by Crepeau, Heybey, and Lee¹² who grew single crystals from the superfluid. Their



FIG. 1. Longitudinal sound velocity in hcp He⁴ at 20.32 cm³/mole as function of the angle α from the *c* axis. The solid line is taken from a calculation at 19.3 cm³/mole (see Ref. 4) and scaled down to give the best fit with the data.

results include data on the transverse branches. These authors also find good agreement with the anisotropy predicted by theory.

Of the five elastic constants of a hexagonal crystal only four $(c_{11}, c_{33}, c_{44}, c_{13})$ determine the longitudinal sound velocity; c_{12} can only be obtained from transverse modes. Of these four constants only two can be obtained from the present set of measurements with sufficient accuracy: c_{33} from the velocity along the *c* axis, and c_{11} from the velocity in the basal plane. We find $c_{33} = 7.1$, $c_{11} = 5.5$ (all elastic constants in units of 10^8 dyn/cm^2) at a molar volume of $20.32 \text{ cm}^3/$ mole, and $c_{33} = 9.8$, $c_{11} = 7.6$ at a molar volume of $19.28 \text{ cm}^3/\text{mole}$. The estimated error is 3%.

In Fig. 2 we present the volume dependence of the longitudinal sound velocity for propagation along the c axis and at 45° to the c axis. We see that fair agreement exists between our data and the experimental data of Crepeau, Heybey, and Lee,¹² as well as results from inelastic neutron scattering.^{13,14} Present theory predicts a much weaker volume dependence than is found experimentally. It can be seen in particular that the previously found agreement⁴ for molar volumes near 20 cm³/mole was somewhat fortuitous.

One can obtain Grüneisen gammas for the longitudinal branch from the present results through the relation

$$\gamma_{I} = -\partial \ln v_{I} / \partial \ln V + \frac{1}{3}.$$

We obtain within our accuracy the same value for propagation along the c axis and at 45° to the c axis, numerically: $\gamma_I = 3.0 \pm 0.1$. Present theory predicts a much smaller value, $\gamma_I \simeq 1.0$ (No-



FIG. 2. Longitudinal sound velocity in hcp He⁴ as a function of molar volume, double logarithmic. The upper and lower solid lines refer to propagation along the *c* axis and at 45° to the *c* axis, respectively. The squares refer to the longitudinal sound velocities along the *c* axis inferred from the neutron data obtained at Brookhaven (Ref. 13) and Iowa State (Ref. 14). The curves labelled NW and GKW are theoretical calculations for propagation along the *c* axis by Nosanow and Werthamer (Ref. 3) and by Gillis, Koehler, and Werthamper (Ref. 4) respectively.

sanow and Werthamer³) or $\gamma_I \simeq 0.7$ (Gillis, Koehler, and Werthamer⁴). A comparison can also be made with the Grüneisen gamma obtained from calorimetric data

$$\gamma_{\rm cal} = -\partial \ln \theta_0 / \partial \ln V,$$

where θ_0 is the Debye temperature at T = 0 K. Sample and Swenson¹⁵ and Edwards and Pandorf¹⁶ give in the density range of our measurements $\gamma_{cal} = 2.6$. It must, however, be remembered that transverse modes contribute mainly to θ_0 . Our results would then indicate that the longitudinal Grüneisen gamma in hcp He⁴ is larger than the transverse gamma by about 0.4. This is not unexpected and is similar to the situation found in many other materials.¹⁷

Comparison of our data with the neutron data of the Iowa State group¹⁴ shows that γ_1 decreases with decreasing molar volume. A similar behavior was also reported for γ_{cal} by Ahlers¹⁸ and

Sample and Swenson,¹⁵ and by Jarvis, Ramm, and Meyer¹⁹ for the Grüneisen gamma defined by

 $\gamma = (V/C_{\nu}) (\partial P/\partial T)_{\nu}$

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THEORY OF THE ONSET OF SUPERFLOW*

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Spontaneous production of quantized vortices in He II by thermal fluctuations is considered. Implications of this theory for superflow in finite channels are discussed.

In a recent communication¹ we examined the problem of the nucleation of quantized vortex rings by ions. There we presented arguments to suggest that the smallest vortex rings are rotons and that larger rings form higher-energy states which are filled by collisions. In order to fix our ideas and notation about superflow, consider first spontaneous vortex production in an unbounded fluid, a concept introduced by Iordanskii,² and applied to the interpretation of the decay of persistent currents by Langer and Fisher,³ Fisher,⁴ and Langer and Reppy.⁵ Rotons in a counterflow will be polarized against the oncoming superfluid and will become rings if they can fluctuate over a saddle point *C* in momentum space constituting a free-energy barrier ΔF . We shall show elsewhere that the probability per unit time of such a diffusion taking place may be estimated by Brownian-motion theory of vortex rings to be

$$P = \frac{\Lambda_c kT}{4\pi \mu^{1/2} p_0^2} \left[\frac{v_s p_0 / kT}{\sinh(v_s p_0 / kT)} \right] \left(\frac{\omega_c}{s_c^2} \right) e^{-\Delta F / kT}, \tag{1}$$

where Λ_c is the diffusivity constant for vortex rings (and includes contributions from rotons, phonons, and solvated He³ atoms as explained in Ref. 1), v_s is the relative velocity of the counterflow, μ and p_0 are the effective mass and momenta of rotons, ω_c and S_c are the principal curvatures of momentum space at the saddle point. The derivation of (1) follows from considerations similar to those leading to Eq. (133) of Donnelly and Roberts.⁶ In a counterflow the density of rotons is given by

$$N_r = 2(2\pi kT)^{3/2} \mu^{1/2} p_0 h^{-3} v_s^{-1} \sinh(p_0 v_s / kT) \exp(-\Delta/kT).$$
⁽²⁾

The rate of production of vortex rings per unit volume is therefore given by

$$\nu \equiv PN_r = (2\pi)^{1/2} (kT)^{3/2} \Lambda_c h^{-3} \omega_c S_c^{-2} \exp(-\Delta F / kT),$$
(3)

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