Interaction of First and Second Sound in Solid ^{4}He: Properties of a Possible Bose Condensate

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A previous experiment in this laboratory found that the addition of a few tens of parts per 10^6 of ^{3}He impurities to solid ^{4}He crystals resulted in a phase transition below 200 mK. In the experiment described here the interaction of acoustic waves with waves generated by heat pulses is measured and shows that there is an energy gap in the excitations of the high temperature phase and that coherent waves are carried by nonphonon excitations at the lowest temperatures.

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We have performed three experiments, in this laboratory, to search for evidence of a possible Bose-Einstein condensation in hcp solid ^{4}He close to its melting curve. Although this possibility has been discussed theoretically for many years [1–3] there is no definite experimental evidence for it. If it exists, it will constitute a previously unobserved state of matter. Our previous experiments found evidence for a wave propagation on excitations other than phonons [4] and a continuous phase transition below 200 mK in ^{4}He crystals containing a few tens of parts per 10^6 of ^{3}He impurities [5]. We were able to interpret the latter experiment as evidence for the presence of a Bose condensate, but the experiments were not sufficient to prove the interpretation. The experiment reported here has further investigated these two phases by measuring the interaction of acoustic waves and waves generated by heat pulses. It provides additional evidence that a non-phonon-type of wave is propagated by heat pulses. In the high T phase of the impure crystals these waves reveal an energy gap in the excitations. These results are also consistent with the Bose condensate model but still do not prove it.

In our first experiment [4], we found that acoustic waves are scattered by a nonphonon family of thermal excitations with activation energy $\Phi \approx 1$ K. They are given the generic name ''defectons'' but may be delocalized vacancies [6]. Our data indicated that these defectons were essentially free particles and suggested that they supported a coherent traveling wave.

In the second experiment [5] we found that the addition of as little as 14 ppm of ^{3}He resulted in a very large, sharp peak in the acoustic attenuation, α, at temperature, T_p, which decreased with increasing ^{3}He concentrations but also varied between crystals grown from the same mixture. The attenuation peak is accompanied by a similarly large increase in velocity, v, upon cooling, centered at T_p. The anomaly is characteristic of a relaxation process in which the velocity, v, and the attenuation, α, vary as

$$\Delta v = v - v_0 = (v_\infty - v_0)\frac{(\omega\tau)^2}{1 + (\omega\tau)^2}, \qquad (1)$$

$$\alpha = \frac{(v_\infty - v_0)}{v_\infty v_0}\left(\frac{\omega^2\tau}{1 + (\omega\tau)^2}\right), \qquad (2)$$

where $\tau = \tau(T)$ is the relaxation time for the response of the system to acoustic waves, ω is the angular frequency of the acoustic waves, and v_0 and v_∞ are, respectively, zero and infinite frequency limits of the velocity. The attenuation peak then occurs at $T = T_P$ such that $\omega\tau(T_P) = 1$. The data show that the T dependence is rapid near T_P so that it appears to result from a continuous phase transition. In that case τ is the relaxation time for the order parameter and τ becomes long as $|T - T_C| \to 0$. A sudden increase in T, near but above T_p, causes an immediate *increase* of α and v, followed by a larger, slow decrease toward the equilibrium state with time constant $\sim$10 h. Thermodynamic analysis [7] of the data indicates that the acoustic velocity is lower in the phase with finite order parameter. This implies that the order parameter is finite above T_P in our system.

In the experiment described here, we measured the propagation of waves generated by heat pulses along a path perpendicular to and intersecting the path of acoustic pulses. These waves were detected by shifts that they caused in α and v of the acoustic waves and by direct detection of the energy deposited by them on a bolometer. Since the dominant equilibrium temperature dependences of α at all T and of v near T_p, measured by the previous experiments, are due to the interaction of acoustic phonons with defectons, the measurements were intended to detect our previously suggested waves propagating on defectons. Since waves carried by defectons would likely travel at different velocity than phonon waves, the shifts were measured as a function of the delay time, τ_{delay}, between the generation of the heat pulse and acoustic pulse so as to determine the velocity of the wave. The shifts were also measured as a function of the heat pulse amplitude at fixed delay time to determine whether they were proportional to the temperature rise in the waves, measured directly by the bolometer. In effect, we have used the acoustic properties as a fast, sensitive, internal probe to measure the

propagation of waves generated by a heat pulse, independent of detection by the bolometer.

As a test of the method, we measured the effect in superfluid liquid ^{4}He at both the saturated vapor pressure and at pressures along the melting curve, since in that system the properties of phonon and roton second sound are well understood [8,9]. These measurements confirmed the method by showing that second sound waves shift the acoustic properties as expected and ballistic phonons have no effect on the acoustic waves.

The sample chamber is a copper cylinder with inside diameter 7.04 mm and inside length 8.43 mm. The acoustic transmitter and receiver are mounted on the inside of the cylinder, at its ends. A hole is drilled through the walls of the cylinder along a diameter at its center. The heater and bolometer are mounted on the ends of copper rods glued into these holes, on opposite sides of the cylinder, but recessed in the holes, such that the distance between them is 12.2 mm. The heater is located 4.7 mm from the axis of the cylinder and the bolometer at 7.5 mm. The heater and bolometer are thin film meander patterns of Pt and Zn, evaporated onto glass. The total area of the lines themselves is 0.35 mm^2. However, since the distance between the lines is small compared to the dimensions of the sample chamber, the 3.3 mm^2 area enclosed by the pattern is the relevant one to determine the power density in the pulses.

The acoustic signals are generated and detected by 9.1 MHz X-cut quartz crystals using the pulsed heterodyne system of our previous work. The heat pulses, for the work presented here, ranged from 0.5 to 2.5 V into the 180 Ω resistance of the thin film heater. Pulse widths between 1 to 4 μ sec were used. Thus the power in the pulses was between about 1 and 30 mW so that it extended up into the range where amplitude induced second sound was found previously [10]. We calibrated the bolometer so that the temperature rise in the heat pulses, projected back to the location where they interacted with the acoustic pulses, was determined to be between 1 and 10 mK for the measurements described here.

Unlike earlier second sound measurements, our sample chamber necessarily has reflecting surfaces and a long heat pulse path length, which allows the second sound waves to reach the bolometer detector by more than one path. This leads to a complicated, temperature dependent, bolometer signal shape (see Fig. 1). Nonetheless, all of the qualitative features of the second sound signals in liquid and solid are reproduced [9,10]. First arrival times are in quantitative agreement. At the lowest temperatures, the signal has a sharp leading edge, similar to ballistic phonons in the liquid, but in this case the first arrival is at 54 μ sec corresponding to a velocity of 226 m/sec, also in agreement with Ref. [10].

However, longitudinal ballistic phonons travel between 450 and 520 m/sec, depending on the direction of propagation relative to the c axis. For this reason the authors of

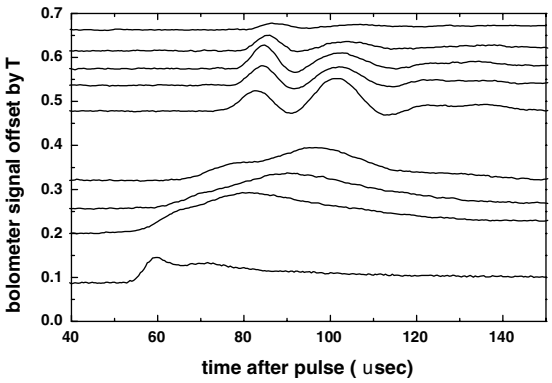

FIG. 1. The temperature dependence of the bolometer signals in the solid in our experiment. The shapes are due to the geometry of the sample chamber. The baselines of the curves are offset by the temperature at which they were obtained.

Ref. [10] argued that this large signal at 226 m/sec was due to focusing of ballistic, slow transverse phonons and that crystals in their sample chamber grew with the orientation required to provide the focusing. Our sample chamber is very different from theirs and variations of our absolute acoustic velocities indicated a 20° variation of the c axis of our crystals relative to the acoustic path. Nonetheless, the signal traveling at 226 m/sec is always at least an order of magnitude larger than any bolometer signal with earlier arrival time. Therefore, the phonon focusing explanation seems improbable for our experiment. The acoustic signal is shifted by this wave (see Fig. 4), so that ballistic phonons could not be responsible for both the bolometer signal and the acoustic shifts unless a significant fraction of them are thermalized in the interaction region and a significant fraction still reach the bolometer. Thermalization does occur for sufficiently large amplitude heat pulses and appears as self-induced second sound at low T. However, in that case the bolometer signal would have a gradual leading edge as between 200 and 400 mK in Fig. 1 rather than the sharp edge observed below 100 mK.

The shifts of α and v as a function of τ_{delay} for an impure crystal, which exhibited a large acoustic anomaly at the phase transition, are shown in Fig. 2 where the transition is between 158 and 198 mK. The peak value of $\Delta\alpha$ is small and negative at 100 mK, then becomes larger, and still negative, at 118 and 138 mK. As T increases further toward T_P the shift begins to change sign and then increases in magnitude up to the highest T measured for this crystal at 283 mK. Thus the peak shift in α is always of opposite sign to that which results from an increase of T at equilibrium, consistent with our observations of short term changes in the previous experiment [5]. Δv is also always of opposite sign to the equilibrium T dependence and increases with increasing T until it reaches a maximum at about 200 mK. There are two effects contributing to the long tails on $\Delta\alpha$ and $\Delta v/v$: (i) reflections and a diffusive tail from the fast

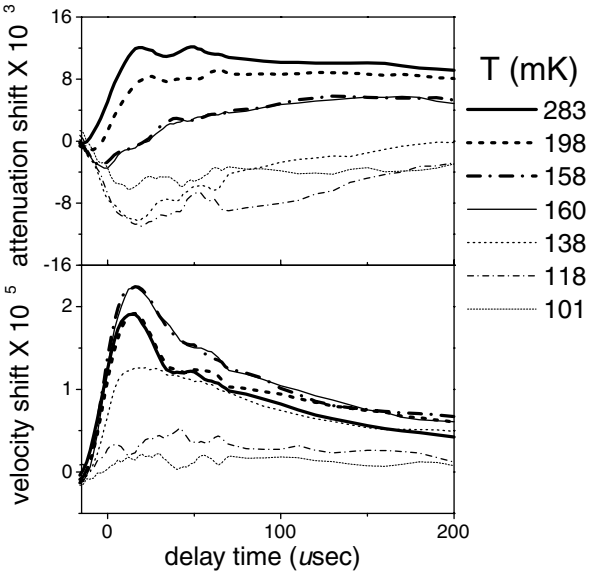

FIG. 2. Fractional velocity and attenuation shifts as a function of delay time for an impure crystal at the temperatures indicated. All heat pulses were 2.7 V.

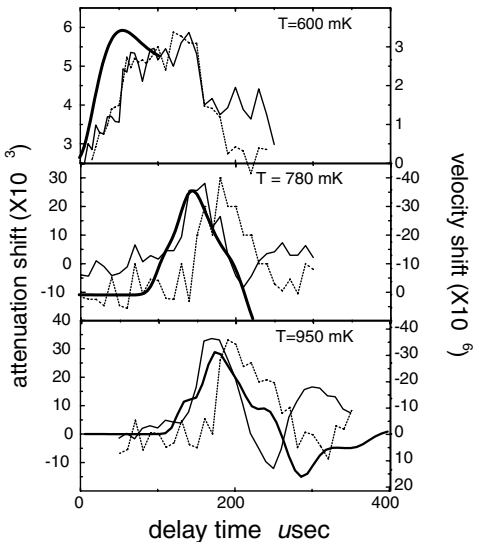

FIG. 3. Fractional shifts of acoustic attenuation and velocity due to interactions with heat pulses in liquid ^{4}He, compared to the convolution of the bolometer signal with the acoustic signal. The thin solid line is the attenuation, the dotted line is the velocity shift, and the heavy solid line is the convolution of the bolometer signal with the acoustic signal.

wave and (ii) phonon second sound waves and their reflections. The effect of the phonon second sound waves increases with increasing T, consistent with their increasing amplitude. The long tail on $\Delta\alpha$ reverses sign at a lower T than does the effect at the early arrival times so that the phonon second sound appears always to increase α. Above T_P the phonon second sound and the high velocity waves both increase α so that they add at long τ_{delay}. Above T_P, $\Delta v/v$ decays more rapidly as a function of τ_{delay} than does $\Delta\alpha$. We know that thermal phonons decrease v so that phonon second sound waves must decrease v. This partly cancels the tail of the increase from the fast waves.

In pure crystals, with low dislocation densities, the shifts in α and v are much smaller, consistent with the much smaller equilibrium T dependences. At all T, the sign and T dependence of the observed shifts correspond to those in impure crystals for $T < T_P$. In our previous experiment, we found that T_p increased with decreasing concentration and concluded that if the phase transition occurs in pure crystals, it is at too high a temperature to be observed by our techniques.

The acoustic pulse is modulated by interaction with the traveling heat pulse so that the shifts of α and v result from a convolution of the two pulses. Therefore, an estimate of the velocity of the wave interacting with the acoustic pulse is obtained by comparing a convolution of the bolometer and acoustic signals to the actual observed shifts. Because of the variety of waves that are propagating at different velocities and with different attenuations, only the leading edge of the convolution is useful. It shows clearly whether the leading edge of the bolometer signal is responsible for the leading edge of the shifts of α and v.

In the liquid the convolution arrives far ahead of the actual shifts in α and v, at low T, where the ballistic phonons yield a bolometer signal but have no effect on the acoustic waves. At temperatures where the heat wave is dominated by fully developed second sound, the convolution coincides with the observed shifts of α (Fig. 3).

Figure 4 shows a similar comparison of the convolution with data for an impure crystal. The shifts in α and v arrive

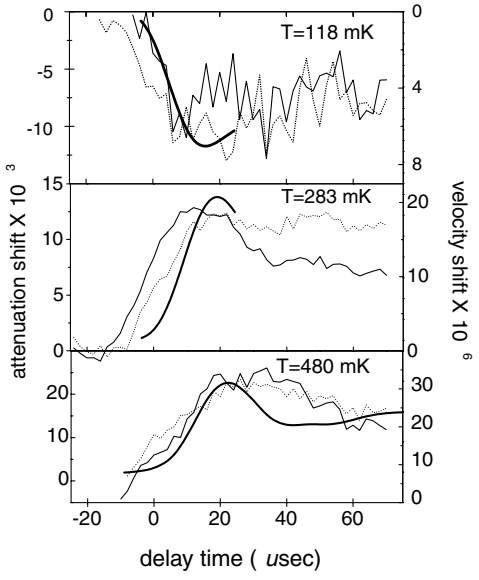

FIG. 4. Shifts of acoustic attenuation and velocity due to interactions with heat pulses in impure solid ^{4}He, compared to the convolution of the bolometer signal. The acoustic shifts occur close to or slightly ahead of the times indicated by the convolution even in the low T limit.

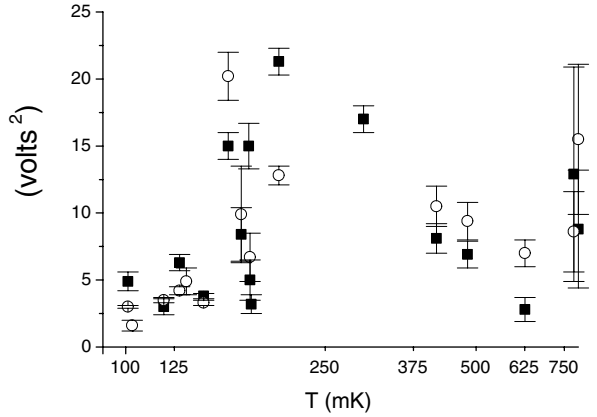

FIG. 5. Values of ϕ from fits of an exponential function to the shifts of α and v vs V_{hp}^2. ■ from attenuation; ∘ from velocity.

at about the same velocity or slightly faster than the convolution of the bolometer signal at all T. If the velocity is the same, then the fast wave interacts with the acoustic waves. If it is faster, then it would indicate that a fast wave which interacts with the acoustic wave is not detected by the bolometer (e.g., it is reflected from and not absorbed by the bolometer surface). If the heat wave propagating at low T were ballistic phonons, as in the liquid, the observed shifts of the acoustic properties would occur later than the convolution of the bolometer signal.

For fixed τ_{delay}, near the peak shifts, and for $T > T_P$, the magnitudes of the shifts as a function of V_{hp} are fit by an exponential function of the heat pulse power so that $\Delta v/v$ and $\Delta\alpha/\alpha \propto \exp[\phi/V_{hp}^2]$. By contrast, for $T < 100$ mK, and at all T in pure crystals the shifts are fit by a linear dependence on V_{hp}^2.

The values of ϕ as a function of T for these data are shown in Fig. 5. ϕ decreases rapidly as T decreases below T_P. The scatter of values near T_P is due to the long time constant for establishing the equilibrium properties at this T and also to slight changes in crystal properties during the two months required to complete all of these measurements.

Conversion of ϕ from units of (volts)^2 to energy units would require knowledge of the effective energy or temperature of the responsible traveling wave as a function of V_{hp}. However, the earliest effect on the acoustics arises from waves that are not detected by the bolometer so that the bolometer signal is not a good measure of their energy. We assume, however, that ϕ must be greater than T at which it is measured.

In summary, interactions between traveling waves generated by heat pulses and sound waves in pure and impure solid ^{4}He have been measured. They reveal a fast traveling wave at low T that appears to be carried by nonphonon excitations. The energy in the heat pulses, which is required to shift the acoustic properties, reveals a gap in the spectrum of these excitations, on the high temperature side of the phase transition discovered in our previous work. All of these results are consistent with our model in which the excitations are long-lived, thermally excited defectons which are Bose condensed when their concentration is sufficiently high. A more detailed description of these results in terms of the model will be published elsewhere; however, additional experiments will be required to prove or disprove the validity of the model. The most decisive experiment would be measurement of the dispersion relation of the nonphonon excitations.

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[1] A. Andreev and I. M. Lifshitz, Sov. Phys. JETP **29**, 1107 (1969).
[2] Y. Imry and M. Schwartz, J. Low Temp. Phys. **21**, 543 (1975).
[3] K. Liu and M. Fisher, J. Low Temp. Phys. **10**, 655 (1973).
[4] G. Lengua and J. M. Goodkind, J. Low Temp. Phys. **79**, 251 (1990).
[5] P.-C. Ho, I. P. Bindloss, and J. M. Goodkind, J. Low Temp. Phys. **109**, 409 (1997).
[6] C. A. Burns and J. M. Goodkind, J. Low Temp. Phys. **95**, 695 (1994).
[7] M. J. Buckingham and W. M. Fairbank, Prog. Low Temp. Phys. **3**, 80 (1961).
[8] V. Narayanamurti, R. C. Dynes, and K. Andres, Phys. Rev. B **11**, 2500 (1975).
[9] V. Narayanamurti and R. C. Dynes, Phys. Rev. B **13**, 2898 (1976).
[10] V. Narayanamurti and R. C. Dynes, Phys. Rev. B **12**, 1731 (1975).