Measurement of anomalous dispersion and the excitation spectrum of He II

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A detailed account of the propagation of superthermal phonons ($\hbar\omega \gg kT$) in He II is presented as a function of pressure up to the solidification point and at low temperatures. Superconducting Al fluorescers and tunnel junctions are used for this study. The measurements show that a well-defined cutoff energy E_c exists, below which the spontaneous decay of a high-frequency phonon via the three-phonon process occurs in He. From measurements of E_c and the group velocity v_g at E_c, we obtain numerical estimates for the magnitude of the anomalous dispersion as a function of pressure. The results show that simple polynomial expansions do not quantitatively describe all the available data and that current models involving two branches in the excitation spectrum describe the situation somewhat better.

I. INTRODUCTION

Over the past few years there has been much discussion over the details of the excitation spectrum of He II in the phonon branch.[1–10] The studies and controversies center on whether the low- to middle-q region (0–0.5 Å^{-1}) is linear, slightly concave up, or concave down. Although on the scale of the other details of the excitation spectrum (rotons, two-roton states, etc.) the differences considered here are small and subtle, they have important implications for the transport properties of He II at temperatures where the phonon branch is dominant (< 0.6 K at saturated vapor pressure). In order to explain discrepancies between theory and experiment in the ultrasonic attenuation at these temperatures, Maris and Massey[1] originally suggested that the phonon branch bent slightly upward, thus allowing the three-phonon process, and obtained better agreement. Since their original suggestion, many investigations—experimental, theoretical, and phenomenological—have ensued attempting to test the validity of this proposal and/or extend the implications to other measurables.

In this paper, we describe our measurements of the propagation properties of superthermal ($\hbar\omega \gg kT$) phonons using superconducting tunnel junctions. We provide direct evidence for the existence of a cutoff energy E_c for spontaneous decay via the three-phonon process and show its dependence and the dependence of the group velocity v_g at E_c, with pressure. These measurements, we believe, provide a strong confirmation of the model of upward dispersion although detailed numerical comparisons with existing schemes still point up some difficulties. In Sec. II we outline the development of the notion of upward dispersion, some of the forms proposed, and the implications of those forms to various experiments. Section III contains a description of the experimental techniques used in this work. Also, a brief description of the relaxation processes involved in a superconductor and the significance of the "double junction" experiment is given. In Sec. IV the results of our measurements and a comparison with various models of upward dispersion are given. Finally, in Sec. V we summarize our results and draw conclusions.

II. ANOMALOUS DISPERSION AND THE THREE-PHONON PROCESS

It has generally been assumed that the excitation spectrum of He II in the phonon branch can be characterized by the polynomial expansion[1,2]

$$\omega(k)=c_0 k(1-\gamma k^2-\delta k^4-\cdots) \ , \qquad (1)$$

where c_0 is the velocity of sound and γ and δ characterize the deviations from linearity (dispersion). A fit to the neutron scattering measurements of Woods and Cowley[3] at the saturated vapor pressure (SVP) yields the parameters $\gamma=(0\pm 2)\times 10^{36}$ g^{-2}cm^{-2} sec^2 and $\delta=(2.4\pm 0.3)\times 10^{75}$ g^{-4}cm^{-4}sec^4. At low k, this then implies linearity or slightly normal (concave down) dispersion. The existing theories of ultrasonic attenuation predicted an attenuation of the form[11]

$$\alpha=\frac{\pi^2}{30}\,\frac{(u+1)}{\rho\hbar^3}\,\frac{(kT)^4}{c_0^6}\times\omega[\arctan(\omega\tau)-\arctan(\tfrac{3}{2}\gamma\langle\vec{k}\rangle^2\omega\tau)] \ , \qquad (2)$$

where ρ is the density, u the Grueneisen constant, τ the thermal phonon scattering time, ω the ultrasonic frequency, and $\langle\vec{k}\rangle$ the average thermal phonon wave vector. This expression is maximized for positive γ when $\omega\tau \gg 1 \gg \frac{3}{2}\gamma\langle\vec{k}\rangle^2\omega\tau$. Experimentally,[8] the ωT^4 dependence appears to be reasonably well obeyed in specific temperature ranges, but the absolute value of the attenuation is greater by as much as a factor of 2. This result led Maris and Massey to predict a *negative* value of γ, thus increasing the upper bound of α by 2 and bringing the predictions into line with experiment.

This original prediction then generated a flurry of experimental and theoretical investigations to verify or expand and modify the notion. Specific heat measurements[4] at SVP were suggestive of a negative value of γ, and from a polynomial fit to the data the number $\gamma = -4.1\times10^{-37}$ g^{-2} cm^{-2} sec^2 was suggested. It was also observed that as a function of pressure, the value for γ became less negative and finally at sufficiently high pressure, became positive. This datum, it was argued, was consistent with neutron scattering measurements of $\omega(k)$ which showed a stronger trend to normal dispersion at higher pressures. It has also been suggested[5] that there is a term linear in k in the expansion (1). This has been shown to be not the case in the ultrasonic regime[9] (~ 90 MHz) by the elegant experiments of Roach *et al.*

Although the differences between anomalous ($\gamma<0$) and normal ($\gamma>0$) dispersion seem small, the transport properties and scattering mechanisms are profoundly influenced. If normal dispersion exists, the lowest-order phonon-phonon scattering mechanism is the four-phonon process (4pp). In order to satisfy energy and momentum requirements four energies and wave vectors are required. On the other hand, a unique type of three-phonon process (3pp) is allowed if the dispersion is linear or upward. In the linear case a 3pp is allowed *only* for collinear processes, i.e., all three phonons are propagating in the same direction. For $\gamma<0$, small-angle scattering (the size of the angle determined by the magnitude of γ) is allowed, which affects the transport properties. This was recognized by Maris, who in a series of three papers[7] employing these concepts, calculated the viscosity due to this 3pp, the velocity shifts and attenuation of ultrasonic waves, and postulated a new type of propagating mode. In the case of linear dispersion ($\gamma=0$) this is a one-dimensional or collinear second sound. Since all scatterings are at zero angle, the second-sound mode has a velocity $\approx c_0$. In addition to the form (1), Maris proposed an alternate form[7] at SVP given by

$$\omega = c_0 k\left(1+\gamma k^2\,\frac{1-(k/k_a)^2}{1+(k/k_b)^2}\right) , \qquad (3)$$

where γ, k_a, and k_b were constants chosen to be consistent with neutron scattering results and the various phenomena he was attempting to describe. He was able to show that with $\gamma\approx10^{38}$ g^{-2} cm^{-2} sec^2, $k_a = 0.542$ Å^{-1}, and $k_b = 0.332$ Å^{-1}, quite good descriptions of the frequency and temperature dependence of the ultrasonic attenuation and ultrasonic velocity shifts could be achieved. In addition, the viscosity measurements of Whitworth[12] were accounted for.

Another aspect of the 3pp, first pointed out by Jackle and Kehr,[2] is the concept of the cutoff momentum. It was pointed out that in the case of an ultrasonic wave ($q\approx0$ compared to thermal phonons), Eq. (1) implies a maximum q that can scatter the ultrasonic wave via the 3pp and still conserve energy and momentum. It is easy to show that this occurs for a wave vector

$$k^2 = -3\gamma/5\delta \ . \qquad (4)$$

Phonons with wave vectors greater than this cannot join in the three-phonon process with ultrasound and must scatter via the much less probable 4pp. This then leads to deviations from simple ωT^4 dependence of attenuation when ω or T is in this energy range. From the pressure-temperature dependences of these deviations they were able to conclude that this cutoff wave vector decreased as a function of pressure, a result in agreement with the earlier specific heat measurements.

In this work we will be primarily concerned with measurements of the cutoff energy for spontaneous decay[13] via the 3pp. Using simple algebraic arguments (involving conservation of energy and momentum), it follows from (1) that the *maximum* wave vector for spontaneous decay is given by

$$k_c^2 = -4\gamma/5\delta \ . \qquad (5)$$

This is the appropriate cutoff for high-frequency ($\hbar\omega \gg kT$) excitations; the difference from (4) comes about because of the restriction of an ultrasonic wave ($k\approx0$) in that case. An important consequence of (5) is that the group velocity v_g of the phonons at k_c is not equal to the sound velocity. Differentiation of (1) yields

$$v_g = \frac{d\omega}{dk} = c_0(1-3\gamma k^2 - 5\delta k^4) \ .$$

Substituting (5) into this gives

$$v_g(k_c) = c_0(1+\gamma k_c^2) \ . \qquad (6)$$

For anomalous dispersion $\gamma<0$ and $v_g(k_c)<c_0$. Hence combining (6) with (5) and the measured values of $v_g(k_c)$ and k_c one can obtain estimates of γ and δ within the framework of this model. For values of $k<(-\frac{3}{5}\gamma/\delta)^{1/2}$ the group velocity is, of course, greater than c_0 and ω and k dependent. A measurement of the group velocity would certainly be a direct and clear test of these models. Unfortunately, because the 3pp is allowed at these lower energies, the mean free path of phonons in this energy range is very short and so time-of-flight velocity measurements are not possible. In an earlier paper,[10] the present authors attempted these direct measurements; due to an unsuspected phenomenon in the device used to generate these high-frequency phonons, erroneous results suggesting linear dispersion resulted. This aspect will be discussed in later sections.

Indirect measurements, and the analyses of these measurements suggested that upward dispersion indeed existed. The explanations by Maris of the transport properties, the Jackle and Kehr cutoff model, the specific heat,[4] heat pulse measurements,[14] and the pressure dependence of these measurements, phonon focusing,[15] phonon interference,[6] the ultrasonic measurements, and x-ray measurements[16] all seemed to suggest this form of dispersion. Various phenomenological expressions, usually of the expansion form of Eq. (1), were proposed, each designed to describe the results of a particular experiment. There seemed to be no general agreement on the form of the expansion or the magnitude of the dispersion, or in fact whether it had been conclusively shown that there was indeed anomalous dispersion.

Recently, Bhatt and McMillan[17] pointed out that it is necessary to calculate the form of the dispersion self-consistently. Starting with an expansion of the form (1) and the simplest continuum model Hamiltonian due to Landau, they calculated the first-order correction self-consistently to the free-particle propagator due to the 3pp. This, then, yields a spectral weight function $S(k, \omega)$ which is still approximately a δ function in ω above k_c and hence is capable of propagating macroscopic distances. Below k_c, however, they showed that $S(k, \omega)$ is quite complicated, displaying two separate propagating modes which dominate for different values of k. The remnant of the mode represented by (1) becomes severely broadened for any finite value of q, and *a* separate collective mode splits off the bottom of this highly damped mode at k_c. This, it is argued, is the mode observed in the experiments reported here and by neutron scattering experiments. A value for v_g at k_c is calculated and as we will see, agrees favorably with the results, particularly at SVP. The pressure dependence of k_c is in accord with their model, as well, but not the numerical values of v_g at higher pressures.

Recently, Aldrich and Pines[18] have pointed out that because of the nature of the van der Waals interaction between atoms, the power-series expansion for the excitation spectrum should contain both even and odd terms in q. However, they also showed that due to the long-range nature of the forces and the form of the potential, an expansion which describes long-wavelength excitations ($q < 0.1$ Å^{-1}) will not be adequate for higher-q values ($q > 0.2$ Å^{-1}). They then propose *two separate* expressions, valid in different q regions which are compatible with the ultrasonic transport properties, the neutron scattering measurements, and the measurements reported in this work. This is an extension of a notion proposed by Lin-Liu and Woo.[19]

In Sec. IV we will compare our measurements of k_c and v_g as functions of pressure with the various proposed models of the excitation spectrum. In this comparison, it will be clear that although there appears to be some convergence to an accepted model for the $\omega(k)$ relation, some detailed difficulties still exist.

III. EXPERIMENTAL TECHNIQUES

In this section we describe the method of generating phonons of energy $\hbar\omega \gg kT$ using thin-film superconductors. Also, we describe the time-of-flight measurement techniques and the accuracy of those techniques.

It has been shown in detail previously[20] that when a superconductor is excited via quasiparticle tunneling or by an incident heat pulse, the spectrum of phonons emitted from that superconductor has a large peak at the superconductor energy gap 2Δ. In the case of a thermal distribution of phonons incident on a superconducting film (a configuration used in this experiment), the phonons of energy $\hbar\omega > 2\Delta$ are subject to strong attenuation via Cooper pair breaking. The two excited quasiparticles then relax via a two-step process. First, they relax to the top of the energy gap via the emission of phonons of energy dictated by their excitation energy. Then they recombine with others at the gap edge, emitting a phonon of energy 2Δ. In this fluorescence scheme, phonons of different energies are, of course, also emitted. For thick films and strong electron-phonon coupled systems, such as Sn, those with $\hbar\omega \gg 2\Delta$ are reabsorbed and 2Δ represents not only a peak in the distribution but also a maximum. In the case of Al, the superconductor used in this study ($2\Delta = 0.38$ meV), down conversion is not complete and leads to results described in Sec. IV. If a superconducting tunnel junction of the same material is used as a detector, it operates in a quantum detector mode. That is, phonons with $\hbar\omega < 2\Delta$ are not detected while for those with $\hbar\omega > 2\Delta$ the detector is essentially "black." For complete down conversion, the scheme is essentially monochromatic and, by the application of a parallel magnetic field, tunable.

As was mentioned earlier, a second method of generating high-frequency phonons from a superconductor is via quasiparticle tunneling.[21] By applying a voltage across a superconductor-insulator-superconductor (Al-Al_2O_3-Al in this case) tunnel junction, quasiparticles can tunnel from one film to the other. The maximum energy above E_F that a quasiparticle injected into a film can have is tunable by the applied voltage V and is given by $eV - \Delta$. In the relaxation process, the injected particles relax again to the energy gap edge via phonon emission and the maximum "relaxation" phonon energy available is again tunable and is given by $eV - 2\Delta$. The particles then recombine into the Cooper sea, giving a phonon of energy 2Δ. If a second similar junction is used as a detector (in a fashion described previously) for bias voltages $eV < 4\Delta$, only the recombination phonons will be detected, but at $eV = 4\Delta$ relaxation phonons can have energies up to 2Δ and an enhanced signal S should result. By current modulation and phase-sensitive detection, one can measure dS/dI vs applied voltage, and a step occurs at $eV = 4\Delta$ as this new channel opens up.[20] For biases $eV > 4\Delta$ phonons of energy greater than 2Δ are created and will either be reabsorbed in a strong electron-phonon coupled case (Pb or Sn) or can escape in the case of Al. This feature then affords the additional convenience of a voltage-tunable source of phonons[22] with energies $> 2\Delta$; by modulation techniques, one can study the effects (in our case in He II) of phonons of energy $eV - 2\Delta$. It is this feature which allows us to measure k_c using this technique since the phonons with $k < k_c$ that are injected into the He II are rapidly scattered by the 3pp and are not detected.

Another relaxation mechanism becomes important when total down conversion is not achieved. In the case where there are a substantial number of quasiparticles excited at the gap edge, either thermally or by tunnel injection, particles injected slightly above the gap edge, say ϵ, can recombine directly with a particle at the gap to emit a phonon of energy $2\Delta + \epsilon$. If there is a finite probability for escape from the superconductor, this will result in a phonon of energy $> 2\Delta$, arising from the direct recombination, which will be detected.

The experiments were performed in an epoxy cell attached by a copper rod to the mixing chamber of a dilution refrigerator. Temperature was measured by a carbon thermometer, previously calibrated against a cerium-magnesium-nitrate thermometer and pressure was applied by a means of a Hoke high-pressure regulator via the fill capillary. Superconducting coax lines were used throughout for transmission of pulse and signal and these were soldered to contact pins in the Teflon holder encasing the generator and detector. The holder was designed to be free of wall reflections, and experiments were performed for propagation distances of ~ 2.5 and 0.7 mm. Data collection was achieved via a transient recorder, and in all cases in this work, the detectors used were Al-oxide-Al tunnel junctions on glass substrates. The Al films were ≈ 600–1000 Å, which is comparable to the penetration depth and hence allowed magnetic tuning of 2Δ.

Two types of generators were used. In the first type, an Al film was pumped by a heat pulse generated by a constantan film of $\approx 3 \times 3$ mm^2 and the fluorescent nature of the Al film was employed. In this case typical power densities dissipated ranged from 1 to 50 mW/mm^2 but above 5 mW/mm^2 amplitude effects were observed. Power densities were kept below this value when quantitative measurements were taken. The second type of generator was a tunnel junction, and quasiparticle injection was used to generate the 2Δ phonons. Because the generator in this case had a normal-state resistance of 2.25 mΩ, typical powers dissipated in this case were $\lesssim 0.5$ mW/mm^2. In all cases, pulse widths were typically 0.2 μsec.

Tunnel junctions used for generation and detection must be of high quality. All devices used in this study had leakage currents (nontunneling) which at the lowest temperatures measured were more than three orders of magnitude lower than the tunneling current at equivalent bias. Also, it was necessary that the detector junctions be of sufficiently low resistance that RC time constant problems be avoided in detection.[23] An order of magnitude estimate suggests that Al junctions 1×1 mm^2 must have normal-state resistances less than 20 mΩ for time responses ≈ 0.1 μsec. Otherwise long tails in the signals were observed and the pulse shapes were not representative of the arriving signal.

Since the absolute value of the group velocity measurement was important in this experiment, it was imperative that an accurate determination of the propagation length be made. It was found that the most reliable method of obtaining this value was by calibrating the cell via careful measurements of the second-sound velocity in He II at ~ 1.10 K. In this temperature range, heat pulses propagate via second sound, and the velocity of second sound at SVP is not overly temperature dependent and is 18.3 m/sec. Precise determination of pulse arrival times and times between echos afford a simple but accurate method of determining cell dimensions to better than 0.3%. This error constitutes $\simeq \frac{1}{3}$ of the error quoted in v_g in the next section.

IV. RESULTS AND DISCUSSION

In the early stages of this experiment, attempts[10] were made to measure the postulated upward dispersion directly by group velocity determinations at $T \sim 0.1$ K at SVP. Magnetic fields applied parallel to the Al films were used to tune the energy gap and hence, it was believed,[20] the frequency of phonons injected into the helium. As a generator, the thin-film heater–Al-film fluorescer was used. Precise calibrations of the cell dimensions were not made since we were looking for relative changes, and so it was assumed that in the low-frequency limit the velocity was c_0. No variation with energy gap was observed. It was concluded

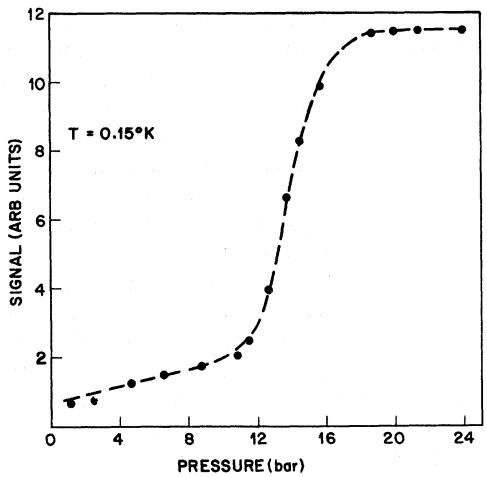

FIG. 1. Plot of signal obtained at the tunnel junction detector as a function of pressure. The fluorescent generator was used in this case. This is a reproduction of the data presented in Ref. 10.

that the dispersion was linear ($\gamma = 0$) over the frequency range considered at SVP. Two strong objections arose to this experiment. First, if the dispersion was indeed linear, the 3pp was operative and all estimates of phonon lifetimes in this frequency range suggested that the phonons should be short lived and not observed. Second, and connected to the first objection, it had not been conclusively shown that the spectrum generated was indeed "monochromatic" or that the detector junctions operated in a quantum fashion. Various suggestions were made as to why the signal was observed in this experiment, and it was vital that these various suggestions be investigated.

In addition, in this earlier work, the pressure dependence of the signal strength (peak height) was plotted and the rapid change in the vicinity of 12–14 bar was interpreted as an attenuation edge, consistent with the onset of normal dispersion at high pressures. These data are reproduced in Fig. 1, where we see a sharp rise in signal at 13 bar. Fitting these data to an exponential form, we estimated the limiting mean free path at SVP to be 0.8 mm. The cell length in this investigation was estimated to be 2.68 mm. After this paper was published, the experiment was repeated for a propagation distance of 0.7 mm, which was less than the estimated mean free path at SVP. The results of the experiment were identical to those illustrated in Fig. 1 and thus indicated that this was not a measure of absorption but must reflect some other pressure-dependent property. As it was clear that the experiment was not operating in the fashion originally conceived (at low pressures), it became vital to determine the spectrum of phonons being generated and more explicitly to know whether phonons of energy $2\Delta = 0.38$ meV $= 4.42$ K were propagating across the sample space.

To investigate this point an Al-oxide-Al tunnel junction was used as a generator, and attempts to measure the structure associated with the spectrum emitted from the tunnel junction were performed at 0.1 K and at several pressures. This experiment and the results of it were described in an earlier publication.[13] The current I through the generator junction was modulated at low frequency and the differential signal at the detector dS/dI vs operator bias V was plotted. The results are illustrated in Fig. 2 for various pressures and a propagation length of 0.7 mm. At high pressures where from pulsed measurements and neutron scattering measurements the dispersion is believed to be normal, we expect the 3pp to be inoperative. We thus expect phonons of energy 2Δ to be long-lived, and this is indeed observed. The curve labelled 24 bar gives clear evidence for generation, propagation, and detection of these phonons. Both the steps at $eV - 2\Delta = 0$ (recombination phonons) and $eV - 2\Delta = 2\Delta$ (relaxation phonons) indicate long-lived phonons at 2Δ. The peak in dS/dI at 0.05 meV is most likely due to direct recombination, as discussed earlier. If this is the

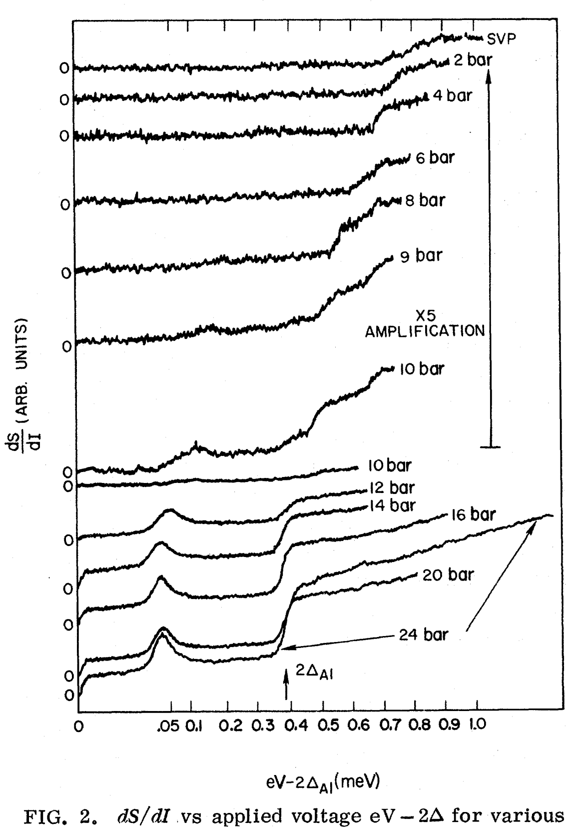

FIG. 2. dS/dI vs applied voltage $eV - 2\Delta$ for various pressures. There is a change of 5 in amplification at pressures below 10 bar.

case, this indicates that phonons of energy $\hbar\omega > 2\Delta$ are escaping and not totally reabsorbed. This is consistent with the observation that in stronger electron-phonon coupled materials, where reabsorption is expected, this peak is substantially smaller or not observed. At energies higher than 2Δ, upon further amplification we observe an additional onset which we ascribe to roton generation and propagation. This will be discussed later in the section.

These data, then, clearly show that at 24 bar phonons of energy 0.38 meV have mean free paths greater than 0.7 mm. From our pulsed measurements, we have determined that these phonons have a group velocity $v_g = 329$ m/sec. Compare that with the measured sound velocity $c_0 = 363$ m/sec and it is clear that we are in the region of normal dispersion; consequently, no 3pp. As we reduce the pressure through 10 bar, the steps at 0 (2Δ recombination phonons) and 0.38 meV (2Δ relaxation phonons) decrease rapidly, indicating attenuation of these phonons. Upon further reduction of the pressure, the 2Δ step begins to move to higher energies, which implies that phonons of energy $>2\Delta$ are escaping the film also and that those of energy less than the step are strongly scattered. Also consistent with this interpretation, the peak ascribed to direct recombination moves to higher energy in a quantitatively similar manner. These curves, then, for pressures between 0 and 10 bar give us a direct measure of the cutoff energy E_c at which the 3pp is no longer operative. Above this energy E_c the generated phonons are long-lived and observed in this experiment.[13,24,25] Below E_c, they are subject to the 3pp and are rapidly scattered. At SVP, we see that $E_c = 0.8$ meV $= 9.5 \pm 0.4$ K. This corresponds to phonons of wavelength 12 Å propagating macroscopic distances while longer-wavelength phonons are scattered. These results indicate that our earlier measurements[10] (Fig. 1) at low pressures were not of phonons of energy 2Δ, but of excitations of energy $E_c > 2\Delta$ until pressures of ≈ 12 bar,[13] at which time the system operated as expected. The signal-vs-pressure curve of Fig. 1 simply reflected the number of phonons of energy greater than E_c which were not down converted by the Al film. This number, of course, increases with increasing pressure because (a) E_c decreases, allowing a wider energy range through and (b) the attenuation in the Al film is also ω dependent. It has been suggested recently[25] that the exponential-like behavior of Fig. 1 can be explained by a model in which efficient down conversion still occurs, but the quasiparticles thermalize at the gap edge to a temperature T_{qp} and recombine from this distribution, thus yielding a high-energy exponential tail in the distribution reflecting this temperature. As the pressure is increased, E_c is decreased and more of this tail is detected. We argue against this model on two counts. Firstly, it is observed that below a certain power density the curve of Fig. 1 is power independent, while T_{qp} would surely depend on power dissipated at all levels. Also, quantitatively, for a cutoff energy ≈ 10 K, a quasiparticle gas temperature ≈ 1 K is necessary, which seems high. Secondly, if complete down conversion occurred, it would be difficult to imagine how this thermal model would produce the steps in dS/dI that vary in such a well-defined way with pressure and measure, we argue, E_c and k_c. Finally, assuming that our description of incomplete down conversion is correct, one can semiquantitatively explain the difference in the ratios of signal observed (in the power-independent regime) at SVP and 24 bar in the experiment by Wyatt *et al.*[25] and this by a simple attenuation model. Assuming on average that a phonon created in the relaxation process must travel one film thickness before escaping into the helium, and taking the film thicknesses quoted in the two experiments and the signal ratios quoted, we determine that the mean free path for a phonon of energy $E_c = 10$ K is approximately 200–400 Å. This is about one order of magnitude less than would be estimated by a free-electron calculation of the mean free path. But in view of the simplicity of the argument (no assumptions made about surfaces and reflection coefficients) and the uncertainties of film thicknesses, the agreement is considered reasonable. It should be noted that the explanation given for the peak in dS/dI at 0.05 meV at 24 bar is not dissimilar to this thermalization argument, and we believe this is happening to some extent. We do not believe, however, that it is responsible for the steps observed at the lower pressures, which we think, for the arguments given above, are due to incomplete down conversion. This is also consistent with work on phonon propagation in solids using Al junctions by Forkel, Welte, and Eisenmenger.[22]

From the data of Fig. 2 one can determine E_c as a function of pressure up to 12 bar. Also, within the small corrections of nonlinear dispersion one can, via the measurements of c_0, determine the critical wave vector k_c. Our estimates of E_c and k_c as functions of pressure are shown in Fig. 3. The points at pressures greater than 12 bar were obtained by magnetically tuning 2Δ, which will be described shortly. It is interesting to note the almost linear dependence of k_c on pressure, a dependence assumed by investigators in the past. We also point out, that when renormalized by $\frac{3}{4}$, these numbers are in reasonable agreement with the estimates of Jackle and Kehr[2] from ultrasonic data.

In Fig. 4 we show the temperature dependence of dS/dI vs V for the data obtained at 24 bar. It is seen qualitatively from these data what happens

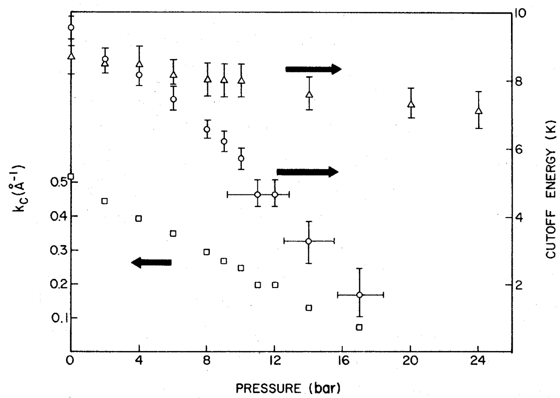

FIG. 3. Cutoff energies and k_c as a function of pressure. E_c vs pressure are the circles; k_c the squares; and the signals ascribed to roton generation and propagation are indicated by the triangles.

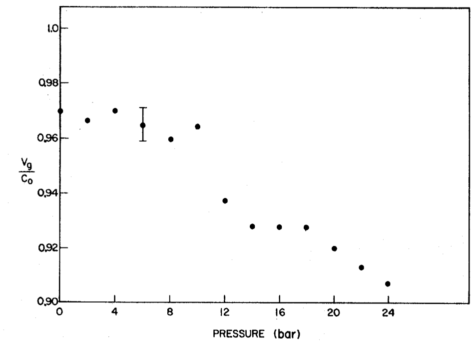

FIG. 5. v_g/c_0 as a function of pressure. This ratio is clearly <1 at low pressures. Above 10 bar the influence of normal dispersion is observed.

when the phonon mean free path becomes less than the propagation length. The detailed structure corresponding to the various mechanisms discussed smear out above ~0.3 K and a smooth rising curve, reminiscent of thermalization effects, is observed. This curve gives us greater confidence that at the lower temperatures (~0.1 K) the results are consistent with our models.

As was described in Sec. II, in addition to measurements of k_c, determinations of the group velocity $v_g\,(d\omega/dk)$ at k_c are very critical in determining the form of the dispersion. As a general result, if $v_g = c_0$, the dispersion can be expected to be linear as the first break from linearity defines k_c. On the other hand, the conditions that $v_g < c_0$, and a k_c is measured, imply that at a lower k, v_g has been greater than c_0 and hence upward dispersion exists. This is, we feel, a general result independent of model. Invoking the model of Eq. (1), it was shown [Eq. (6)] that $v_g = c_0(1+\gamma k_c^2)$. We have measured v_g as a function of pressure, and the results are shown in Fig. 5. Here we see that over the entire pressure range, we have $v_g < c_0$. Up to 10 bar this is, we know from Fig. 2, a measure of $v_g(k_c)$ and hence implies positive dispersion. From (6) and (5) we estimate the parameters γ and δ, which are given in Table I. The value obtained for $|\gamma|$ at SVP is substantially lower than that required by Maris to describe transport properties and is more in line with the estimates of Bhatt and McMillan. A more complete comparison of various models will be made below. An especially disturbing feature of this analysis is the result that as the pressure is increased, the parameter describing the anomalous dispersion γ also increases negatively. k_c is reduced by an even more rapid increase in δ. It is difficult to imagine that this is really happening as it predicts stronger anomalous dispersion (for low k) at higher pressures. We believe that this unexpected dependence reflects the fact that the polynomial model of $\omega(k)$ does not adequately reflect the physics of the situation for large k ($\gtrsim 0.2$ Å) and the more recent attempts invoking more complicated descriptions are on the right track. Above 10 bar, the data of Fig. 5 reflect simply the normal dispersion as observed by neutron scattering measurements. For example, at 24 bar, we measure the group velocity v_g(0.38

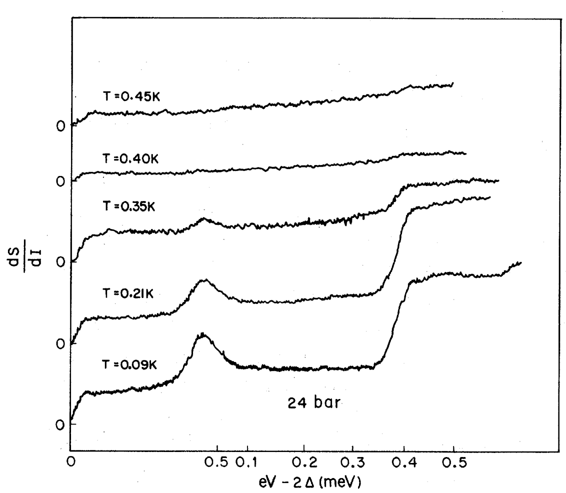

FIG. 4. Temperature dependence of dS/dI vs V at 24 bar presented in Fig. 2. It is clear that at 0.5 K thermal scattering (the 4pp) dominates.

TABLE I. Values of γ and δ derived from measurements of k_c, v_g, and Eq. (5) and (6).

Pressure (bar)	γ(Å^2)	δ(Å^4)
0	−0.12	0.36
2	−0.17	0.69
4	−0.19	0.97
6	−0.29	1.89
8	−0.49	4.66
10	−0.58	7.86

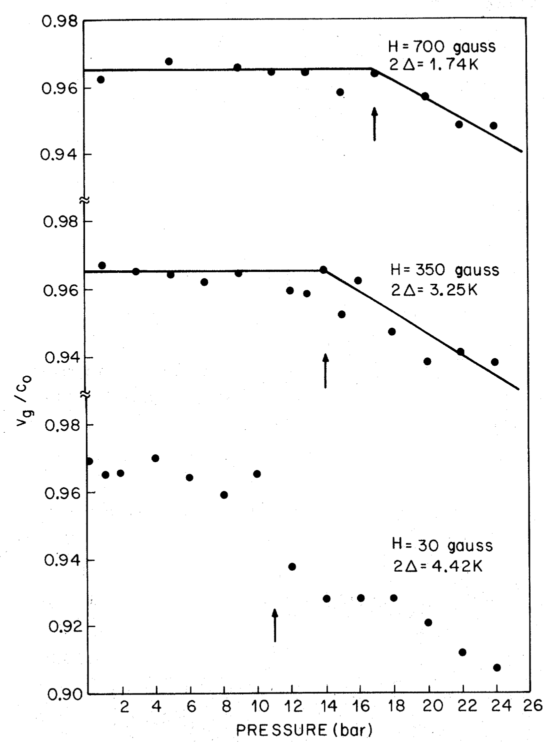

FIG. 6. v_g/c_0 for various tuned energy gaps including the data shown in Fig. 5. The arrows indicate our choice of the cutoff pressure.

meV) to be 329 m/sec. The values of v_g reported here differ from those of Lockerbie *et al.*[26] We believe this difference arises because the latter authors normalized their SVP data to the sound velocity as we had done originally.[10] However, our new length calibrations and measurements of v_g when the superconductor goes gapless (when v_g approaches the heat pulse value of 236 m/sec, see below) convince us that the numerical values are correct within about 2 m/sec.

As was mentioned earlier, these experiments were repeated with an applied magnetic field in an attempt to tune the energy gap 2Δ and hence measure k_c and E_c at higher pressures. In Fig. 6 we show the data obtained from these measurements for three values of 2Δ measured from the *I-V* characteristics of the tunnel junction detector. Again we see the break in the ratio v_g/c_0 at the point in pressure where E_c becomes less than 2Δ; our choice for those critical points are illustrated by the arrows. It is from these data that the three points at 11, 13, and 17 bar in Fig. 3 are derived. Again at higher pressures in Fig. 6 we see the results of normal dispersion.

Plots similar to Fig. 1, but for different gap energies 2Δ, are illustrated in Fig. 7. Here we see two interesting aspects. Firstly, with decreasing 2Δ, the sharp rise in the signal moves to higher pressure. This is consistent with the results of Fig. 6. The values obtained by these two different methods agree within error. Secondly, we observe that the ratio of the signal at 24 bar to SVP becomes less and approaches 1 at the lowest value of 2Δ. Although we do not understand this at the moment, it is consistent with the observation that at smaller values of 2Δ it is more difficult to obtain these curves independent of power dissipated in the pulse. For higher pulse powers at zero field, one can obtain a curve similar to the one labeled $2\Delta = 2.67$ K. This behavior is reminiscent of the pressure dependence of our heat pulse measurements[14] where heater and bolometer respond to a broad-band spectrum of phonons and suggests that the magnetic field on the superconductor destroys the quantum nature of the device. However, this is probably not the case since the velocities measured in these experiments do not approach those measured in the heat pulse case until the superconductor becomes gapless. At that point v_g rapidly changes to the value measured in the heat pulse experiments.

The various models proposed for the excitation spectrum can be compared with our experimental measurements of k_c and $v_g(k_c)$. The models of which we are aware and the predictions of k_c and v_g that can be made are listed in Table II. It is clear from this table that there are several proposed schemes, some of which do better than others in this regard. We also point out, that in addition to the predictions at SVP, any successful model should be capable of explaining the pressure dependence of k_c and the pressure dependence of v_g/c_0. For example, using the measured k_c, the

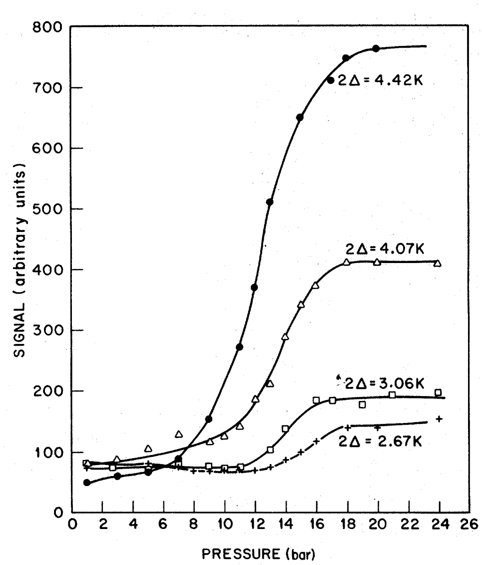

FIG. 7. Signal as a function of pressure for various energy gap values. Propagation length in this case is ~0.7 mm.

TABLE II. Comparison of the predictions of various models for $\omega(k)$ with experimental values obtained in this investigation at SVP. This table includes only models which are appropriate, i.e., several investigations quote only leading terms in the expansion (γ in our notation) and not δ. This type of expansion would not predict cutoffs and v_g values. Experimental values are $k_c = 0.51 \pm 0.02$ Å^{-1}, $v_g = 231 \pm 1.5$ m/sec.

Model	Predicted k_c (Å^{-1})	v_g at predicted k_c (m/sec)	v_g at measured $k = 0.51$ Å^{-1} (m/sec)
Model C	0.43	229	207
Model D (Ref. 7) [Eq. (3)]	0.42	221	206
Molinari and Regge (Ref. 5)	0.29	239	201
Brooks and Donnely (Ref. 27)	0.43	217	199
Lin Liu and Woo (Ref. 19)	0.51	229	229
Bhatt and McMillan (Ref. 17)	Measured k_c used as parameter in model		228
Jackle and Kehr (Ref. 2) [Eq. (1)]	0.49	212	202
Aldridge and Pines (Ref. 18)	0.5	203	203

model of Bhatt and McMillan appears, from Table II, to successfully determine v_g at SVP. However with increasing pressure, it is predicted that v_g/c_0 should approach 1 at ≈ 20 bar, which is not observed. The model of Aldrich and Pines quite successfully determines the pressure dependence of k_c. On the other hand, v_g determinations are outside our errors. These authors do warn, however, that reducing their calculations to a polynomial expansion is not meaningful and could lead to errors of this nature. It should also be pointed out that the expression of Lin-Liu and Woo appears to predict quite accurately the experimental values at SVP. This expression, it is argued, is consistent with x-ray scattering and neutron scattering and should be most accurate in the region 0.3 Å^{-1} $< q < 0.7$ Å^{-1}. The message to be gotten from this table is that there is a general qualitative agreement at this stage as to the nature of the dispersion in $\omega(k)$ and that a cutoff of the 3pp exists. However, no model has yet been shown to explain quantitatively the transport data, the neutron scattering measurements, and the determinations of k_c and v_g over the entire pressure range. The general feeling that seems to be emerging is that if polynomials (or some sort of analytic expressions) are to be used to describe $\omega(k)$, it is possible that different regions of k must be treated differently and that possibly two different types of excitations (low k and high k) could be considered possible.

From this experiment there are some still unmentioned implications. The fact that it has been shown that phonons of energies up to 10 K can be generated in Al films and be transmitted into He II at low temperatures makes at least qualitative statements about Kapitza resistance models and measurements. The wavelength of a 10 K phonon in He II is ≈ 12 Å, which is only a few atomic spacings. Various models and measurements of Kapitza resistance have suggested things like rapid mode conversion at the interface, density gradients, damage layers, phonons emitted only at normal incidence, etc. We suggest that from these and similar measurements, there is now clear evidence that at least some portion (and probably a substantial percentage) of high-frequency excitations are transmitted through the interface in an elastic fashion. We also suggest that perhaps one of the cleanest and most undamaged (although polycrystalline) surfaces available for studies of this nature is that of an evaporated film with its as-grown surfaces exposed to the He. Although we are merely speculat-

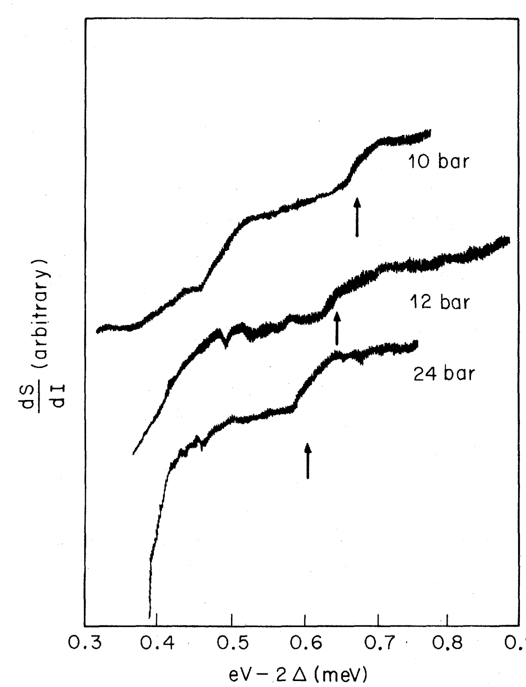

FIG. 8. dS/dT vs $eV-2\Delta$ at higher energies for representative pressures. The steps indicated by the arrows we ascribe to roton generation and propagation and the energy values are given as the triangles in Fig. 3.

ing at this point, the evidence is strong. The use of low temperatures in this study may also be important.

Finally, consistent with this discussion, we point out that in addition to the steps of Fig. 2, from which we measure E_c and k_c, further amplification of dS/dI yields another series of thresholds; some representative examples of the data are shown in Fig. 8. Here at higher pressures, a second, weaker step is consistently observed, which apparently merges with the E_c step at lower pressures. Our best estimates of the pressure dependence of this step are shown as the triangles in Fig. 3. It is seen that at 24 bar, this threshold occurs at ≈ 7.1 K and increases with decreasing pressure. Within error this pressure dependence is the same as that measured by inelastic neutron scattering measurements[28] for the roton branch of $\omega(k)$. In addition, from heat pulse measurements[14,29] we know that phonons from a heater and bolometer do couple to the rotons since propagating rotons have been observed. For this reason, we tentatively assign these onsets to the generation, propagation, and detection of rotons.

This discussion of Kapitza resistance and roton generation is, of course, qualitative in nature, but it is our belief that these results are significant. Further experimentation and investigation of these phenomena is clearly called for.

V. CONCLUSIONS

By generating high-frequency ($\hbar\omega > kT$) phonons using pulsed superconducting films and tunnel junctions, it has been shown that the concept of upward dispersion in the He II excitation spectrum is valid. We have shown that interpretation of our earlier measurements in which we concluded that the dispersion curve was linear at SVP in the region 0–4 K was incorrect, due to incomplete down conversion of higher-frequency phonons. From double-junction experiments, magnetic field tuning, and pulsed time-of-flight measurements, we have measured the pressure dependence of the critical wave vector at which the 3pp cuts off and the group velocity of the phonons at that wave vector. It is found, consistent with other investigations, that the region in k space of anomalous dispersion decreases with increasing pressure, going to zero at approximately 20 bar. In comparing these measurements with postulated forms of the dispersion relation, we conclude that there is general agreement as to the qualitative shape of $\omega(k)$ and to the order of magnitude deviations from linearity. Quantitatively, however, it is felt that no one model describes adequately all the data available: perhaps some of the more complicated descriptions based on physical models involving two branches are getting closer to a proper description.

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