

## High-Frequency Ultrasonic Attenuation in Superfluid Helium Under Pressure\*

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(Received 22 July 1971)

We attribute the formation of a shoulder in the high-frequency ultrasonic attenuation in superfluid helium under pressure to a restriction on the three-phonon process with thermal phonons. The restriction is proposed to follow from the pressure dependence of the phonon dispersion at zero temperature. The steep rise above the shoulder due to phonon-roton and roton-roton interactions also indicates large dispersion effects under pressure.

Recently Roach, Ketterson, and Kuchnir<sup>1</sup> have measured the high-frequency ultrasonic attenuation ( $\alpha$ ) in superfluid helium under pressure ( $P$ ). At low pressures the temperature dependence of  $\alpha$  is approximately described by a  $T^4$  law as under vapor pressure, whereas at higher pressures ( $\geq 10$  atm) the  $\alpha(T)$  curve is drastically changed and a shoulder occurs. They showed that this feature cannot be explained by the known relaxation times arising from the interactions of phonons and rotons since these relaxation times either are too small or do not have the required  $T$  and  $P$  dependence. Therefore they suggested that the shoulder might indicate the existence of a new relaxation mechanism. Rather than using such a hypothesis we show in this Letter that the high-frequency ultrasonic attenuation in He II under pressure can be understood in terms of the known interaction processes between phonons and rotons.<sup>2,3</sup> Our basic idea is that the anomalous behavior of the attenuation results from a deformation of the phonon spectrum under pressure. In this way we obtain information about the pressure dependence of the phonon dispersion from the sound-absorption data.

We point out that the measured anomalous attenuation under pressure is smaller than the theoretical  $\omega T^4$  attenuation derived from the three-phonon process for dispersionless phonons in the collisionless regime (Fig. 1). So, we explain the formation of the shoulder in  $\alpha(T)$  by assuming that the three-phonon process (3PP) is allowed only for very long-wavelength phonons ("partially allowed three-phonon process"). With this assumption we also can describe the rapid increase of  $\alpha$  for  $T$  above about 0.6 K. In this range thermal phonons of shorter wavelength also contribute to the three-phonon process as a

result of the width which increases rapidly because of phonon-roton interactions ("lifetime-induced three-phonon process"). It turns out

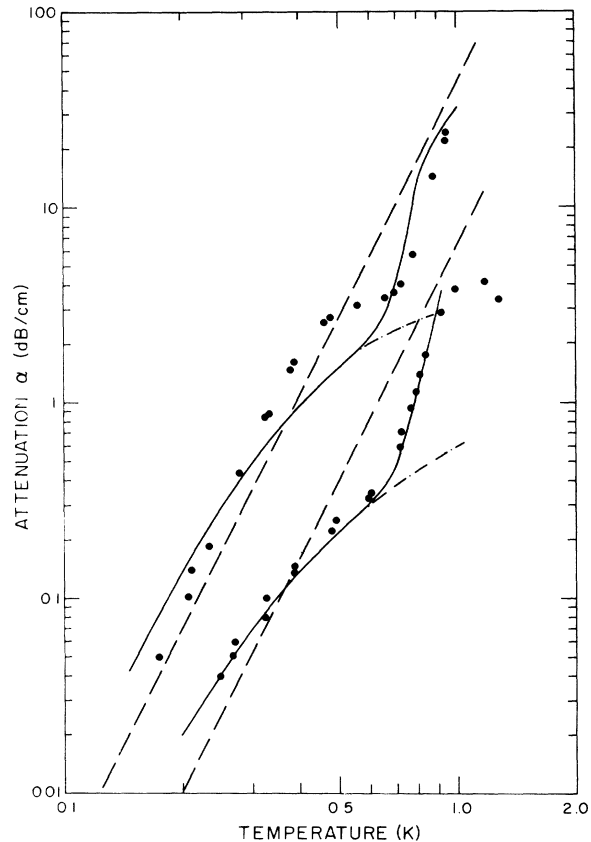


FIG. 1. Ultrasonic attenuation for 16.4 atm. Lower curves for 15 MHz; upper curves for 105 MHz. Solid points, experimental data of Roach, Ketterson, and Kuchnir. Dashed lines, three-phonon result  $\frac{1}{2}A\omega T^4$ . Dashed-dotted lines, partially allowed 3PP. Solid line for 15 MHz, sum of partially allowed 3PP's and lifetime-induced attenuation. Solid line for 105 MHz, the same, plus contribution of the roton viscosity.

that above 0.6 K, contributions to the attenuation from the roton viscosity become important as well.

We assume that a cutoff momentum  $q_c$  exists for the 3PP such that an ultrasonic phonon (with frequency  $\omega$ ) can be absorbed only by thermal phonons with momentum  $q \leq q_c$ . The onset of the shoulder in  $\alpha(T)$  is characterized by the shift of the dominant thermal phonons into the momentum region above  $q_c$  as the temperature is increased. Our assumption is that  $q_c$  varies strongly with pressure. At low pressures (below about 10 atm)  $q_c$  is above the thermal phonon range so that the 3PP is fully effective. At higher pressures  $q_c$  is within the momentum range of the thermal phonons so that the attenuation is reduced and a shoulder appears. With increasing pressure  $q_c$  decreases and the shoulder shows up at lower temperatures. The ultrasonic attenuation by the partially allowed 3PP is given by

$$\alpha(\omega, P, T) = A\omega T^4 F(z)/F(\infty), \quad (1)$$

where  $z = cq_c/(k_B T)$  and  $F(z) = \int_0^z dx x^4 f(x)[1+f(x)]$ ,  $f(x)$  being the Bose function. For low temperatures ( $k_B T \ll cq_c$ ) one obtains  $\alpha = A\omega T^4$ , the well-known three-phonon result<sup>4</sup> (allowing for a factor 2 if the spectrum is concave at long wavelengths<sup>5</sup>). For high temperatures ( $k_B T \gg cq_c$ ) the 3PP is restricted and one has

$$\alpha = \frac{1}{3}A(cq_c/k_B)^3 \omega T. \quad (1')$$

A linear  $T$  dependence of  $\alpha$  on the shoulder is found in various experimental curves at 45 and 105 MHz.<sup>1,6</sup> Note that Eq. (1) depends on the frequency only through the factor  $\omega$ . Thus the  $\alpha(T)$  curves for different frequencies should simply be shifted in proportion to  $\omega$  but not changed in shape. This prediction is rather well confirmed by the curves at 45 and 105 MHz whereas at 15 MHz the curves are considerably different in shape compared with those at 105 MHz.

Assuming a value of 1.60 for  $cq_c/k_B$  at 16.4 atm, we have plotted Eq. (1) for 15 and 105 MHz (Fig. 1). At 15 MHz the experimental points<sup>6</sup> are reproduced, whereas 105 MHz the measured  $\alpha(T)$  shows a more pronounced onset of the shoulder and is larger than  $\alpha$  from Eq. (1'). By fitting  $q_c$  one obtains similar results for the other pressures (Table I).

How can a cutoff momentum  $q_c$  and its pressure dependence be explained? The 3PP is very sensitive to the shape of the phonon spectrum. A thermal phonon can absorb an ultrasonic phonon only when—with an accuracy given by the ef-

TABLE I. Values of the cutoff momentum  $q_c$  obtained from fit.

$P$ (atm)	$cq_c/k_B$ (K)
14.0	2.05
15.5	1.85
16.4	1.60
17.8	1.25
19.0	0.86

fective width of the thermal phonons—its group velocity is not smaller than the ultrasonic phase velocity. We suggest that the cutoff momentum and its  $P$  dependence result from the  $P$  dependence of the phonon spectrum  $\epsilon(q)$  at  $T=0$ , neglecting, in a first approximation, effects of  $T$  renormalization on  $\epsilon(q)$  and effects of the thermal linewidth. With the energy of long-wavelength phonons at  $T=0$  given by

$$\epsilon(q) = cq(1 - \gamma q^2 - \delta q^4 - \dots), \quad (2)$$

the existence of a cutoff momentum is readily explained if  $\gamma$  is negative and  $\delta$  is positive;  $q_c$  is then given by

$$q_c^2 = -3\gamma/5\delta. \quad (3)$$

Inelastic neutron scattering from helium at vapor pressure and 1.1 K gave<sup>7</sup>  $\delta = 2.4 \times 10^{75} \text{ g}^{-4} \text{ cm}^{-4} \text{ sec}^4$ , and an upper bound of  $2 \times 10^{36} \text{ g}^{-2} \text{ cm}^{-2} \text{ sec}^2$  for  $|\gamma|$ , which results in an upper bound of 3.86 K for  $cq_c/k_B$ . The actual value of  $cq_c/k_B$  should not be much smaller since at vapor pressure no reduction of the three-phonon result for  $\alpha(T)$  is observed. Unfortunately there are no neutron data of comparable accuracy available for higher  $P$ .

Recently Phillips, Waterfield, and Hoffer<sup>8</sup> investigated the specific heat  $C_v(T)$  in superfluid helium and found the first correction to the leading  $T^3$  term to be negative below about 6 atm and positive at 20.8 atm. While it is an open question whether the data can be analyzed using the specific-heat formula for noninteracting phonons with a spectrum given by Eq. (2), the results indicate that pressure indeed has a remarkable effect on the phonon dispersion. Phillips, Waterfield, and Hoffer<sup>8</sup> deduced negative values of  $\gamma$  for low  $P$  but positive values of  $\gamma$  for  $P \geq 6$  atm, contrary to our assumption. At  $P=20.8$  atm, a value of  $\gamma$  as large as  $19.6 \times 10^{37}$  is derived. Since the phonon contribution to  $C_v(T)$  could be measured only in a very small temperature interval, the possibility is not excluded that there

is still a negative  $\gamma$  which is dominated over by a larger positive  $\delta$ . Choosing  $\gamma = -3.6 \times 10^{37}$  and  $\delta = 1.8 \times 10^{78}$  we are able to reproduce both the assumed  $cq_c/k_B = 0.86$  K at 19.6 atm and  $C_v(T)$  at 20.8 atm. One then expects, below  $T^2 = 0.016$  K<sup>2</sup>, a negative slope of the curve  $C_v/T^3$  vs  $T^2$  which was plotted in Ref. 8. With such large  $\gamma$  and  $\delta$  a fit of neutron data will not be possible without higher-order terms in  $\epsilon(q)$  [Eq. (2)].

To summarize, the partially allowed 3PP explains qualitatively, at the lowest frequency even quantitatively, the shoulder in the attenuation  $\alpha$  under pressure. The explanation of the cutoff momentum  $q_c(P)$  should be elaborated; effects of the temperature renormalization of the phonon spectrum and of the thermal linewidth should be included especially.

We now discuss the steep rise above the shoulder in the temperature region between 0.6 and 1.0 K. Obviously this rise is connected with the rapidly increasing number of thermally excited rotons. In their extensive treatment of sound propagation in He II, Khalatnikov and Chernikova<sup>9</sup> derived general expressions for the attenuation. From this theory, Roach, Ketterson, and Kuchnir<sup>1</sup> extract a contribution proportional to  $\rho_{np}\tau_{pr}^{-1}$ , where  $\tau_{pr}$  is the phonon-roton collision time<sup>2,10</sup> and  $\rho_{np} \propto T^4$  is the phonon part of the density of the normal component. This is essentially a contribution of the first viscosity arising from the phonon-roton interactions. It is obtained under the condition  $\tau_{pr}^{-1} \ll \omega \ll \tau_{pp}^{-1}$ , where  $\tau_{pp}$  is the phonon-phonon collision time associated with wide-angle scattering. It has the right  $T$  dependence but is much too small compared with experiment. The steep rise of  $\alpha$  occurs in a collisionless regime both with respect to  $\tau_{pr}$  ( $\omega\tau_{pr} \gg 1$ ) and  $\tau_{pp}$  ( $\omega\tau_{pp} \gg 1$ ). In this limit, with allowance for an energy deficit in the 3PP, the expressions of Khalatnikov and Chernikova reduce to

$$\alpha' = \frac{3}{4}(u+1)^2 \frac{\omega}{c} \frac{\rho_{np}}{\rho} \arctan\left(\frac{1}{r\omega\tau}\right), \quad (4)$$

$$\tau^{-1} = \tau_{pr}^{-1} + \tau_{pp}^{-1},$$

where  $r = (c-v)/v > 0$  is the relative difference between the ultrasonic velocity  $c$  and the mean thermal group velocity  $v$ ;  $\alpha'$  is the amplitude attenuation coefficient related to  $\alpha$ , in decibels per centimeter, by  $\alpha = 8.68\alpha'$ ;  $\rho$  is the density; and  $u$  is the Grüneisen constant  $\partial \ln c / \partial \ln \rho$ . We note that this result is identical to that obtained in Ref. 4 in second-order perturbation theory with dressed phonon propagators, provided that the

thermal phonon lifetime is given by the same  $\tau$ .

For  $r > 0$ , Eq. (4) describes the rate of the 3PP which is due to the finite lifetime of the thermal phonons ("lifetime-induced 3PP"). In the case  $r\omega\tau \gg 1$ , in which the energy deficit in the 3PP is large compared to the linewidth,  $\alpha$  rises steeply with  $T$ , in proportion to  $(u+1)^2 T^4 / (r\tau)$ . In Fig. 1 a fit of Eq. (4) to the measured data for 16.4 atm and 15 MHz is shown with  $r = 0.26T^4$ . We used  $\tau_{pr}^{-1} = 1.5 \times 10^{11} T^{9/2} \exp(-7.5/T) \text{ sec}^{-1}$ ,<sup>11</sup>  $\rho = 0.1653 \text{ g cm}^{-3}$ ,  $u = 2.33$ , and  $c = 3.33 \times 10^4 \text{ cm sec}^{-1}$ .<sup>12</sup> Combining this fit of  $r$  with the fit of  $q_c$  (Table I) we can determine the dispersion parameters  $\gamma$  and  $\delta$ . For 16.4 atm we get<sup>13</sup> the rather large values  $\gamma = -1.3 \times 10^{38}$  and  $\delta = 2.2 \times 10^{78}$ . Using this fit of the relative energy difference  $r$  obtained from the 15-MHz data we cannot, however, reproduce the magnitude of the data at the higher frequencies. For example, in order to reproduce the 105-MHz data one would have to assume values of  $r$  which are about 5 times smaller. This discrepancy is related to the fact that the measured attenuation is roughly proportional to  $\omega$ , whereas Eq. (4) is almost independent of  $\omega$  in the region of the steep rise.

We show that at higher frequencies the contribution of the roton part of the viscosity helps to explain the measured data. We consider the impressed sound wave as an external periodic disturbance with a velocity field  $\vec{v}_s(\vec{r}, t)$  shifting the energy of a roton of momentum  $\vec{p}$  by  $\vec{p} \cdot \vec{v}_s(\vec{r}, t)$ . As stated by Khalatnikov<sup>10</sup> the deformation-type coupling is small compared to the  $\vec{p} \cdot \vec{v}_s$  coupling and can be neglected. We have considered the linearized Boltzmann equation for rotons in a conserving-collision-time approximation. We only take the roton-roton collision time  $\tau_{rr}$  into account, which is short compared to the roton-phonon collision time  $\tau_{rp}$ . We find

$$\alpha' = (\omega^2/2\rho c^3) \left(\frac{4}{3}\eta_r + \zeta_{2,r}\right) [1 + (\omega\tau_{rr})^2]^{-1}, \quad (5)$$

where the shear viscosity  $\eta_r$  is given by<sup>2,10</sup>

$$\eta_r = (\rho_0^2/15\mu)n_r\tau_{rr}, \quad (6)$$

with  $\rho_0$  and  $\mu$  the usual roton parameters and  $n_r$  the roton number per unit volume. The second viscosity  $\zeta_{2,r}$  equals  $\frac{2}{3}\eta_r$ , and  $\tau_{rr}$  is determined from the roton part of the measured shear viscosity according to Eq. (6)<sup>10</sup>; so all quantities in Eq. (5) are known.

The result is smaller than the measured attenuation at 15 MHz but of comparable magnitude at 105 MHz. The roton contribution Eq. (5) becomes important compared to the phonon contri-

bution Eq. (4) because the roton lifetime  $\tau_{rr}$  is much smaller than the phonon lifetimes  $\tau_{pr}$  and  $\tau_{pp}$ . For example, at 16.4 atm,  $\tau_{rr}^{-1} = 1.0 \times 10^{13} T^{1/2} \times \exp(-7.5/T) \text{ sec}^{-1}$ , so that at 105 MHz the transition from the high-frequency to the low-frequency regime, marked by  $\omega\tau_{rr} = 1$ , occurs at a temperature as low as 0.78 K. In Fig. 1 we have plotted for 105 MHz the sum of the partially allowed 3PP's, the lifetime-induced absorption Eq. (4), and the contribution of the roton viscosity Eq. (5). The theoretical curve even exceeds the measured points and shows a bendover around the temperature where  $\omega\tau_{rr} = 1$ .

Further improvement between theory and experiment in the region between 0.6 and 1.0 K will probably only be achieved by treating the coupled system of the Boltzmann equations for the photons and the rotons. The main conclusion we want to draw from the region of the steep rise is the indication for large energy deficits for the three-phonon processes in this range under pressure. Those large energy deficits should be the result of large dispersion of the phonon spectra under pressure. This can be tested by direct measurements of  $\epsilon(q)$ ; thus neutron-scattering measurements and high-frequency sound-velocity measurements under pressure are most desirable.

We should like to thank G. V. Chester, C. P. Enz, J. B. Ketterson, J. A. Krumhansl, and P. N. Trofimenkoff for discussions. We are very grateful to P. R. Roach, J. B. Ketterson, and M. Kuchnir for having provided us, prior to publication, with data on the attenuation under pressure at different frequencies, and to O. W. Dietrich, L. Passell, E. Graf, and C. H. Huang for

having communicated to us neutron-scattering results on the roton spectrum under different pressures.

\*Work supported in part by the National Science Foundation through Grant No. GP-27355.

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## Spectral-Function Sum Rules for Hard-Core Potentials\*

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(Received 6 July 1971)

It is shown that the third frequency moment of the dynamic density-density correlation function  $\chi''(k, \omega)$  diverges for quantum and classical systems with hard-core potentials. Some information is obtained about the leading term of the divergence in the hard-core limit.

The dynamic density-density correlation function, or spectral function  $\chi''(k, \omega)$ , is defined by<sup>1</sup>

$$\chi''(k, \omega) = (1/\rho) \int dr dt e^{-i(kr - \omega t)} \langle [\rho(r, t), \rho(0, 0)] \rangle, \quad (1)$$

where  $\rho(r, t)$  is the number-density operator for a system of  $N$  particles in a volume  $\Omega$ , given by

$$\rho(r, t) = \sum_{i=1}^N \delta(r - r_i(t)) = \psi^\dagger(r) \psi(r), \quad (2)$$