are underway.

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¹R. T. Hodgson and R. W. Dreyfus, Phys. Rev. Lett. 28, 536 (1972); W. Waynant, Phys. Rev. Lett. 28, 533

(1972).

²M. A. Duguay and P. M. Rentzepis, Appl. Phys. Lett. 10, 350 (1967).

³S. E. Harris and R. B. Miles, Appl. Phys. Lett. <u>19</u>, 385 (1971).

⁴J. F. Young, G. C. Bjorklund, A. H. Kung, R. B. Miles, and S. E. Harris, Phys. Rev. Lett. <u>27</u>, 1551 (1971).

⁵R. B. Cairns, H. Harrison, and R. I. Schoen, J. Chem. Phys. 51, 5440 (1969).

⁶C. R. Vidal and J. Cooper, J. Appl. Phys. <u>40</u>, 3370 (1969).

⁷J. F. Ward and G. H. C. New, Phys. Rev. <u>185</u>, 57 (1969).

⁸R. B. Miles and S. E. Harris, "Optical Third Harmonic Generation in Alkali Metal Vapors" (to be published).

Thermodynamic Evidence Against a Quadratic Term in the Phonon Spectrum of Superfluid Helium*

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A quadratic term, characterized by the coefficient α_1 , in the phonon spectrum $\epsilon(k) = c\hbar k (1 - \alpha_1 k - \alpha_2 k^2 - \cdots)$ is shown to be inconsistent with data on the low-temperature specific heat of liquid He II. Consistency obtains if α_1 is set identically equal to zero.

The continuing controversy over the analytic form of the low-momentum excitation spectrum of liquid He II was initiated by the suggestion of Maris and Massey¹ that the coefficient γ in the expression

$$\epsilon(p) = cp(1 - \gamma p^2 - \delta p^4 - \cdots), \tag{1}$$

which was generally believed to be positive, may in fact be negative. This would allow a reconciliation of the experimental data on the attenuation and velocity of sound in liquid He II with the current theories of superfluidity. Further evidence for negative γ has come from the analysis of lowtemperature specific-heat data² and from x-ray scattering measurements.3 In the meantime, Feenberg⁴ has shown that in the case of a dilute, weakly interacting Bose gas, if the interatomic potential falls off asymptotically as $(1/r)^6$, then the energy spectrum of elementary excitations may contain both odd and even powers of p-with the exception of a term in p^2 . On the other hand, Molinari and Regge, 5 who analyzed the neutron scattering data of Woods and Cowley⁶ to test various analytic properties of the phonon dispersion curve, have concluded that a p^2 term may as well be present. So, quite generally, one may write

$$\epsilon(k) = c\hbar k (1 - \alpha_1 k - \alpha_2 k^2 - \cdots). \tag{2}$$

The conclusion of Molinari and Regge has been supported by the recent work of Anderson and Sabisky, who have studied dispersion in the phase velocity of first sound in superfluid helium at 1.38 K in the frequency range 20–60 GHz; their value of α_1 agrees favorably with the one obtained by Molinari and Regge, viz., 0.27 Å. Most recently, however, Roach et~al.8 have carried out a direct measurement of the velocity of 30- and 90-MHz sound waves in helium below 0.1 K and have shown that the value of α_1 cannot be larger than 0.01 Å. Accordingly, the controversy has now shifted from the cubic to the quadratic term in the spectrum.

To help resolve this controversy we undertook a re-examination of the specific-heat data of Phillips, Waterfield, and Hoffer² by including a nonzero quadratic term in the phonon spectrum and comparing the resulting values of the various parameters of the excitation spectrum with the values obtaining from other, more direct, sources (such as Abraham $et\ al.^9$ and Donnelly¹⁰). In particular, we studied the density dependence of these parameters. It turns out that the inclusion of a quadratic term in the phonon spectrum leads to a set of parameters whose behavior, as a function of density, is too erratic to be acceptable.

On the other hand, setting this term identically equal to zero leads to a set of parameters whose behavior, as a function of density, is in line with the one expected on the basis of the aforementioned sources. We regard this as evidence in favor of the findings of Roach $et\ al.^8$ and, hence, against the presence of a quadratic term in the phonon spectrum of superfluid helium.

Following the customary approach, 11,12 we obtain for the phonon specific heat

$$C_{V_{\bullet} ph} = A_0 T^3 + A_1 T^4 + A_2 T^5 + \cdots,$$
 (3)

where

$$A_{0} = \frac{Vk_{B}^{4}}{2\pi^{2}c^{3}\hbar^{3}} 4! \zeta(4), \quad A_{1} = \alpha_{1} \frac{2Vk_{B}^{5}}{\pi^{2}c^{4}\hbar^{4}} 5! \zeta(5),$$

$$A_{2} = (\alpha_{2} + 3\alpha_{1}^{2}) \frac{5Vk_{B}^{6}}{2\pi^{2}c^{5}\hbar^{5}} 6! \zeta(6);$$
(4)

here, $\zeta(n)$ is the Riemann ζ function while the other symbols have their usual meanings. The roton contribution is given by

$$C_{V, \text{rot}} = RT^{1/2} \exp(-\Delta/k_B T)$$

 $\times [(\Delta/k_B T)^2 + \Delta/k_B T + \frac{3}{4}],$ (5)

where

$$R = V(2 p_0^2 / \mu) (\mu k_B / 2\pi \bar{h}^2)^{3/2}.$$
 (6)

The combination of (3) and (5), in the form

$$C_{V}/T^{3} \simeq A_{0} + A_{1}T + A_{2}T^{2} + RT^{-5/2}\exp(-\Delta/k_{B}T)$$

$$\times \left[(\Delta/k_{\rm B}T)^2 + \Delta/k_{\rm B}T + \frac{3}{4} \right], \tag{7}$$

was fitted to the specific-heat data of Phillips, Waterfield, and Hoffer, taken at four densities (corresponding to the molar volumes 27.58, 27.11, 26.23, and 23.79 cm³). We carried out a five-parameter fit with the unknowns A_0 , A_1 , A_2 , R, and Δ/k_B , as well as a four-parameter fit with the unknowns A_0 , A_2 , R, and Δ/k_B (with $A_1 \equiv 0$). The latter was essentially a repeat of the analysis carried out by Phillips, Waterfield, and Hoffer and provided a check on ours; the former was, of course, intended to detect the presence of a quartic term in C_v which arises from the quadratic term in the phonon spectrum. The results of our analysis are shown in Figs. 1 and 2, where the filled circles denote the values resulting from the four-parameter fit while the unfilled circles denote the values resulting from the five-parameter fit. For comparison, we have included empirical curves which indicate the expected behavior of the parameters involved. The curve in Fig. 1 is based on the measurements of Abraham

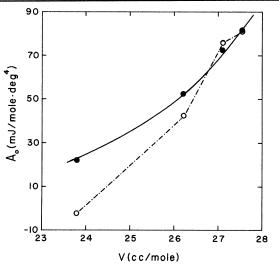


FIG. 1. Parameter A_0 of the phonon spectrum as a function of the molar volume V: closed circles, four-parameter fit; open circles, five-parameter fit. Solid curve, based on the work of Abraham $et\ al$., Ref. 9.

et al.⁹ on c(P), the velocity of first sound in liquid He II under pressure, and $\rho(P)$, the density of the liquid; the ones in Fig. 2 are based on the equations, given by Donnelly, ¹⁰ for the density dependence of the parameters p_0 , μ , and Δ of the roton spectrum, viz.,

$$p_0(\rho)/\hbar = 3.64 \rho^{1/3} \text{ Å}^{-1},$$
 (8)

$$\mu(\rho)/m = 0.32 - 1.103\rho,$$
 (9)

$$\Delta(\rho)/k_{\rm B} = (16.99 - 57.31\rho)^{\circ} K,$$
 (10)

where ρ is the density in grams per cubic centimeter. In each case, the values resulting from the four-parameter fit show a significantly better agreement with the empirical curves than the ones resulting from the five-parameter fit. At the same time, the improvement in the "goodness of fit," arising from the introduction of a nonzero A_1 , is only marginal—in fact, too marginal to justify the sacrifice one would have to make in regard to the standard parameters A_0 , R, and Δ .

The corresponding values of α_1 and α_2 , which follow from A_1 and A_2 with the help of (4), are shown in Table I. We note that while the α_2 resulting from the four-parameter fit has a reasonable dependence on density, the α_1 and α_2 resulting from the five-parameter fit behave rather abnormally. Not only that, the values of α_1 and α_2 , for ρ_0 itself, differ markedly from the values expected on the basis of the Molinari-Regge form of the spectrum! In view of these results, it seems that the quadratic term in the phonon spec-

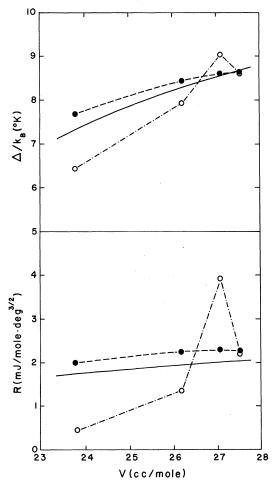


FIG. 2. Parameters R and Δ/k_B of the roton spectrum as a function of the molar volume V: closed circles, four-parameter fit; open circles, five-parameter fit. Solid curves, based on the work of Donnelly, Ref. 10.

trum of superfluid helium is probably not present.

Before we conclude it appears worthwhile to make the following comments.

Although the values of the various parameters,

with $\alpha_1 = 0$, are comparatively reasonable, deviations from expected values are not insignificant. This was also noted by Phillips, Waterfield, and Hoffer, especially in relation to the parameter A_0 . We suspect that these deviations are, to some extent, due to approximations inherent in the formulas (3)–(7), viz., the ones arising from the fact that the evaluation of the phonon and roton contributions to C_v is done over ranges of momenta that overlap. This approximation may introduce significant errors in the analysis¹³ and may lead to somewhat inaccurate values of the parameters involved. We propose to investigate this question in detail and hope that a more careful evaluation of the quantities $C_{\gamma, ph}$ and $C_{\gamma, rot}$ will improve the final results.

Secondly, it is possible that the temperature of the liquid plays a role. The experiments of Woods and Cowley, on which the Molinari-Regge analysis was based, and of Anderson and Sabisky were carried out above 1°K. On the other hand, the experiments of Phillips, Waterfield, and Hoffer, on which our analysis is based, and of Roach et al. were done at considerably lower temperatures. It is conceivable that the coefficients of the excitation spectrum, especially α_1 , are strongly temperature dependent, so that the two sets of investigations may turn out to be complementary, rather than contradictory. There are hints of such a dependence in the literature 10,14; however, no one has carried out the Molinari-Regge type of analysis for temperatures below 1 °K.

Finally, we recall that several authors have commented on the density dependence of the coefficient γ in Eq. (1), 2,15,16 especially on the passage of γ from negative to positive values as density increases. The general impression left behind is that there is no theoretical evidence for this behavior of γ . In this context, we wish to point out that if one takes the Brueckner-Sawada

TABLE I. Coefficients α_1 and α_2 of the phonon spectrum.

V (cm 3 /mole)	Five-parameter fit, $\alpha_1 \neq 0$		Four-parameter fit, $\alpha_i \equiv 0$
	α ₁ (Å)	α_2 (Å ²)	$egin{array}{c} lpha_1 \equiv 0 \ lpha_2 \equiv \gamma \hbar^2 \ (\mathring{\mathrm{A}}^2) \end{array}$
27.58	0,01 (0.27) a	$-0.47 (-0.71)^a$	-0.45
27.11	-0.21	0.17	-0.42
26.23	1.09	-7.64	-0.05
23.79	8.63	-267	2.16

^aValues expected on the basis of the Molinari-Regge form of the spectrum.

spectrum for an imperfect Bose gas, namely12

$$\epsilon(p) = \left[\left(\frac{p^2}{2m} \right)^2 + \frac{1}{2} \Lambda \left(\frac{\hbar}{ma} \right)^2 p^2 \frac{\sin(pa/\hbar)}{pa/\hbar} \right]^{1/2}, \quad (11)$$

where Λ is a monotonically increasing function of the density of the system and is determined by the implicit relationship

$$\frac{4\pi^2\rho a^3}{m} = \Lambda \int_0^\infty \frac{x \sin^2 x}{x^3 + \Lambda \sin x} dx, \tag{12}$$

and expresses it in the form (1), one obtains

$$c = (\frac{1}{2}\Lambda)^{1/2}\hbar/ma$$
, $\gamma = \frac{1}{12}(a/\hbar)^2(1-3/\Lambda)$. (13)

At low densities, where $\Lambda \simeq 8\pi \rho a^3/m \ll 1$, one obtains the Bogoliubov expressions¹⁷

$$c \simeq (4\pi \rho a \hbar^2/m^3)^{1/2}, \quad \gamma \simeq -m/32\pi \rho a \hbar^2.$$
 (14)

Equations (12)-(14) display the same qualitative trend for γ as the empirical values do. Of course, the density at which the zero of γ occurs is much smaller than the one encountered in liquid helium nevertheless, the appearance of a γ which is negative at low densities and becomes positive at higher densities is strongly suggestive of the possibility that an improved theory of elementary excitations in a Bose liquid may explain the observed behavior of γ quantitatively as well.

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220 (1970); see also H. J. Maris, Phys. Rev. Lett. <u>28</u>, 277 (1972).

²N. E. Phillips, C. G. Waterfield, and J. K. Hoffer, Phys. Rev. Lett. 25, 1260 (1970).

³R. B. Hallock, Phys. Rev. A 5, 320 (1972).

⁴E. Feenberg, Phys. Rev. Lett. <u>26</u>, 301 (1971).

⁵A. Molinari and T. Regge, Phys. Rev. Lett. <u>26</u>, 1531 (1971).

⁶A. D. B. Woods and R. A. Cowley, Phys. Rev. Lett. <u>24</u>, 646 (1970), and Can. J. Phys. <u>49</u>, 177 (1971).

C. H. Anderson and E. S. Sabisky, Phys. Rev. Lett. 28, 80 (1972).

⁸P. R. Roach, B. M. Abraham, J. B. Ketterson, and M. Kuchnir, Phys. Rev. Lett. <u>29</u>, 32 (1972).

⁹B. M. Abraham, Y. Eckstein, J. B. Ketterson, M. Kuchnir, and P. R. Roach, Phys. Rev. A <u>1</u>, 250 (1970).

¹⁰R. J. Donnelly, Phys. Lett. 39A, 221 (1972).

¹¹L. D. Landau and E. M. Lifshitz, Statistical Physics (Pergamon, Oxford, England, 1958).

¹²R. K. Pathria, "Statistical Mechanics" (Pergamon, Oxford, England, to be published).

¹³A. D. Singh, Can. J. Phys. <u>46</u>, 2607 (1968).

¹⁴K. Yamada and K. Ishikawa, Progr. Theor. Phys. <u>47</u>, 1455 (1972).

15J. Jackle, Phys. Rev. Lett. 27, 654 (1971).

¹⁶P. R. Roach, J. B. Ketterson, and M. Kuchnir, Phys. Rev. A 5, 2205 (1972).

¹⁷The thermodynamics of an imperfect Bose gas, with a negative γ , has been studied in detail by R. K. Pathria and A. D. Singh, Proc. Nat. Inst. Sci. India, Part A 26, 520 (1960). Equation (40a) of this reference is identical with Eq. (3) of the present paper, with the Bogoliubov parameters $\alpha_1 \equiv 0$, $\alpha_2 \equiv -m/32\pi pa$, and $c = (4\pi pa\hbar^2/m^3)^{1/2}$. See also T. D. Lee and C. N. Yang, Phys. Rev. 112, 1419 (1958).

Phase Diagram of a Charged Bose Gas

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The phase diagram of a charged Bose gas is drawn. We discuss the domains of existence of the solid, fluid, and superfluid phases. It is predicted that superdense helium can be superfluid at densities higher than $10^6~\rm g/cm^3$.

The purpose of this note is to draw the phase diagram of a system of charged bosons of mass m and charge e; embedded in a uniform background of opposite charge which ensures overall electrical neutrality. This model might be of astrophysical interest. In a white dwarf or in the outer layers of a neutron star, the atoms are pressure ionized, and the electrons form a very inert² uniform Fermi sea; if the nuclei are bosons, as it is the case for instance for helium,

we do have a charged Bose gas in a uniform background. The density-temperature diagram that we obtain is drawn in Fig. 1, and will now be explained.

When classical statistical mechanics applies, there are two possible phases, fluid and solid. A simple dimensional argument³ says that the transition between these phases occurs when the thermal energy kT is some well-defined fraction Γ^{-1} of a characteristic Coulomb energy e^2/\overline{r} (\overline{r}

¹H. J. Maris and W. E. Massey, Phys. Rev. Lett. 25,