Phonon Dispersion and Contribution to the Heat Capacity in Liquid He\textsuperscript{4}

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The temperature dependent shift of the phonon excitation spectrum in liquid He\textsuperscript{4}, due to the phonon interactions in quantum hydrodynamics, is calculated over a wide range of momentum. It is found that in a limit of the small momentum the result is in agreement with previous papers, and in another limit of the large one the shift decreases logarithmically with increasing momentum and turns into negative value beyond a critical momentum.

Further a correction term to the specific heat, which is brought by this temperature dependent shift of the spectrum and gives a deviation from the $T^3$-law, is evaluated from the lowest order thermodynamical potential. The predicted contribution is proportional to $T^7$ and results in negligible magnitude at low-temperature and pressure compared with the term $\gamma T^3$ (see Eq. (1.2)), which is dominant in the analysis of the experiments for the heat capacity by Phillips et al. As a result, the positive dispersion ($\gamma<0$) at zero temperature has not been altered by the evaluation obtained here.

§ 1. Introduction

The recent report by Phillips, Waterfield and Hoffer\textsuperscript{1}) supports the positive dispersion in phonon spectrum in He II from the measurements of temperature and pressure dependence in the constant volume heat capacity $C_v$. Supposing the non-interacting phonon gas with the assumed dispersion relation,

$$\varepsilon_p = cp\left(1 - \gamma p^3 - \delta p^4\right),$$

the low temperature expression for $C_v$ is given by\textsuperscript{8})

$$C_v = A \left[ T^3 + \frac{25}{7} \gamma \left(\frac{2\pi K}{c}\right)^3 T^5 + 7 (4r^2 + \delta) \left(\frac{2\pi K}{c}\right)^4 T^7 \right],$$

where $A = \pi (2\pi K)^4/15 h^3 c^3$. On the basis of (1.2) they analyzed the experimental data, which exhibits small deviation from $T^3$-law, and showed that the value of $\gamma$ for the best fit, retaining the first two terms, is $-4.1 \times 10^7$ in cgs units at the saturation vapor pressure, nearly zero at certain intermediate one and finally changes to the positive value at higher pressure. This negative $\gamma$ at low pressure may serve to resolve\textsuperscript{3}) the discrepancies between theory and experiment\textsuperscript{5}) for the propagation of sound in collisionless regions. It might be also useful for elucidation\textsuperscript{6}) of the new behaviors in the first sound attenuation under pressure, recently

\textsuperscript{8}) The third term in Eq. (2) in Ref. 1 includes a trivial error in the numerical factor.
K. Yamada and K. Ishikawa reported by Roach, Ketterson and Kuchnir.\textsuperscript{5)}

The consistency between the thermodynamical quantities, like heat capacity, calculated with use of the excitation spectrum at finite temperature observed by neutron diffraction, and their experimental data was shown by Bendt, Cowan and Yarnell.\textsuperscript{6)} Their procedure is based on the quasi-particle picture for phonons at finite temperature. The purpose of the present paper is to examine an effect on $C_v$ due to the shift of phonon spectrum at non-zero temperature from that at $T=0$, given in (1·1).

It has been extensively studied both experimentally and theoretically that expression (1·1) is subject to small but observable change at $T\approx 0$ by the phonon-phonon interactions. The shift of sound velocity in the ultrasound wave is obtained, in the collisionless region, from the real part of self-energy of low momentum phonon to the lowest order as to the phonon interactions in the quantum hydrodynamics.\textsuperscript{7)} To evaluate the correction to $C_v$ mentioned above at low temperature, we need only the lowest order contribution of thermodynamical potential $\Omega$, due to the phonon interactions. We find that $\Omega$ can be represented as a summation over momentum of the self-energy. Then it is required to obtain the lowest order self-energy valid over the entire range of momentum, differently from the case for the ultrasound.

In § 2 we calculate the self-energy, taking account of all contributions in the lowest order as to three and four phonon interactions in quantum hydrodynamics of Landau. We find there that in the low momentum limit the sound velocity shift agrees with the results by Carroll\textsuperscript{8)} and Andreev and Khalatnikov.\textsuperscript{9)} We also give the dispersion coefficient $\gamma(T)$ and compare with $\gamma$ at $T=0$. In the higher momentum range, $p\gtrsim KT/c$, it is found that the velocity shift decreases logarithmically with increasing $p$ and turns into the negative value beyond a critical $p$.

In § 3 the thermodynamical potential is expressed in terms of this self-energy and the correction to $C_v$ is calculated. We see that this correction is proportional to $T^3$ and is negative at low temperature, which has the same order of absolute magnitude as the $\gamma^3$-term but much less than $\gamma$-term in (1·2). Consequently the conclusion obtained by Phillips et al.\textsuperscript{10)} remains valid at lower and higher pressures, but at certain intermediate one, in which $\gamma$ becomes nearly zero as is seen from the analysis of heat capacity experiments, the $T^3$-term calculated here may play a role in the deviation from the $T^4$-law. The last section is devoted to some discussions for the value of $\gamma$.

§ 2. Self-energy at finite temperature

The relevant Hamiltonian of phonon interactions, in the second quantized form of hydrodynamics,\textsuperscript{10)} is

$$H_s=\sum V_s(p,p,p) \{ c_{p}^{\dagger}c_{p'}^{\dagger}c_{p''}\delta_{p_1+p_2-p_3} + c_{p'}^{\dagger}c_{p'}^{\dagger}c_{p''}\delta_{p_1+p_2-p_3} \} + \text{h.c.}, \quad (2·1)$$
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\[ H_4 = \sum V_4(p_1, p_2, p_3) \left( c_{p_1}^+ + c_{-p_1} \right) \left( c_{p_2}^+ + c_{-p_2} \right) \left( c_{p_3}^+ + c_{-p_3} \right) \delta_{p_1+p_2+p_3-p_4}, \]

(2.2)

where

\[ V_4(p_1, p_2, p_3) = \left( \frac{c_{p_1} p_2 p_3}{32 \rho V} \right)^{1/3} \left( 2u - 1 + \frac{p_1 \cdot p_2}{p_1 p_2} + \frac{p_2 \cdot p_3}{p_2 p_3} + \frac{p_3 \cdot p_1}{p_3 p_1} \right), \]

\[ V_4(p_1, p_2, p_3, p_4) = \frac{(u-1)^2 + \omega}{48 \rho V} \left( p_1 p_2 p_3 p_4 \right)^{1/3} \]

with two phenomenological parameters

\[ u = \frac{\rho}{c} \frac{\partial c}{\partial \rho}, \quad \omega = \frac{\rho}{c} \frac{\partial^2 c}{\partial \rho^2}. \]

Now the self-energy of second order in $H_4$ is given by\(^{11,12}\)

\[ \Sigma_4(p, \varepsilon_p) = 2 \sum_{p'} \left[ 2V_4^{\frac{3}{2}}(p, p', p+p') \frac{n_{p'-p}}{\varepsilon_{p'} + \varepsilon_{p'} - \varepsilon_{p} + \varepsilon_{p'}} + V_4^{\frac{3}{2}}(p, p', p-p') \right. \]

\[ \times \left. \frac{n_{p'-p} + n_{p'-p} + 1}{\varepsilon_{p' - \varepsilon_{p'}} + \varepsilon_{p} - \varepsilon_{p'}} - V_4^{\frac{3}{2}}(p, p', -p-p') \right], \]

(2.3)

where $n_p = [\exp(\varepsilon_p/KT) - 1]^{-1}$. The self-energy of the lowest order in $H_4$ is

\[ \Sigma_4(p, \varepsilon_p) = 24 \sum_{p'} V_4(p, -p, p', -p')(n_{p'} + \frac{1}{2}). \]

(2.4)

For later convenience we write down the sum of (2.3) and (2.4) as follows:

\[ \Sigma(p) = \Sigma^{(0)}(p) + \Sigma^{(T)}(p), \]

(2.5)

where we used the notation $\Sigma(p)$ in place of $\Sigma(p, \varepsilon_p)$ since $\Sigma(p, \varepsilon_p)$ is a function of only $|p| = \rho$ as will be seen later. $\Sigma^{(0)}(p)$, independent of temperature, denotes the zero point energy, which are associated with the factors 1 in (2.3) and 1/2 in (2.4). The second term, depending on temperature through $n_p$, can be rewritten as

\[ \Sigma^{(T)}(p) = \sum_{p'} f(p, p') n_{p'}, \]

(2.6)

with

\[ f(p, p') = 4 \left[ \frac{V_4^{\frac{3}{2}}(p, p', p+p')}{\varepsilon_{p'} + \varepsilon_{p'} - \varepsilon_{p} + \varepsilon_{p'}} - \frac{V_4^{\frac{3}{2}}(p, p', p-p')}{\varepsilon_{p} - \varepsilon_{p'} + \varepsilon_{p'} - \varepsilon_{p} + \varepsilon_{p'}} + \frac{V_4^{\frac{3}{2}}(p, p', p-p')}{\varepsilon_{p} - \varepsilon_{p'} + \varepsilon_{p'} - \varepsilon_{p} + \varepsilon_{p'}} \right] + 24V_4(p, -p, p', -p'). \]

(2.7)

Eckstein and Varga\(^{13}\) showed that $\Sigma^{(0)}(\rho)$ coming from (2.3) leads to a positive $\gamma$ as well as a velocity shift at $T=0$. The values obtained, however, depend on the upper cut off momentum. So the perturbational treatment of these temperature-independent terms is not completely reliable and this difficulty re-
presents a weak point of the approaches basing on the quantum hydrodynamics. Then in what follows we ignore the contribution from $\Sigma^{(p)}(\rho)$ and confine ourselves to that from $\Sigma^{(T)}(\rho)$, and we assume to regard $c$ and $\gamma$ at $T=0$ as parameters to be determined experimentally, as Phillips et al. did.

In the same way as in the earlier work, we use the linear spectrum, $cp$ for $\varepsilon_p$, apart from using (1.1) $(\delta=0)$ to avoid the divergence at small angles in denominators in (2.3). Summation over $p'$ can be carried out by an integral in the spherical coordinates. Performing the integration over the angular variables, we get

$$\Sigma^{(T)}(\rho) = ST^4(\alpha(T)p + \beta(p,T)), \quad (2.8)$$

where

$$S = \pi^2 \frac{(u+1)^2}{30} \left(\frac{K}{c}\right)^4, \quad (2.9)$$

$$\alpha(T) = \ln \left( \frac{2}{3\gamma} \left(\frac{c}{KT}\right)^2 \right) - \frac{3}{2} + \frac{3\omega-4}{6(u+1)^2}, \quad (2.10)$$

$$\beta(p,T) = \frac{15}{2\pi^2} \frac{KT}{c} \int_0^\infty dx x^2 \left( x - \frac{cp}{KT} \right)^3 \ln \left| x + \frac{cp}{KT} \right| \left| x - \frac{cp}{KT} \right|.$$  \hspace{1cm} (2.11)

(2.8) is an odd function of $p$.

In the small $p$ limit, (2.8) gives

$$\Sigma^{(T)}(\rho) \approx ST^4 \left[ \ln \frac{2}{3\gamma} \left(\frac{c}{KT}\right)^2 - 4.8852 + \frac{3\omega-4}{6(u+1)^2} \right] \rho - \frac{\zeta(2)}{6\pi^2} \frac{(u+1)^2}{\rho h^3} \left(\frac{K}{c}\right)^3 T^2 \rho^2, \quad (2.12)$$

where the numerical value $-4.8852$ results from $-37/6 + 2\gamma_c (-1.1544) + 2(90/\pi^4) \times \sum_{n=1}^{\infty} (\ln n/n^4) \approx -0.1271$ ($\gamma_c$: Euler constant) and $\zeta(2) = 1.645$ is the zeta function. The first term in (2.12) is in complete agreement with the recent result by Andreev and Khalatnikov, and also with that by Carroll if we neglect the contribution from $\Sigma_0$, that is, if we subtract $((u-1)^2 + \omega)/2(u+1)^2$ from the square bracket in (2.12). This agreement with the latter would be natural, because his treatment is essentially the same as ours although he uses the density and velocity fields in place of the annihilation and creation operators used here. The agreement with the former, however, might be surprising at first sight, because it has not yet been necessarily clear how can be derived the phenomenological two-fluid kinetic theory from the Hamiltonian (2.1) and (2.2). However we can show that the two-fluid kinetic theory in the collisionless region is consistent with the dynamical behavior of the system with (2.1) and (2.2) in an
appropriate approximation, although we here do not go into the details.\(^\text{*})\)

The first term in (2·12) differs in the numerical value from the earlier results,\(^8,13,14\) as was pointed out in Refs. 9 and 5), where the explicit estimates of it have not been given. We here give numerically \(\Delta c/c = \Sigma^{(9)}(p)/c\rho\), using the parameters\(^9,13,15\)

\[ u = 2.84, \quad \omega = 8.26, \quad \rho = 0.145, \]

\(c = 238\ \text{m/sec}\) and

\(|\gamma| = 4.1 \times 10^{27} \text{ cgs units}, \quad (2·13)\)

and show it in Fig. 1, which was represented by Abraham et al.\(^5\) The agreement with the experiments is certainly improved, although the predicted values in (2·12) are still slightly large and also there is no frequency dependence which is seen in the experiments.

The second term in (2·12) gives the temperature dependent and positive \(\gamma(T)\):

\[ \gamma(T) = \frac{1.645}{6\pi^2} \left(1 + \frac{1}{\rho}\right)^2 (KT)^2 \quad (2·14) \]

\(< 0.358 \times 10^{27} T^{-1} \text{ cgs units}.\)

The value at \(T = 1^\circ\)K is less by one order of magnitude than \(\gamma = -4.1 \times 10^{27} \text{ cgs units}\) estimated from the heat capacity experiments, and is the same order as \(\gamma = (0 \pm 2) \times 10^{26} \text{ cgs units}\) estimated from the neutron scattering (see § 4).

In the limit of large \(p\), (2·8) results in

\[ \Sigma^{(7)}(p) = ST^4 \left[ \left( \ln \frac{2}{3\gamma p^2} - \frac{5}{2} + \frac{3\omega - 4}{6(u + 1)^2} \right) p - \frac{40\pi^5}{63p} \left( \frac{KT}{c} \right)^4 \right] \quad (2·15) \]

We find in this case that the first term includes the logarithmically decreasing term and turns into negative value when \(\gamma p^2 \geq 6.92 \times 10^{-2}\) or \(p \geq 3.94 \times 10^7 \text{ cm}^{-1}.\)**

\(^*\) In the low momentum region, the second and third terms in \(\Sigma^4((2·3))\) cancel out exactly the factor \((u - 1)^2\) in \(\Sigma^4\) (see (2·4) and the expression of \(V^2\)).

\(^{**}\) See Note added in proof.
where (2·13) was used. To see more clearly this situation we carry out the numerical integration of (2·11) and show in Fig. 2 the variation of $\Sigma^{(\tau)}(p)/c_p$, the velocity shift obtained from (2·8). Although the validity for applying the quantum hydrodynamical approaches up to the large momentum range like this case should be critically considered, the behavior mentioned above, if it would be, might be checked by the inelastic scattering of neutrons. Cowley and Woods [16] observed the temperature variations in $T=1.0$ K of excitation spectrum, but it is not so clear whether the predicted behavior is realized or not, although it appears that we could see qualitatively this behavior, for instance, in the range $0.2 \sim 0.6 \AA^{-1}$ of Fig. 20 in Ref. 16).

§ 3. Corrections to $C_\tau$

To estimate the contribution of phonon interactions to $C_\tau$ we calculate thermodynamical potential $\mathcal{Q}$. This can be carried out in a similar way to that in the anharmonic crystal, which was done by Cowley. [10] By rearranging $\mathcal{Q}$ calculated in the lowest order in $H_3$ and $H_4$, it can be expressed by means of the self-energy obtained in § 2:

$$\mathcal{Q} - \mathcal{Q}_0 = \frac{1}{2} \sum_p \Sigma(p) (n_p + \frac{1}{2}) + E_0,$$

where $\mathcal{Q}_0$ denotes one for the harmonic parts of phonon and $E_0$ is some constant.

Using (2·5), (2·6), the symmetric property $f(p, p') = f(p', p)$, which is easily seen from (2·7), and a relation $\sum_p f(p, p') = \Sigma^{(0)}(p)$, we obtain

$$\frac{d}{dT} \left\{ \frac{1}{2} \sum_p \Sigma(p) \left(n_p + \frac{1}{2}\right) \right\} = \sum_p \Sigma(p) \frac{dn_p}{dT}.$$

Fig. 2. Variation of $\Sigma^{(\tau)}(p)/c_p$ versus momentum. The limiting values in $p \rightarrow 0$ are plotted on the ordinate. Scale in the ordinate represents in $10^{-3}$ units for $T=2.0, 1.5, 1.0, 0.5$ K and in $10^{-4}$ units for $T=0.3, 0.2$ K.
Then the internal energy $U = \mathcal{Q} - T(\partial \mathcal{Q} / \partial T)$ results in

$$U - E_0 = \sum_p \left( \varepsilon_p + \frac{1}{2} \Sigma(p) \right) \left( n_p + \frac{1}{2} \right) - T \sum_p \Sigma(p) \frac{dn_p}{dT}. \tag{3·3}$$

Observing the relation $-Tdn_p/dT = \varepsilon_p dN_p/d\varepsilon_p$, (3·3) may be considered as the form retained to the lowest order of $\Sigma(p)$ in an expression

$$\sum_p (\varepsilon_p + \frac{1}{2} \Sigma(p)) (\bar{n}_p + \frac{1}{2}), \tag{3·4}$$

where $\bar{n}_p = n(\varepsilon_p + \Sigma(p))$. (3·4) would be the expected form in the quasi-particle picture for the weakly interacting phonons.

With use of (3·2) heat capacity $C_v = \partial U / \partial T$ is given by

$$C_v = \sum_p (\varepsilon_p + \Sigma(p)) \frac{dn_p}{dT} + \frac{d}{dT} \sum_p \varepsilon_p \Sigma(p) \frac{dn_p}{d\varepsilon_p},$$

where $C_v = \sum_p \varepsilon_p dN_p/dT$ leads to (1·2) and

$$\Delta C_v = \left( -\frac{1}{T} + \frac{d}{dT} \right) \sum_p \varepsilon_p \Sigma(p) \frac{dn_p}{d\varepsilon_p}, \tag{3·5}$$

where we used again the apparent relation used in the above. For the same reason mentioned in §2, we here again ignore the term with $\Sigma^{(0)}(p)$ in (3·5).\(^*\)

As can be easily seen by substituting $\Sigma^{(T)}$, (2·12) expanded in powers of $p$, into (3·5), the contributions from each term with the different powers of $p$ have all the same power of $T$, $T^T$. Therefore to evaluate (3·5) we need to use for $\Sigma^{(T)}(p)$ the original expression (2·8) valid for arbitrary magnitude of momentum.

We finally obtain

$$\Delta C_v = A \frac{S}{c} T^T \left[ -7 \alpha(T) + \frac{15}{4} + 29, 20 \right], \tag{3·6}$$

where $A$, $S$ and $\alpha(T)$ are given in (1·2), (2·9) and (2·10), respectively. (3·6), proportional to $T^T$, is the same order of magnitude as $\gamma^2$-term in (1·2) but negative sign at low temperature. These two terms, however, is negligible at $0.5^\circ K > T$ comparing with $\gamma$-term in (1·2) because of the difference in the temperature dependence. Consequently the results by Phillips et al.,\(^3\) analyzed with use of the first two terms in (1·2), are not subject to any essential modification at low and high pressure, but when $\gamma$ goes to nearly zero at certain intermediate pressure (about 5 atm), (3·6) may contribute to the small deviation from $T^\gamma$-law in $C_v$.

\(^*\) The contribution to $C_v$ from $\Sigma^{(0)}(p)$ gives a term proportional to $T^\gamma$. This is considered to be included into the main term in (1·2), in which the front factor is regarded as a parameter.
§ 4. Discussion

The temperature dependent shift of the phonon excitation spectrum was examined over the wide momentum range. However we should note that these results depend crucially on the dispersion of the spectrum. This holds also for the sound attenuations and other relaxation processes. As seen in the last section, the result $\gamma = -4.1 \times 10^{37}$ cgs unit, obtained from the experiments of the heat capacity, was retained without alterations.

On the other hand, Woods and Cowley showed that $\gamma$ was nearly zero, $(0 \pm 2) \times 10^{36}$ cgs units from the neutron experiments. This result, as was shown by Pines and Woo, was consistent with the liquid structure factor $S(k)$ measured in X-ray, together with the theoretical calculation of the deviation from Feynman formula by Lai, Sim and Woo. Although there is at present the apparent difference for $\gamma$'s value, it seems that zero or negative $\gamma$ is more favorable than the positive one accepted before (e.g., $\gamma \approx 3 \times 10^{37}$ cgs units). If it is the case, we have the finite lifetime even at zero temperature:

$$\frac{\hbar}{\tau_p} = \frac{(\mu + 1)^3}{240\pi} \rho \hbar^6,$$

(4·1)

which is derived from twice the imaginary part of the second term in (2·3).*

This is negligibly small for the wave length in the ultrasound, but amounts to the measurable order in the range of the inelastic scattering of neutrons: e.g., while $\hbar/\tau_p = 3.5 \times 10^{-5}$K for $\rho/\hbar = 0.2 \AA^{-1}$, $\hbar/\tau_p = 0.85$°K for $0.6 \AA^{-1}$. However as can be seen in Fig. (17) in Ref. 16) we cannot observe the energy width with a tendency increasing intensively by the factor $\rho^3$, but the width at 1.1°K is equal for the range $0.2 \sim 0.6 \AA^{-1}$. Then it is required to examine the validity for the theoretical expression of the lifetime, at the same time further experimental investigation is hoped.

Finally our discussion and many other investigations have been based on the assumption that the phonon spectrum $\varepsilon_p$ can be expressed in an odd power series in the wave vector $p$. This assumption, however, does not necessarily have the rigorous theoretical base. Recently Feenberg pointed out that, under conditions of low density and weakly interacting Bose gas, if the interatomic potential falls off asymptotically as $r^{-6}$, then $\varepsilon_p$ contains both odd and even powers of $p$. Another recent paper by Gould and Wong, who developed the microscopic analysis of a weakly interacting Bose gas to the higher orders, showed that $\varepsilon_p$ is non-analytic function of $p$ like $\varepsilon_p = \rho (c_0 + c_1 p^3 + c_2 p^4 \ln(1/p))$ and the low temperature expression for $C_v$ is $T^3 [C_0 + C_1 T^2 + C_2 T^4 \ln(1/T)]$. Although these two theories were

* The lifetime proportional to $p^0$ has been given by M. Nelkin, Phys. Rev. 127 (1962), 979. The difference of the factor between (4·1) and one by Nelkin comes from the differences in the coupling of the phonon interactions. The microscopic theories also give the lifetime of this kind (see the references cited in the Nelkin's paper).
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developed only for the weakly interacting Bose gas and hence were not directly applicable to the real He⁴, they might be suggestive in the investigations of phonon spectrum and specific heat.

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References

4) Y. Hiki, private communication.

Note Added in Proof:

After this paper was prepared, we found that the value of w, 8.26 in (2·13) should be changed to 0.194 [B. M. Abraham et al., Phys. Rev. A2 (1970), 550]. This modification reduces about 7 per cent at T=0.3°K the calculated value in Fig. 1. Two relations following (2·15) should read (μ/2π) ≥ 5.26×10⁻² or ρ ≥ 3.40×10⁶ cm⁻¹ and curves represented in Fig. 2 are modified slightly.