

Phonon-Like Excitations in Liquid Helium

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An attempt is made to formulate a theory of phonon-like excitations in liquid helium by generalizing the theory of phonons in quantum crystals to quantum disordered systems. By using experimental data for pair correlation functions, dispersion curves of longitudinal phonon modes are calculated, with a result for their over-all behavior in fairly good agreement with phonon-roton dispersion curves obtained by neutron scattering measurements. It is shown that the temperature-dependence of the calculated roton-minimum is in good agreement with experiment for liquid helium I, but not for liquid helium II. A brief discussion is given on the nature of rotons in liquid helium. It is suggested that, within certain restrictions, the roton spectrum in liquid helium is likely to be a general feature of the dispersion of elementary excitations in liquids, classical or quantum.

§ 1. Introduction

Since the pioneering work of Landau,¹⁾ many attempts have been made to study the physical properties of elementary excitations in liquid helium.²⁾ On the other hand, several works, both experimental and theoretical, have implied that the over-all behavior of the dispersion of elementary excitations in classical simple liquids bears some resemblance to that of phonon-roton excitations in liquid helium.^{3)~6)} It has also been shown that high-frequency collective modes in classical liquids are rather similar to phonons in amorphous or polycrystalline solids.^{6)~9)} These results suggest that the dispersion of elementary excitations in non-crystalline solids and liquids, classical or quantum, is likely to have several things in common and may evoke a renewed interest in the nature of rotons in liquid helium. There is, however, a good reason to believe that such an analogy between classical and quantum disordered systems, as it stands, may only be meaningful for liquid helium I. Recently a soft-mode behavior of rotons in liquid helium II has been noted.^{10),11)}

The principal purpose of this paper is to develop a theory of phonon-like excitations in liquid helium by generalizing the theory of phonons in quantum crystals.^{12),13)} In a previous paper,⁵⁾ to be referred to as I, the present authors have studied the general properties of elementary excitations in non-crystalline solids and liquids by employing a phonons-in-amorphous-solids approach. Particular attention was paid there to the interrelationship between the roton-like spectra

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and the local ordering in disordered systems. This paper is intended to be as compact as possible because a generalization of the theory developed in I to the present case, which may be called a phonons-in-amorphous-quantum-solids approach, is quite straightforward. Attention is called to the fact that a phonon dispersion curve in liquid helium obtainable by such a method is in fairly good agreement with phonon-roton excitations observed by neutron scattering measurements.¹⁴⁾

§ 2. Preliminary discussions

As a preliminary for later discussions, we first study phonons in a classical non-crystalline solid composed of atoms of a single species within the framework of the harmonic approximation. According to the results obtained in a previous paper^{5),9)} the dynamical matrix \mathcal{D} giving the phonon eigenfrequencies $\omega \equiv \omega(k)$ as a function of wave vector k is given by^{*}

$$\mathcal{D} \equiv \mathcal{D}(k) = (\rho/M) \int dR g_{,}^*(R) \nabla \nabla v(R) [1 - \exp(ik \cdot R)]. \quad (1)$$

Here M and ρ are the atomic mass and the number density of the atoms, respectively, $v(R)$ is the "bare potential" describing the interaction of a pair of atoms and $g_{,}^*(R) \equiv g^*(R, \omega)$ is an "effective pair correlation function" in which the effect of higher order correlations other than pairs for the distribution of the atoms are exactly taken into account.⁹⁾ In this paper we neglect the difference between $g_{,}^*$ and $g_{,}$, a "bare pair correlation function" for the sake of simplicity. Physically this is mainly to neglect the damping of phonon modes arising from the structural disorder. In the previous paper,⁹⁾ Eq. (1) was applied to an analytically tractable one-dimensional model disordered system. The principal result obtained by such a calculation is that a partial disorder characterizing a non-crystalline solid gives rise to a frequency gap Δ_L in the phonon dispersion curve at the value of k corresponding approximately to the first peak in the structure factor and that the quantity Δ_L decreases with increasing local order and eventually vanishes in the limit of complete ordering corresponding to a crystal lattice. It has been shown⁹⁾ that if we make an attempt at identifying Δ_L with the roton minimum Δ in liquid helium, the above result is in qualitative agreement with the pressure and the temperature dependence of Δ observed experimentally.^{9),14)}

§ 3. Generalization to a non-crystalline quantum solid

We next generalize Eq. (1) to a non-crystalline quantum solid, although the present authors are not aware of the existence of such a disordered system. As in the case of phonons in quantum crystals,^{15),16)} we do this by modifying the bare pair potential v in Eq. (1) by an effective potential v^* in which the effect of

^{*}) Equation (1) is obtained by combining Eqs. (3.11) and (2.3) in Ref. 5) with each other. The quantity $\nabla \nabla v(R)$ here corresponds to the force constant K in Eq. (2.3).

the anharmonicity of vibrations and the short-range correlations arising from the large zero-point atomic motion and the strong repulsive part of the bare potential, respectively, are taken into account. Here we follow the method suggested by Nosanow and co-workers^{15)~17)} to approximate v^* by^{*)}

$$v^*(R) = (A/2\pi)^{1/2} (1/R) \int_0^\infty dR' R' W(R') \{ \exp[-(A/2)(R' - R)^2] - \exp[-(A/2)(R' + R)^2] \}, \quad (2)$$

where

$$W(R) = f(R)^2 [v(R) - (\hbar^2/2M) \nabla^2 \ln f(R)] \quad (3)$$

in which

$$f(R) = \exp[-K\{(\sigma/R)^{12} - (\sigma/R)^6\}] = \exp[-(K/4\epsilon)v(R)] \quad (4)$$

is the Jastrow factor. In the above equations A and K are adjustable parameters and σ and ϵ are constants.

Before calculating Eq. (1) numerically, along the line described above we remark that various formulas giving the eigenfrequencies of short-wavelength collective modes in simple liquids have forms similar to Eq. (1)^{18), 19)} and that the results of numerical calculations yield dispersion curves which bear some resemblance to phonon-roton-like excitations as obtained in our previous paper for phonons in one-dimensional non-crystalline solid.⁵⁾ The underlying fact in such a physical situation would be that for short-wavelength modes we could expect the liquid to show a solid-like behavior.²⁰⁾ In this meaning Eq. (1), when applied to classical liquids, may be called a phonons-in-amorphous-solids approach.

§ 4. Application to liquid helium and numerical calculations

In this paper we have made an attempt at applying Eq.(1) with v replaced by v^* to liquid He⁴, confining ourselves to those eigenvalues of $\mathcal{D}(k)$ which correspond to longitudinal phonons. This may be called a *phonons-in-amorphous-quantum-solids approach* to liquid helium. The numerical calculation has been done using the values of the pair correlation function g , obtained by Henshaw²¹⁾ and by Gordon et al.²²⁾ at $T=1.06^\circ\text{K}$, 2.40°K and 4.20°K . Following Nosanow and his co-workers,^{15)~17)} we put $\sigma=2.556\text{\AA}$ and $K=0.178$. The numerical value of A in Eq. (2) has been determined to fit roughly the velocity of sound waves v_s . It is taken to be 1.30\AA^{-2} , 1.20\AA^{-2} and 0.90\AA^{-2} for $T=1.06^\circ\text{K}$, 2.40°K and 4.20°K , respectively (the values of v , thus obtained are 252 m sec^{-1} , 226 m sec^{-1}

^{*)} Equation (2) is readily obtained from Eqs. (18) and (19) in Ref. (17). We do not intend here to reiterate the cluster variation method, upon which Eq. (2) is based, employed in the theory of phonons in quantum crystals. For a detailed discussion on this, see, for example, Refs. (15) and (16) and also (13).

and 171 m sec^{-1} , respectively). These numerical values are smaller than those used in the case of solid He⁴. This choice of the parameters K and A seems to be reasonable in the sense that the anharmonicity of atomic vibrations in liquid phase is generally larger than in solid phase (the quantity A is a measure of the inverse of mean square displacement of atoms), while the difference of a quantity characterizing hard cores in solids and liquids seems to be negligible. The result of numerical calculations is shown in Fig. 1, together with the experimental result of Henshaw and Wood at 1.12°K .¹⁴⁾ It is seen that the agreement of the over-all behavior of dispersion curves between the theory and the experiment¹⁴⁾ is fairly good. The numerical values of the roton minimum energy Δ as a function of temperature are also plotted in Fig. 2. It is seen that the quantity Δ decreases linearly with temperature and that it well agrees with experimental result¹⁴⁾ above the superfluid transition temperature. It is, however, to be noted that its temperature dependence below the λ -point T_λ is very different from that obtained here. This is probably due to new mechanisms or interactions arising from the onset of superfluidity which have not been taken into account in our treatment. There is a good reason to believe that our method of phonons-in-amorphous-quantum-solids approach cannot be used, as it stands, for liquid helium II.

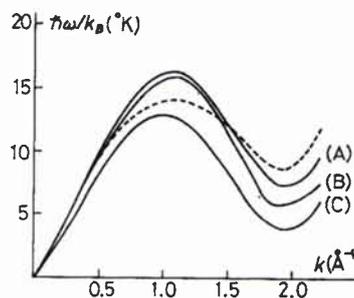


Fig. 1. Dispersion curve of elementary excitations in liquid helium. The dashed curve shows the experimental result of Henshaw and Woods (Ref. 14) at $T=1.12^\circ\text{K}$ obtained by neutron scattering measurements. Curves (A), (B) and (C) denote calculated values of phonon eigenfrequencies using the values of g_s obtained by Henshaw (Ref. 21) at $T=1.06^\circ\text{K}$ (curve (A)) and those by using the values of g_s obtained by Gordon et al. (Ref. 22) at $T=2.40^\circ\text{K}$ (curve (B)) and $T=4.20^\circ\text{K}$ (curve (C)).

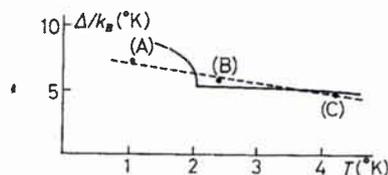


Fig. 2. Temperature dependence of the roton minimum. The continuous line denotes the experimental result (Ref. 14). The solid circles (A), (B) and (C) show the calculated values of the roton minimum obtained from the curves (A), (B) and (C) in Fig. 1, respectively.

§ 5. Discussion

Aside from their over-all behavior, the main difference of the result obtained here from that of Feynman expressed by the formula^{23),*)}

*) The quantity $S(k)$ is the structure factor of liquid helium.

$$\epsilon(k) \equiv \hbar\omega(k) = \hbar^2 k^2 / 2MS(k) \quad (5)$$

or that of the more elaborate work of Feynman and Cohen³⁴⁾ is that the former, as we can show from Eq. (1), gives the second maximum in ω -versus- k curve, while the latter yield a monotone increasing function for $k > k_0$. Here k_0 is the value of k at which the roton minimum takes place. The experimental result of Henshaw and Woods,¹⁴⁾ although indefinite, seems to suggest the existence of such a second maximum. This difference in the behavior of dispersion curves are illustrated in Fig. 3. It is worthy of note in this connection that the second maximum in the dispersion of elementary excitations has been observed for classical liquids.^{3), 4), 8), 9), 18), 19), 20)}

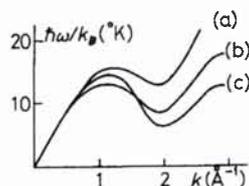


Fig. 3. Dispersion curves of elementary excitations obtained by Feynman and Cohen (curve (a), at $T=0^\circ\text{K}$), by Henshaw and Woods (curve (b), at $T=1.12^\circ\text{K}$) and in the present paper (curve (c), at $T=1.06^\circ\text{K}$).

In view of these facts, it appears that the dispersion of elementary excitations in liquid helium looks something like that in classical liquids. This point has been discussed previously by Singwi, Sköld and Tosi,⁹⁾ by Murase⁴⁾ and by the present authors.⁵⁾ The claim made by Singwi, Sköld and Tosi was that the plateau beyond the roton minimum might be a general feature of liquids. In this paper it is suggested that the behavior of excitations in the roton region itself does reflect some general features of liquids. To study this point, in particular, the nature of rotons, a systematic investigation of the temperature and the pressure dependence of dispersion curves of elementary excitations in classical as well as in quantum liquids is required for a wide range of momentum transfer utilizing neutron scattering measurement. Very recently, such an experimental study has been made by Dietrich et al. for liquid helium.³⁶⁾ Theoretical studies of this point have also been made by Schneider and Enz¹⁰⁾ and by Ruvalds.¹¹⁾ These workers remarked

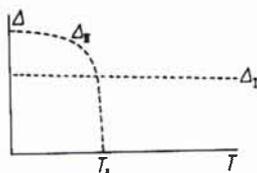


Fig. 4. A tentative illustration of the temperature dependence of the roton minimum Δ as being composed of two branches Δ_I and Δ_{II} .

soft-mode behaviors of rotons in liquid helium II with respect to their dependence on external pressure and temperature. Qualitatively the experimental result for the temperature dependence of the roton minimum $\Delta \equiv \Delta(T)$ as shown in Fig. 2¹⁴⁾ might be understood by noting that the discontinuity near $T = T_\lambda$ is the result of the joining of two distinct branches of the excitation spectra of the system (see Fig. 4). Then the result obtained here (see Fig. 2) and that of Ruvalds may correspond to $\Delta_1(T)$ and $\Delta_{II}(T)$, respectively. He has shown that the behavior of the roton minimum $\Delta_{II}(T)$, particularly its temperature dependence, can be accounted for by taking into account roton-roton interactions giving the renormalization of the roton energy $\Delta_{II}(T) = \Delta_0 + \Sigma(T)$. Here Δ_0 is the value of $\Delta_{II}(T)$ without roton-roton interactions. For example, neutron data at $T = 1.1^\circ\text{K}$ gives $\Delta_0 = 8.65^\circ\text{K}$. The quantity $\Sigma(T)$ is the roton self-energy. The result obtained by him for $\Sigma(T)$ can be expressed in the form of self-consistent equations:

$$\Sigma(T) = 2gN_{\text{R}}(T) \quad (6)$$

with

$$N_{\text{R}}(T) = [2p_0^3(\mu k_{\text{B}}T)^{3/2}/(2\pi)^{3/2}\hbar^3] \exp[-\{\Delta_0 + \Sigma(T)\}/k_{\text{B}}T]. \quad (7)$$

Here g is the roton coupling parameter, $N_{\text{R}}(T)$ denotes the number of rotons, k_{B} is the Boltzmann constant and μ is the effective mass of rotons when expressed in the form of the Landau formula:¹⁾

$$\epsilon \equiv \epsilon(p) = \Delta + [(p - p_0)^2/2\mu]. \quad (p = \hbar k, \quad p_0 = \hbar k_0) \quad (8)$$

Based upon these formula, he was also successful in explaining the temperature dependence of the superfluid density. By such a calculation, $\Delta_{II}(T)$ tends to zero as T approaches T_λ from below, thus giving a soft-mode behavior. As shown in Fig. 4, the underlying fact here would be that the nature of rotons below and above T_λ must be quite different.

It is not yet clear what is the interrelationship between $\Delta_{II}(T)$ obtained by Ruvalds and $\Delta_1(T)$ obtained here. In view of the results obtained in this paper and also of the temperature dependence of the width as well as the energy of the roton minimum,¹⁴⁾ it is suggested as an alternative conjecture that a roton-minimum curve is likely to be composed of a single branch (rather than two branches as shown in Fig. 4), undergoing an abrupt change in the vicinity of $T = T_\lambda$. In this meaning it appears that rotons below and above T_λ , although their behaviors are qualitatively different, are likely to be of similar nature, somewhat reflecting a general feature of liquids. To confirm whether this is true or not, a further examination and a generalization of the method developed herein taking into account the effect of Bose-Einstein condensation are probably required.

§ 6. Concluding remarks

The main results obtained in this paper can be summarized as follows: (a) Equation (1) with $v(R)$ replaced by the effective potential $v^*(R)$ (cf. Eq. (2))

gives dispersion curves of phonon-like excitations in liquid helium with their over-all behavior below and above the transition temperature T_λ in fairly good agreement with experiment. (b) Upon closer examination it is shown that the temperature-dependence of the roton-minimum obtained here is in good agreement with experiment for liquid helium I, but not for liquid helium II. (c) It appears that the important properties of the roton minimum for $T < T_\lambda$ are determined by quantum effects other than those taken into account in this paper. (d) In this sense quantum effects to be taken into account in quantum liquids seem to be different from those in quantum solids. (e) The present theory is, therefore, not applicable, as it stands, to liquid helium II.

References

- 1) L. Landau, J. Phys. USSR 5 (1941), 71; 11 (1947), 91.
- 2) For a general survey of the problem of liquid and solid helium see, for example, J. Wilks, *Properties of Liquid and Solid Helium* (Oxford University Press, 1967); W. K. Keller, *Helium-3 and Helium-4* (Plenum Press, 1966), and also references cited therein.
- 3) K. S. Singwi, K. Sköld and M. P. Tosi, Phys. Rev. Letters 21 (1968), 881 and references cited therein.
- 4) C. Murase, J. Phys. Soc. Japan 29 (1970), 549.
- 5) S. Takeno and M. Goda, Prog. Theor. Phys. 47 (1972), 790 and references cited therein.
- 6) Y. Obata, JAERI (Japan Atomic Energy Research Institute) 1157; *Conference on Inelastic Neutron Scattering in Solids and Liquids* (Tokai, Japan, 1967), p. 114.
- 7) K. E. Larsson, *Proceedings of a Symposium on Neutron Inelastic Scattering* (Copenhagen, 1968), Vol. 1, p. 397.
- 8) J. Hubbard and J. L. Beeby, J. Phys. C2 (1969), 556.
- 9) S. Takeno and M. Goda, Prog. Theor. Phys. 45 (1971), 331.
- 10) T. Schneider and C. P. Enz, Phys. Rev. Letters 27 (1971), 1186.
- 11) J. Ruvalds, Phys. Rev. Letters 27 (1971), 1769.
- 12) For a review, see N. R. Werthamer, Am. J. Phys. 37 (1969), 763.
- 13) See also, R. A. Guyer, *Solid State Physics*, edited by F. Seitz, D. Turnbull and H. Ehrenreich (Academic Press New York and London, 1969), p. 413.
- 14) D. G. Henshaw and A. D. B. Woods, Phys. Rev. 121 (1961), 1266.
- 15) L. H. Nosanow, Phys. Rev. 146 (1966), 120.
- 16) J. H. Hetherington, W. J. Mullin and L. H. Nosanow, Phys. Rev. 154 (1967), 175.
- 17) F. W. de Wette, L. H. Nosanow and N. R. Werthamer, Phys. Rev. 162 (1967), 824.
- 18) See, for example, J. Chihara, Prog. Theor. Phys. 41 (1969), 285 and also references cited therein.
- 19) See, for example, K. S. Singwi, K. Sköld and M. P. Tosi, Phys. Rev. A1 (1970), 454 and also references cited therein.
- 20) J. Frenkel, *Kinetic Theory of Liquids* (Oxford University Press, London, 1946).
- 21) D. G. Henshaw, Phys. Rev. 119 (1960), 14.
- 22) W. L. Gordon, C. H. Shaw and J. G. Daunt, J. Phys. Chem. Solids 5 (1958), 119.
- 23) R. P. Feynman, Phys. Rev. 94 (1954), 262.
- 24) R. P. Feynman and M. Cohen, Phys. Rev. 102 (1956), 1189.
- 25) K. Sköld and K. E. Larsson, Phys. Rev. 161 (1967), 102.
- 26) O. W. Dietrich, E. H. Graf, C. H. Huang and L. Passell, Phys. Rev. A5 (1972), 1377.