THEORY OF LIQUID HELIUM

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by

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Abstract

A quasi-thermodynamic theory of liquid helium is developed based on four very
general postulates. The results are in excellent agreement with experiment, in
particular with the measurements of the temperature waves (second sound), the
existence of which was predicted by the theory. The Bose-Einstein liquid intro-
duced earlier is a possible molecular model for the theory. Landau's hydrodynamic
theory has certain features which are in conflict with the present results and also
are contradicted by experiment. A modified hydrodynamic theory, however, might
provide a satisfactory molecular model. A decision between the two molecular models
requires further experimental and theoretical study.
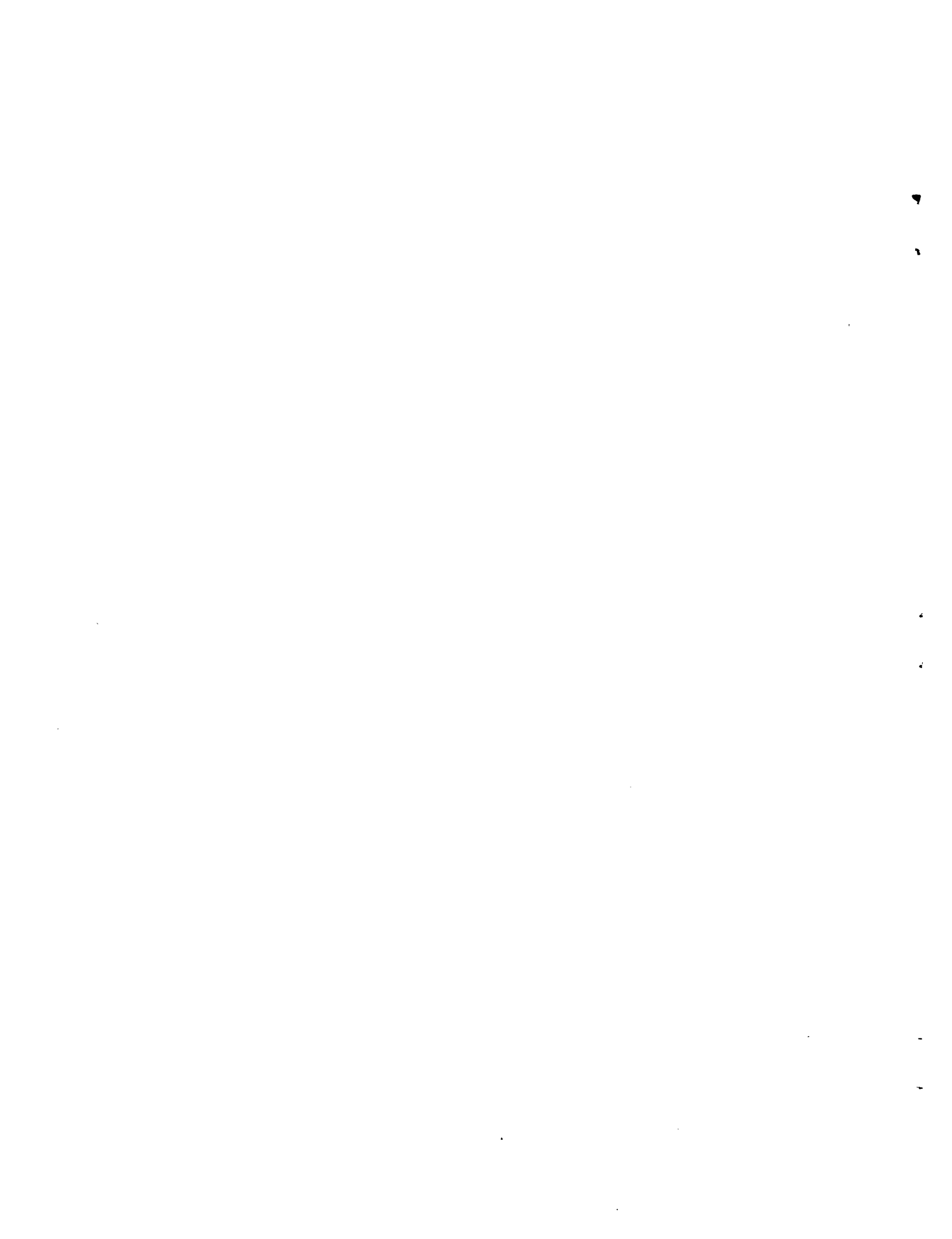
THEORY OF LIQUID HELIUM

1. Introduction

Several years ago F. London[1] gave some evidence to support the idea that the peculiar phase transition of liquid helium at $2.19°K$ ("λ-point") might be regarded as due to the condensation mechanism characteristic of the ideal Bose-Einstein gas, distorted, of course, by the presence of molecular forces which cannot be ignored in the case of a liquid. He observed that the various earlier attempts to explain the λ-transition by the familiar mechanisms of phase transitions of the second kind (order-disorder transition in ordinary space) are incompatible with the weak Van der Waals forces of helium and with the requirements of quantum mechanics. In this situation the example of an order in momentum space, as presented by the Bose-Einstein condensation, seemed to offer a very welcome and suggestive new possibility. Moreover, the Bose-Einstein condensation demonstrated a very peculiar feature. It led to a kind of phase equilibrium of two phases interpenetrating in ordinary space but separated in momentum space. There were good reasons to suspect that the "condensed" fraction of the atoms was the seat of a superfluid mass transfer.

Actually the author was able to show[2] that the kinetic effects to be expected in an ideal Bose-Einstein gas revealed a close similarity to the well-known effects in helium II. In spite of this fact, the theory did not seem very convincing since it was difficult to understand how the properties of a liquid could be interpreted even qualitatively in terms of a gas theory. The rigorous treatment of this problem would require the solution of the quantum mechanical many-body problem. The mathematical difficulties involved have proved so far to be so tremendous, that up to the present it has been impossible to substantiate these ideas by the development of a rigorous molecular theory.

In view of this situation, the author has tried to avoid these difficulties by developing a somewhat more macroscopic theory of the various thermo-hydrodynamic effects in helium II.[3,4] (See also H. London[4a]. For an account of these experiments we refer to Keesom's monograph on helium[5]). The main point of the theory was the definition of the "Bose-Einstein liquid", uniting some properties of a liquid and some of a Bose-Einstein gas in a self-consistent scheme. Whereas the discussion of such a "hybrid" system could not be carried out with quantum mechanical methods, it proved to be easily manageable from the much more schematic quasi-thermodynamic point of view. In particular the notion of "condensation in momentum space" was extended to the Bose-Einstein liquid. Hence, below the condensation temperature, this system was supposed to have a heterogeneous character implying the existence of two velocity fields. The existence of two fields rather than one resulted in an "internal convection" carrying energy and entropy, but not associated with any net transfer of matter. Hence the macroscopic hydrodynamics of helium II proved to be of greater

complexity than that valid for other substances and allowed the correlation of phenomena which appeared paradoxical from the point of view of ordinary hydrodynamics. The theory led also to various predictions which have been subsequently verified by experiment. The most important of these was the conclusion that inhomogeneities of the temperature would propagate according to a _wave equation_ rather than the usual _parabolic equation_ of heat conduction. The velocity of propagation of these "_temperature waves_" was computed as a function of the temperature. It was also indicated that an experimental verification could be obtained by exciting the temperature waves through periodical heating of helium II. A short report of these results appeared in 1938 in the form of preliminary notes[3], but the detailed account[4] (quoted henceforth as l.c.) was, because of wartime conditions, not generally available until recently.

Landau - apparently unaware of these results - criticised the Bose-Einstein theory on the basis of the earlier note[2] and advanced instead a theory in which he maintained the assumption of the heterogeneous nature of helium II, but tried to justify it from a different molecular point of view based on quantum hydrodynamics[6]. As soon as the possibility of the two velocity fields was granted, the development of the macroscopic hydrodynamic equations had to proceed on the same lines as those based on the Bose-Einstein theory. In particular Landau concluded that there should be two different modes of sound propagation in helium II, with different velocities for the "first" and "second" sound. Landau did not discuss the physical meaning of this second sound wave. According to the recent account of Peshkov[7], it was only after an attempt to excite the second sound by mechanical means had failed, that the question was reconsidered by Lifshitz[8], who concluded that the excitation of second sound could best be obtained by periodical heating of the liquid. Thus the second sound proved to be identical to the temperature waves predicted earlier on the basis of the Bose-Einstein theory. The existence of these temperature waves has been experimentally demonstrated by Peshkov[7] who measured also their velocity of propagation as a function of temperature. These results have been recently confirmed by an interesting indirect method suggested by Onsager[9]. The comparison of the experimental and theoretical results will be found in Section 4. It may be noted that the theoretical curves do not contain any adjustable constants, but are evaluated with the empirical values of the entropy measured previously by Kapitza[10].

The aim of the present paper is to clarify the relation between the theories and experiment. It seemed advantageous to proceed as much along phenomenological lines as possible. The theory of the Bose-Einstein liquid was already more phenomenological than the original Bose-Einstein gas theory. The present paper is a further step in this direction. This procedure — although rather abstract — seems necessary in view of the existence of Landau's theory. It will help to avoid the pitfall of justifying any special molecular model by experimental results, which in reality can be also understood under broader assumptions.

There are good reasons to expect that a quasi-thermodynamic method should be adequate to handle the problem of helium II. The peculiar properties of helium II are

usually described by the statement that this substance is superfluid and superheat-conducting. Actually it is important to realize that the behavior of helium II cannot be characterized by assuming extremely small or large values of kinetic coefficients such as viscosity and heat conductivity. It is rather that the usual differential equations governing capillary flow and the transfer of heat have to be replaced by equations adapted to the particular mechanisms effective in helium II. This change in differential equation rather than the assumption of an extreme value of the conductivity is characteristic also of superconductivity, as is apparent from the well-known theory of F. and H. London. While kinetic coefficients depend very sensitively on the nature of the molecular forces, this is usually not the case for the differential equations themselves.

Our procedure will then be to advance certain general assumptions regarding the energy spectrum of liquid helium, expressing the fact that this system unites in a peculiar way some characteristics of a liquid and of a gas (Section 2). The lowest state of the system has liquid characteristics. The excited states can be classified into compressional modes which are also liquid-like (Debye phonons) and shear modes corresponding to the rearrangements at constant volume of the molecules, which are supposed to have a gas-like character. The technique of drawing conclusions from these assumptions will be essentially thermodynamical (Sections 3 and 4). Such an extension of classical thermodynamics seems to be the adequate method of dealing with second-order transitions. We hope to come back to this question in a later publication. A discussion from the molecular point of view is of course of the greatest interest and will be outlined in Section 5 as far as it seems possible at present. Landau's theory will be discussed in Section 6.

2. The Fundamental Assumptions

Let us consider a macroscopic system of helium atoms enclosed in a box of volume V. The characteristic values of the Schroedinger equation of the system, in particular the lowest energy E_0, are determined by the properties of the helium atoms and in addition depend on the volume V as a parameter. Two assumptions will now be made regarding the properties of this lowest state of the system. After a short discussion of the meaning of these assumptions two others will follow regarding the excited states.

(i) The function $E_0(V)$ has a minimum for some value $V = V_0$ of the volume:

$$\left(\frac{dE_0}{dV}\right)_{V = V_0} = 0 \qquad \left(\frac{d^2 E_0}{dV^2}\right)_{V = V_0} > 0 \qquad (1)$$

(ii) The state $E_0(V)$ is characterized by a vanishing microscopic rigidity.

Assumption (i) expresses the fact that at a temperature of absolute zero and with a

vanishing external pressure $P = \dfrac{-dE_o}{dV} = 0$, the system is in a condensed and not in a gaseous state. Assumption (ii) specifies further that this state is liquid rather than solid.

The postulate that a system should have a vanishing coefficient of rigidity at _absolute zero_ is very far reaching. Indeed, according to classical statistics one would expect every system which is in thermodynamic equilibrium at absolute zero, to be in a crystalline state. Hence it is reasonable to define a system satisfying postulates (i) and (ii) as a _quantum liquid_. Among liquids in the conventional sense of the word only helium fulfills this condition. There is also the possibility of considering heavy atomic nuclei as quantum liquids.

At first it might seem that the discussion of the implications of (ii) will be hampered by the difficulties inherent in the theoretical understanding of the liquid state. Actually the reverse is true, since the properties of helium (we think here chiefly of helium I) are in some respects simpler than those of ordinary, or classical liquids. The comparison of ordinary liquids with helium I can throw new light on some elusive features of the theory of the former.

The terminology of _microscopic rigidity_ as a counterpart to _macroscopic_ rigidity has apparently been first introduced by Brillouin[11], although essentially the same ideas form the basis of the theories of liquids by Frenkel, Andrade, and Eyring, (cf. particularly the recent detailed account of Frenkel[12]).

If a solid is deformed (shearing strain), it reacts with a proportional shearing stress, the proportionality constant being the shear modulus, or macroscopic rigidity. It may be said also that the free energy of the solid is a function of the shearing strain. This is, of course, not so in case of a liquid. In contrast to the free energy, the potential energy of configuration depends on the strain also in a liquid; in other words, in the course of a shearing deformation the molecules have to surmount barriers of potential energy, but these barriers constantly collapse under the impact of thermal agitation. The observable consequences of this situation are the so-called visco-elastic effects. Suppose the average life time of a potential barrier is τ . If a shearing stress is applied at a high frequency $\omega \gg \tau^{-1}$, the system will react as a solid and propagate transverse elastic waves. For liquids of low viscosity the frequency required for such a behavior shifts to the very high values of the thermal vibrations (hypersonic in contrast to ultrasonic waves). The specific heats of monatomic liquids near the freezing point show good evidence for the presence of transverse hypersonic waves[13].

At low frequencies $\omega \ll \tau^{-1}$ for which the rate of change of the shearing strain will be proportional to the stress, the microscopic rigidity gives rise to viscosity. This _liquid type_ viscosity should be distinguished from the _gas type_ or _transport_ viscosity well known from the kinetic theory of gases. In order to avoid

confusion we will call them _dynamic_ and _kinetic viscosity_, respectively. The two viscosities give rise to the same type of macroscopic effects, but they can be clearly identified from their temperature dependence.

For the kinetic viscosity one has

$$\eta_{kin} \sim \rho \bar{c} \ell \tag{2}$$

where ρ is the density, $\bar{c}$ the mean thermal velocity, and ℓ the mean free path; η_{kin} increases slowly with temperature.

For the dynamic viscosity one has a strongly negative temperature coefficient, since

$$\eta_{dyn} \sim e^{A/kT} \tag{3}$$

where A is an activation energy clearly indicating the presence of potential barriers. The situation is illustrated in Figure 1 where the dynamic viscosity of liquid hydrogen is compared with the kinetic viscosity of hydrogen gas. The case of hydrogen is typical of any classical liquid.

The most spectacular effect of the _microscopic_ rigidity is _freezing_. As the thermal agitation decreases with temperature, the potential barriers organize into a crystalline pattern and give rise to the _macroscopic_ rigidity of the solid.

The important point for the present purpose is that the three phenomena discussed above (propagation of transverse hypersonic waves, dynamic viscosity, and freezing) are all due to the same factor: the microscopic rigidity. As helium does not solidify at vanishing pressure, one must conclude that the microscopic rigidity vanishes, as it was postulated in (ii). According to the above discussion this implies also the vanishing of the dynamic viscosity and the absence of transverse hypersonic waves. The first conclusion finds a striking verification in the measurement of viscosity (Figure 2 to be compared with Figure 1). Attention is drawn here to the viscosity of helium gas and liquid helium I, since in the case of helium II additional complications arise, which will be discussed in the next section. The kinetic character of the viscosity of helium is obvious. It should be pointed out that in our terminology it is helium, not just helium II, that is a quantum liquid. This terminology based on (i) and (ii) is well justified, e. g., on the basis of the viscosity measurements. None the less it is at variance with the rather generally adopted view that helium I is "classical" or a "normal" liquid. Actually the spectacular properties of helium II have overshadowed the fact that helium I is distinctly different from classical liquids.

A qualitative discussion of postulate (ii) from the point of view of quantum mechanics will be found in Section 5. At present we turn to the discussion of the excited states of the system.

The excited states of a continuum or quasi-continuum can be classified into compressional and shear modes of motion. The density fluctuations connected with the

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first type can be analyzed into longitudinal waves. The infinite spectrum of longitudinal modes of the continuum has to be "cut" in the usual fashion to obtain the correct number of degrees of freedom. The elastic quanta (briefly, phonons) obtained by quantization of these waves are the "elementary excitations" of the compressional modes of motion.
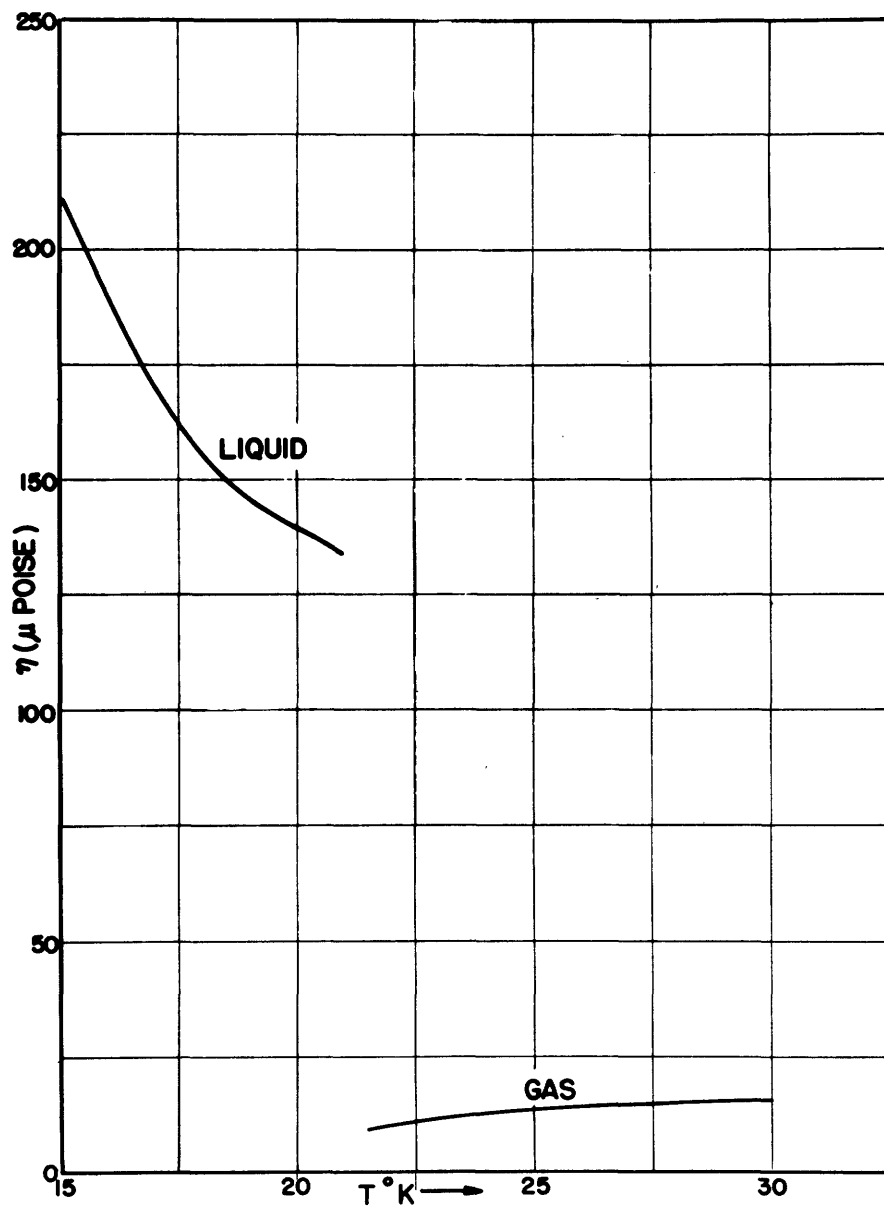

Figure 1. Viscosity of liquid and gaseous hydrogen, from
W. H. Keesom, G. E. MacWood, Physica 5, 745 (1938).

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Regarding the shear modes postulate (ii) leads only to the negative state-
ment that they cannot have the character of waves. This gap will be filled by the
next assumption:

(iii) <u>The shear modes of motion of liquid helium have a gas-
eous character. The "elementary excitations" corre-
spond to translations of atoms (or groups of atoms) with
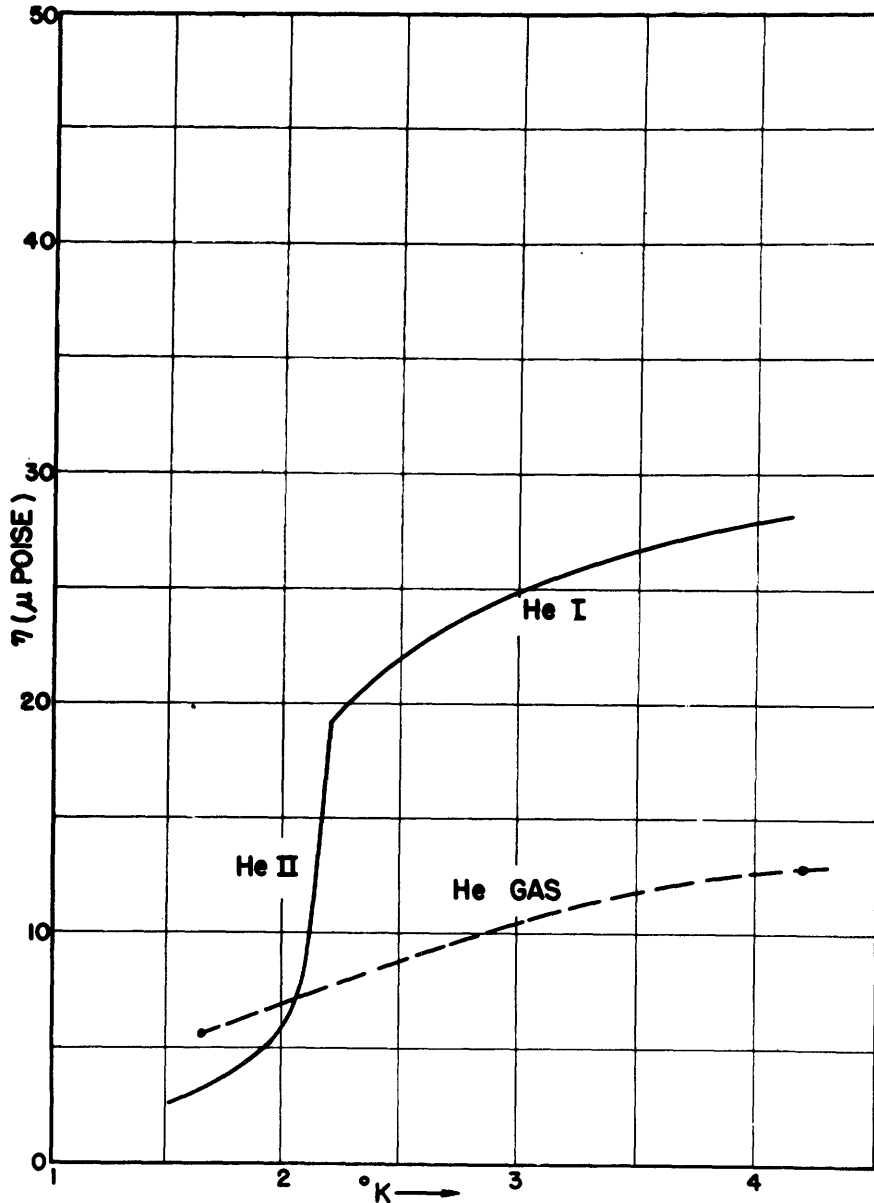definite values of mass, momentum, and energy.</u>

Figure 2. Viscosity of liquid and gaseous helium, from pp. 106
and 267-8 of Reference 5.

In accordance with the program formulated in the Introduction the postulate
(iii) is kept in very general terms, since the macroscopic equations to be derived in
the next two sections do not depend on the molecular interpretation of the "gas" in-
troduced in (iii). This molecular interpretation is of course important for the in-
tuitive understanding of the meaning of the theory, and will be discussed in Sections
5 and 6. At present only a few results of that discussion should be anticipated.

The first molecular model compatible with (iii) is the Bose-Einstein liquid.
In this case the "elementary excitations" correspond to helium atoms in translational
Bloch-type states[1,4]. Between these states there is a strong correlation, inasmuch
as they occur at constant density (shear modes of motion). This model has been con-
sidered in great detail in l.c.

The second model will be referred to as the vortex model. It was obtained
through modification of Landau's hydrodynamic theory which in its original form is
contradicted by experiment. In this model the elementary excitations of (iii) are
groups of atoms possessing in addition to their translational momentum also an internal
angular momentum. Although the theory of these "vortex molecules" has not been worked
out so far, progress in this direction is conceivable. Actually the two models are very
closely related to each other. In the Bose-Einstein case the "molecules" of the gas of
postulate (iii) are monatomic, while in the vortex model they are polyatomic. There are
many hydrodynamic phenomena (such as those considered in this paper) for which the dif-
ference between a polyatomic and a monatomic gas is irrelevant.

The most important property of the Bose-Einstein liquid is its "condensation
in momentum space"[1,4]. The essential features of this condensation phenomenon will now
be postulated for our more general case.

While the elementary excitations introduced in (iii) have the main attributes
of molecules in the kinetic theory of gases (constant mass, momentum, and energy), this
is true only if a definite quantum state is considered. At absolute zero no "molecules"
are present, but can be created by means of thermal excitation. (While at absolute
zero, there is no translational motion, the atoms still possess a zero point kinetic
energy, cf. Section 5.) In every quantum state one can distinguish the total mass of
the system. Upon averaging over a canonical ensemble, the same kind of subdivision is
obtained as a function of temperature. Hence the density of the system will be sub-
divided into two parts.

$$\rho = \rho_n + \rho_s \qquad (4)$$

where ρ_n is the density connected with the "molecules" of the gas and ρ_s refers to the
"background" in which the molecules are moving. The subscripts refer to "normal"
and "superfluid", a terminology which will be explained below. The ratio ρ_n/ρ is a
very important property of the system. The last assumption of the theory will be con-
cerned with it:

(iv). <u>There exists a temperature T_0 such that</u>

$$\frac{\rho_n}{\rho} \left\{ \begin{array}{lll} = 0 & \text{for} \quad T = 0 & \text{He II} \\ < 1 & 0 < T < T_0 & \text{He II} \\ = 1 & T \geq T_0 & \text{He I} \end{array} \right.$$

with ρ_n/ρ monotonically increasing between $T = 0$ and T_0.
T_0 is to be identified with the λ-point of helium.*

Let us consider now a volume element in helium II which is small compared to macroscopic dimensions, but big enough to contain not only a large number of helium atoms, but also a large number of "molecules". Averaging over the translational motion of the "molecules", one obtains a drift velocity $\underset{\sim}{v}_n$ which will not in general coincide with the velocity of the "background" $\underset{\sim}{v}_s$ In contrast to ordinary liquids and also to helium I, one has two densities and correspondingly two velocity fields. The total mass current density is

$$\underset{\sim}{j} = \rho_n \underset{\sim}{v}_n + \rho_s \underset{\sim}{v}_s \tag{5}$$

According to (iii) every "molecule" has a momentum. Averaging over this momentum current one gets, through the procedure well known from the kinetic theory of gases, a stress tensor. The diagonal elements will be equal to a pressure P_n (which is to be distinguished from the liquid pressure P) while the non-diagonal elements represent a viscous momentum flow. This is, of course, of the kinetic type and is uniquely associated with the flow $\underset{\sim}{v}_n$. No kinetic viscosity is associated with the flow of the background $\underset{\sim}{v}_s$. Since the dynamic viscosity was found to be totally absent in helium, none of the mechanisms of viscosity is effective, and this flow may be called <u>superfluid</u>. In particular at absolute zero where $\rho_n = 0$, $\rho = \rho_s$, helium II as a whole is superfluid.

The characters of the two currents will be essentially different; this is reflected most clearly in the boundary conditions for the components of the velocities at a solid wall at rest:

$$v_{n\|} = 0 ,$$

$$v_{n\perp} = 0 , \qquad\qquad\qquad \text{whereas only } v_{s\perp} = 0 . \tag{6}$$

The kinetic viscosity of the gaseous component entails the "normal" boundary condition for the velocity $\underset{\sim}{v}_n$, while no condition is obtained for the tangential component of the superfluid flow. The terminology "normal" and "superfluid" is thus explained.

The superfluid component can "slip" along a solid surface, which is its most essential characteristic, more so than the absence of dissipation. Actually, according

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* Actually ρ_n/ρ will be also a function of the pressure P and this would have to be taken into account in a refinement of the present theory.

to experimental evidence to be discussed below, the superfluid flow may be associated with a dissipation of the turbulent type as soon as a critical velocity is exceeded.

The slip of the superfluid component of helium II is most effectively demonstrated in the well-known creeping of the Rollin film[5]. This is, however, a complex phenomenon for quantitative discussion, corresponding to the fact that the general hydrodynamic equations of helium II are most unwieldy. It is very satisfactory that the form (6) of the boundary conditions is sufficient to obtain conclusions which can be quantitatively checked by experiment. This will be seen in the next section.

Finally we have to consider the specific entropy s of the system. (Specific energy and specific heat follow of course at once from s.) According to our classification of the excited states, s consists of two terms

$$s = s_n + s_{phon} \tag{7}$$

corresponding to the shear and compressional modes of motion. As indicated by the subscript, s_n is associated with the normal component ρ_n of the liquid while the contribution s_{phon} of the Debye phonons is associated with the liquid as a whole. We return to this question in Section 6. It will be seen in the next section that in the temperature range from the λ-point to almost $1^\circ K$, $s_{phon} \ll s_n$. Under this condition, i. e., if s_{phon} can be neglected, it may be said that the superfluid component of the liquid has zero entropy. This result is of great importance for the theory, but its approximate character should be kept in mind.

3. Helium II as a Mixture

The proper understanding of the heterogeneous nature of helium II is made somewhat difficult by the fact that it does not conform to the terminology of the classical phase rule with its clean-cut distinction between "phase" and "independent component". In one connection it is best to consider helium II as a two-phase system and in another rather as a two-component system. Both analogies have to be handled with certain qualifications.

The two-phase point of view has been emphasized by London[1] in connection with the ideal Bose-Einstein gas. Below the condensation temperature this system can be considered as a gas in equilibrium with a condensed phase, the phase separation taking place in momentum space rather than in ordinary space. The saturation gas pressure $P_n = P_n(T)$ is a function of the temperature alone. In the liquid the situation is complicated by the fact that in addition to the gas pressure P_n one has a liquid pressure P. The relation between P and P_n can be better understood in terms of the second analogy: the pressure P_n appears then as an "osmotic pressure" of the normal component dissolved in the superfluid. Because of the existence of semipermeable membranes, P_n is subject to direct observation and this analogy will serve us as a

useful guide throughout this section. Two qualifying remarks have to be observed, however. In the theory of solutions the total amount of each component is constant. In helium II this is true only if adiabatic conditions are maintained. In case of heat transfer there is a transition of one component into the other which has no analogue in the case of ordinary solutions. This requirement of maintaining adiabatic conditions is of importance also from the practical point of view; we will come back to it in connection with the discussion of experiments.

There is a second point in which P_n differs from the conventional osmotic pressure. A gradient of the osmotic pressure in -- say a salt solution -- gives rise to a diffusion (second-order effect). In virtue of postulate (iii), however, a gradient in P_n gives rise to a current, (first-order effect); i. e., in this connection P_n has the character of a gas pressure. This idea will be developed in the next section.

The simplest and most direct experimental method for the verification of the heterogeneous nature of helium II and of the boundary conditions (6) consists in the measurement of the moment of inertia of a rotating vessel containing helium II. As the superfluid component will not take part in the rotation, the apparent moment of inertia of the liquid will be

$$I = I_0 \rho_n / \rho$$

where I_0 is the moment of inertia above the λ-point. This effect was first predicted by Landau[6] and the decrease of the moment of inertia was experimentally demonstrated by Andronikashvilli[14]. This experiment constitutes the most direct, although not the most accurate determination of the important quantity ρ_n / ρ. The experimental results can be approximately represented in the form

$$\rho_n / \rho = (T/T_0)^r \qquad (8)$$

where r is a constant around 6. More precise measurements discussed below give r = 5.5. It should be emphasized that this is a purely empirical relation which cannot be derived within the framework of the quasi-thermodynamic theory.

A related, though somewhat more involved case is the measurement of the viscosity by means of the oscillating disk method. Although the discussion goes somewhat beyond the scope of the quasi-thermodynamic method, it should be given here because of the special interest of the problem.

As in the previous case, the presence of the superfluid component can be ignored. The results can be understood in terms of the elementary formula (2) valid for the kinetic viscosity, provided the density ρ is replaced by ρ_n. In order to obtain the temperature dependence of η , one has to realize that $\rho_n l$ = const. This is equivalent to the statement that the "molecules" of the normal liquid collide only with each other. The justification and meaning of this assumption can be discussed only from the molecular point of view. The question will be taken up again in Section 5. It should be mentioned also that in l.c. the author has used at this point the ad hoc

assumption ρl = const., which is in conflict with the fundamental assumptions of the theory. This was pointed out to the author by Prof. Onsager to whom he is indebted also for the correct interpretation of the experiments. The earlier discussion of the viscosity measurements has been also rightly criticized by Keesom[5]. The present assumption leads to the same temperature dependence of η for both helium 1 and II$_\lambda$ given by $\eta \sim \bar{c} \sim T^{\frac{1}{2}}$. This result seems to conflict with the measurements of Keesom and MacWood[15] reproduced in Figure 2, according to which the temperature dependence of the viscosity of helium II is essentially given by $\eta \sim \rho_n/\rho \sim (T/T_o)^r$. An analysis of the experiment reveals, however, (c.f. formula (1) in Reference 15) that the quantity actually measured is the viscosity multiplied by the density which is in the case of helium II $\eta \rho_n$. Keesom and MacWood have evaluated their measurement under the assumption of having obtained $\eta \rho$. Hence the curve in Figure 2 has to be corrected by multiplication with the factor ρ/ρ_n, which is in agreement with the above result of the theory. Landau's interpretation of the viscosity measurement[16] is in line with the present discussion.

The next special case to be considered is that of a very thin capillary in which the flow velocity of the normal component is negligibly small. Such a capillary acts as a semipermeable membrane, being permeable to the superfluid component alone. Making use of the analogy discussed above, a semipermeable membrane can be used for the measurement of P_n in much the same way as the osmotic cell leads to a qualitative understanding of the well-known fountain phenomenon in helium II. In this experiment an open reservoir is connected to a helium bath by a semipermeable capillary. If the liquid in the reservoir is heated, ρ_n and P_n increase over the corresponding values in the bath and the superfluid component is sucked into the reservoir -- like the solvent into an osmotic cell.

We proceed now to investigate the "osmotic cell" with thermodynamic methods. In order to avoid complications connected with the liquid-vapor interface (evaporation and consequently nonadiabatic transition of the normal component to the superfluid state), an osmotic cell shall be constructed as follows: A small part of a long cylinder filled with helium II is compressed adiabatically by means of a semipermeable piston. This can actually be realized using a porous material, the pores playing the role of the thin capillaries. During the compression the superfluid component flows across the piston, thus keeping the total density ρ constant, while the values of ρ_n, P_n and consequently also of the temperature T are increased over their values in the helium bath.* In order to distinguish an adiabatic compression with a semipermeable piston from an ordinary compression with an impermeable piston, the former process will be called an isopycnic compression (since ρ is kept constant).

The basis for the application of thermodynamics to the isochoric compression is the assumption that the flow across the piston carries along no entropy and is reversible.

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* The small thermal expansion is being neglected throughout this section.

The assumption that the superfluid component has the entropy zero has been first advanced by the author[2], who pointed out also the approximate nature of this assumption (l.c). In fact, as indicated above, the phonon contribution to the entropy s_{phon} is associated with the whole liquid and is carried along in particular also with the superfluid component. Hence the above assumption means that $s_{phon} \ll s_n$, and $s \sim s_n$. The following considerations depend essentially on this assumption, the limits of validity of which will be considered below. It will prove to be correct for not too low temperatures.

The assumption of reversibility of the superfluid flow implies that the flow across the capillary should be slow. This is of course a special case of the general requirement that thermodynamics should be applied only to quasistatic processes. Actually in the present case the situation is more favorable than usual, since experience shows that the dissipation connected with the flow sets in only above a finite critical velocity. We assume that the flow velocities always stay below this limit.

Let us now compress the cell adiabatically by applying the pressure dP on the semipermeable piston. The volume of the cell will decrease until the osmotic pressure difference balances the external pressure $dP_n = dP$. The condition of equilibrium with respect to the superfluid mass transfer is the equality of the chemical potentials:*

$$\mu (T,P) = \mu (T + dt, P + dP) ;$$

expanding the right-hand side and noting that

$$\left(\frac{\partial \mu}{\partial T}\right)_P = - s \simeq - s_n \qquad \left(\frac{\partial \mu}{\partial P}\right)_T = v$$

(v is the specific volume), one has

$$\frac{dP_n}{dT} = \frac{s_n}{v} = s_n \rho \tag{9}$$

This relation was first derived by H. London[4a]. The fact that a temperature difference is associated with an osmotic pressure difference is referred to as the thermomechanical effect.**

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* This follows at once from the principle of virtual work. It is somewhat unusual that the equality of chemical potentials is obtained for states at different temperatures. This difficulty is resolved by comparison with the case of the true osmotic cell. The virtual process in this case is the mass transfer of the solvent which is of course associated with transfer of energy (entropy). Hence the equality of temperatures follows simultaneously with the equality of the chemical potentials. In the present case the superfluid flow carries no entropy or energy in excess over the zero point value, thus the equality of the chemical potential does not imply the equality of temperatures.

** It may be noted that the zero of the entropy is fixed by the assumption that the entropy of the superfluid component vanishes.

It is instructive to compare (9) with the thermodynamic relation

$$\left(\frac{dP}{dT}\right)_V = \frac{\alpha}{\kappa} \tag{10}$$

where α is the coefficient of thermal expansion and κ the isothermal compressibility. The quantity (10) can be observed if the liquid is heated at constant volume. Since α is very small, it is seen that P is not very much dependent on temperature. It is, of course, mainly a function of the density ρ. The opposite is true for P_n. While the derivation of (9) was worded in the terminology of the two-component system, an interpretation from the two-phase point of view is equally possible. Equation (9) is obviously the Clausius-Clapeyron relation expressing the fact that the same condensed (superfluid) phase is in equilibrium with the gas or normal phase both at the values P,T and $P + dP$, $T + dT$ of pressure and temperature.*

Regarding the experimental verification of (9), its limits of validity should be kept in mind: the capillary must be thin enough so as to keep the normal component from transporting entropy and the flow velocity must be below the critical value, and adiabatic conditions should be maintained. Apparently only Kapitza's[10] experiments have been carried out under satisfactory conditions. Kapitza measured all quantities occurring in (9) and found satisfactory agreement within the experimental error. He claims that the best value for the entropy is obtained from the measurement of the left-hand side of Equation (9). These entropy values are in only rough agreement with those obtained by Keesom and collaborators from the measurement of the specific heat[5].

An analytical expression for the entropy will prove very useful. Actually, Kapitza's result can be expressed with good accuracy by the expression (Figure 3).

$$s_n = s_0 \left(T/T_0\right)^r \tag{11}$$

with $s_0 = 0.405$ cal/gm-deg. and $r = 5.5$. Of course it should be kept in mind that (11) has no theoretical foundation and can be considered at best as an approximation.

One more important relation can be derived in connection with the osmotic cell. In the course of the isopycnic compression the normal component and the entropy associated with it are "trapped" in the cell. If the volume of the cell is V, these statements lead to the following relations:

$$d(\rho_n V) = \rho_n dV + V d\rho_n = 0;$$

hence

$$\frac{d\rho_n}{\rho_n} = -\frac{dV}{V}. \tag{12}$$

---

* It is also clear that (9) should contain the entropy difference of the two phases, i.e., s_n rather than s.

Similarly

$$d(s_n \rho V) = \rho(s_n dV + V ds_n) = 0$$

and

$$\frac{ds_n}{s_n} = - \frac{dV}{V} \cdot$$
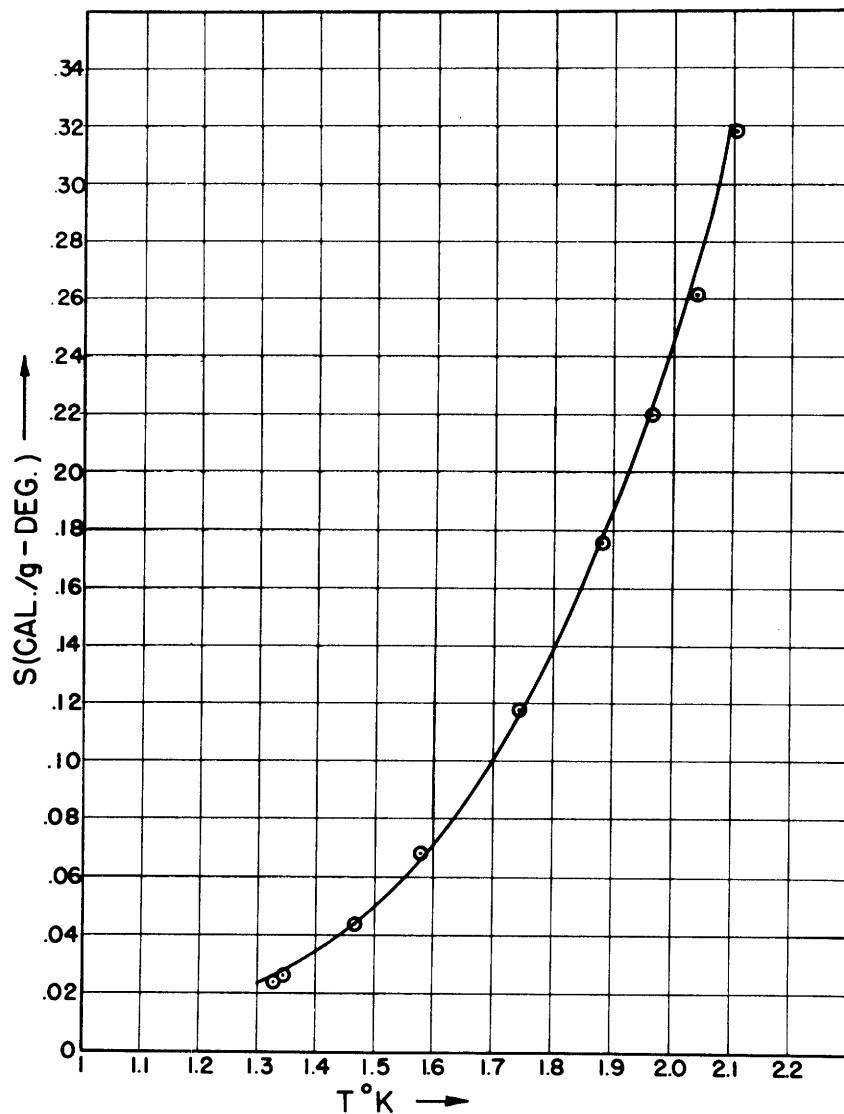
(13)

Figure 3. Approximation of Kapitza's entropy measurements[10] (circles) with formula (11) (curve).

From (12) and (13) one has

$$\frac{d\rho_n}{\rho_n} = \frac{ds_n}{s_n} .$$ (14)

Or integrating,

$$\frac{\rho_n}{\rho} = \frac{s_n}{s_o}$$ (14a)

since at the λ-point $\rho_n(T_o) = \rho$ and $s_n(T_o) = s_o$. This relation has also been obtained by F. London[17]. Hence the empirical formula for the entropy (11) implies

$$\rho_n/\rho = (T/T_o)^r$$ (15)

with

$$r = 5.5 .$$

The result is compatible with the direct determination[14] of this function discussed earlier in this section, but the accuracy of the direct determination is much less than that obtained by using the relations (9) and (12).

We are now in a position to discuss the limits of validity of our assumption $s_{phon} \ll s_n$. The phonon entropy can be tentatively represented by a Debye formula

$$s_{phon} = \frac{4\pi^4 k}{15 \, m} (T/\theta)^3$$ (16)

which is a factor 3 times smaller than the Debye formula for solids, since in the latter case one has two transverse waves for every longitudinal one. Computing θ in the usual way from the sound velocity (240 m/sec), one gets $\theta \approx 18°$ and

$$s_{phon} \approx 2 \times 10^{-3} T^3 \quad cal/gm\text{-}deg.$$ (17)

Picard and Simon[18] have actually found an approximate T^3 law for the specific heat in the temperature range $0.2° - 1.0°$, but their values seem to be about 4 times larger than one would expect according to (17). Their measurements correspond to the Debye temperature $\approx 11°$. Picard and Simon claim that their measurements should be described by the Debye temperature $\theta \approx 15.5°$ which is $(3)^{1/3}$ times too high, since they evaluated their measurements according to the Debye formula valid for solids. Although the Debye theory is not rigorous enough to expect an exact coincidence of the Debye temperatures computed from elastic and caloric data, the specific heat computed from $\theta \approx 11°$ seems definitely too high. Also Keesom and Westmijze (p. 220 of Ref.5) report lower values in a preliminary report. While new measurements of the specific heat around and below $1°$ would be desirable, it is possible to draw the following conclusions: In the neighborhood of the λ-point it is certainly $s_{phon} \ll s_n$. Assuming that (11) can be extrapolated to low temperatures, the curves (11) and (17) must intersect, and near absolute zero the opposite relation $s_{phon} \gg s_n$ must hold. In this extreme low-temperature region the above conclusions lose their validity. The effect of the normal component can be neglected

and the liquid becomes essentially homogeneous.[*] The main characteristic of this region is its superfluidity, no more complicated by the viscosity of the normal component and the thermomechanical effect. The transition region cannot be precisely located because of the above-mentioned uncertainties in the specific heat measurements, but it is probably between 0.6° and $1^{\circ}K$. Caloric measurements at extreme low temperatures would be desirable.

4. Macroscopic Hydrodynamics in Helium II

It was seen in the preceding section that from the λ-point down to about $1^{\circ}K$ helium II has to be considered as a mixture of two components with two distinct velocity fields; the entropy being associated only with the normal component. We will now derive the general macroscopic (coarse grained) hydrodynamic equations of this system, whereby only the so-called first approximation will be considered and dissipation effects will be neglected. In ordinary hydrodynamics the dissipation is due to viscosity and heat conductivity. In the present case there are also other effects which will be listed below. The restriction to the first approximation means in particular that non-adiabatic transitions between the two components are excluded. The hydrodynamics of our mixture is related to ordinary hydrodynamics, as a two-body to a one-body problem, and can be best handled by the so-called Lagrangian method. Since the method does not seem to be in general use in texts of hydrodynamics, a brief presentation of ordinary hydrodynamics in this form seems to be in order.

The liquid will be described by a density ρ and a vector field $\underset{\sim}{\xi}$ denoting the displacement of every elementary mass of the liquid from a standard state of constant density ρ_0. The equation of continuity in these variables is

$$\frac{\Delta\rho}{\rho} + \nabla \cdot \underset{\sim}{\xi} = 0 \tag{18}$$

where $\Delta\rho = \rho - \rho_0$ is assumed to be small compared to ρ_0. In fact, differentiating (18) with respect to time, and noting that the velocity is $\underset{\sim}{v} = \frac{d\xi}{dt} = \dot{\underset{\sim}{\xi}}$, one gets the continuity equation in its more conventional form.

The density of the kinetic energy is $\frac{1}{2}\rho\,\dot{\xi}^2$. The system possesses also a potential energy in connection with fluctuation of the specific volume v at constant entropy. The potential energy is the minimum work required to bring forth this fluctuation. Although for the present case $\Delta s = 0$, for later reference we give the general expression for the minimum work as a function of Δv and Δs. According to the well-known principles of thermodynamics this is

$$\frac{1}{2}\left(\frac{\partial^2 u}{\partial v^2}\right)_s (\Delta v)^2 + \frac{\partial^2 u}{\partial v \partial s}(\Delta v)(\Delta s) + \frac{1}{2}\left(\frac{\partial^2 u}{\partial s^2}\right)_v (\Delta s)^2 \tag{19}$$

---

[*] In particular the cooling method suggested by Kapitza[10] cannot be expected to lead to temperatures much below $1^{\circ}K$.

where u is the energy per unit mass. The first term in (19) can be written as

$$-\frac{1}{2}\left(\frac{\partial P}{\partial v}\right)_s (\Delta v)^2 = \frac{1}{2}\left(\frac{\partial P}{\partial \rho}\right)_s \left(\frac{\Delta \rho}{\rho}\right)^2 \tag{20}$$

By using (18), the Lagrangian of the system is obtained through integration over the volume

$$\mathcal{L} = \int L dV = \int \left[(\rho/2)\dot{\xi}^2 - \rho/2\left(\frac{\partial P}{\partial \rho}\right)_s (\nabla \cdot \underline{\xi})^2\right] dV. \tag{21}$$

The corresponding equation of motion is

$$\frac{d}{dt}\frac{\partial L}{\partial \dot{\xi}} + \nabla\frac{\partial L}{\partial(\nabla\cdot\underline{\xi})} - \frac{\partial L}{\partial \underline{\xi}} = 0;$$

by making use of (21)

$$\frac{d^2\xi}{dt^2} - \left(\frac{\partial P}{\partial \rho}\right)_s \nabla^2\underline{\xi} = 0. \tag{22}$$

Using $\frac{d}{dt} = \frac{\partial}{\partial t} + \underline{v}\cdot\nabla$ and (18), one sees that (22) is identical to the Euler equation:

$$\frac{\partial \underline{v}}{\partial t} + \underline{v}\cdot\nabla\underline{v} = -\left(\frac{\partial P}{\partial \rho}\right)_s \frac{1}{\rho}\nabla\rho = -\frac{1}{\rho}\nabla P \tag{23}$$

For small velocities quadratic terms in $\underline{v}$ may be neglected and (22) reduces to the wave equation for the propagation of sound

$$\frac{\partial^2\underline{\xi}}{\partial t^2} - \left(\frac{\partial P}{\partial \rho}\right)_s \nabla^2\underline{\xi} = 0 \tag{24}$$

with the well-known expression for the sound velocity

$$c_1 = \left[\left(\frac{\partial P}{\partial \rho}\right)_s\right]^{1/2} \tag{25}$$

These results will now be generalized for the case of helium II. The state of the liquid will be described by two vector fields $\underline{\xi}_n$, $\underline{\xi}_s$. The density of the kinetic energy is $\frac{1}{2}\rho_n \dot{\xi}_n^2 + \frac{1}{2}\rho_s \dot{\xi}_s^2$. The potential energy per unit mass will now be given by the full expression (19), since the specific entropy can change in a first-order process by an internal convection of the normal component.

It should be recalled that the entropy thus transferred is only the entropy s_n and for the validity of our conclusions it is essential that $s \approx s_n$; i. e., the phonon entropy should be negligible.

The first term of (19) has been transformed thermodynamically (cf. (20)). A similar transformation yields for the other terms:

$$\frac{\partial^2 u}{\partial s_n^2}(\Delta s_n)^2 = \frac{\partial T}{\partial s_n}(\Delta s_n)^2 = -\frac{\partial T}{\partial(1/s_n)}\left(\frac{\Delta s_n}{s_n}\right)^2 \tag{26}$$

and

$$\frac{\partial^2 u}{\partial s_n \partial v} = -\left(\frac{\partial P}{\partial s_n}\right)_v = -\left(\frac{\partial P}{\partial T}\right)_v \left(\frac{\partial T}{\partial s_n}\right)_v = -\rho\alpha\left(\frac{\partial P}{\partial \rho}\right)_T \frac{T}{c_v} \tag{27}$$

where α is the coefficient of thermal expansion, and c_v the specific heat at constant volume.[*] Equation (26) can be further transformed by noting that from (9) and (14)

$$-\frac{dT}{d(1/s_n)} = \frac{\rho_n}{\rho}\frac{dP_n}{d\rho_n} ; \tag{28}$$

hence

$$\rho\frac{\partial^2 u}{\partial s_n^2}(\Delta s_n)^2 = \rho_n\frac{dP_n}{d\rho_n}\left(\frac{\Delta s_n}{s_n}\right)^2 . \tag{26a}$$

The following transformation of the coordinates $\underline{\xi}_n$, $\underline{\xi}_s$ should now be introduced:

$$\underline{\xi} = \frac{1}{\rho}\left(\rho_n\underline{\xi}_n + \rho_s\underline{\xi}_s\right) \tag{29}$$

$$\underline{\eta} = \underline{\xi}_n - \underline{\xi} = \frac{\rho_s}{\rho}\left(\underline{\xi}_n - \underline{\xi}_s\right)$$

The "center of mass" coordinate $\underline{\xi}$ corresponds to the net flow of matter and has identical properties to the coordinate of ordinary hydrodynamics. In particular, it satisfies the equation of continuity (18). The "relative coordinate" $\underline{\eta}$ satisfies the equation of continuity

$$\frac{\Delta s_n}{s_n} + \nabla\cdot\underline{\eta} = 0 \tag{30}$$

This relation expresses the fact that the entropy changes uniquely by means of the flow of the normal component with respect to the "center of mass".

Equation (30) is of course an approximate relation valid only so long as the "two-component" concept can be maintained. "Sources" of entropy and of the normal component arise whenever heat is absorbed or rejected by the system and also as a consequence of dissipation effects neglected here.

If the heating occurs across a solid wall, the sources of the two components can be conveniently accounted for by the boundary conditions for the perpendicular components of $\underline{\xi}$ and $\underline{\eta}$:

$$\dot{\xi}_\perp = 0 , \qquad \dot{\eta}_\perp = \frac{W}{Ts_n\rho} \tag{31}$$

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[*] Strictly speaking, the specific heat diminished by the Debye contribution.

where W is the amount of heat absorbed by the liquid across a unit area per unit time.

Making use of the equations of continuity (18) and (30), one gets the Lagrangian in the new coordinates:

$$\mathcal{L} = \int \left[\tfrac{1}{2}\rho\,\dot{\xi}^2 + \tfrac{1}{2}\rho\frac{\rho_n}{\rho_s}\,\dot{\eta}^2 - \tfrac{1}{2}\rho\left(\frac{\partial P}{\partial\rho}\right)_s (\nabla\cdot\xi)^2 - \frac{\alpha T s_n}{c_v}\,\rho\left(\frac{\partial P}{\partial\rho}\right)_T (\nabla\cdot\xi)\,(\nabla\cdot\eta) \right.$$
$$\left. - \tfrac{1}{2}\rho_n\frac{dP_n}{d\rho_n}\,(\nabla\cdot\eta)^2 \right]\, dV \tag{32}$$

The equations of motion corresponding to (32) are

$$\frac{d^2\xi}{dt^2} - \left(\frac{\partial P}{\partial\rho}\right)_s \nabla^2\xi = \left(\frac{\partial P}{\partial\rho}\right)_T \frac{\alpha T s_n}{c_v}\,\nabla^2\eta \tag{33}$$

and

$$\frac{d^2\eta}{dt^2} - \frac{\rho_s}{\rho}\frac{dP_n}{d\rho_n}\nabla^2\eta = \frac{\rho_s}{\rho_n}\left(\frac{\partial P}{\partial\rho}\right)_T \frac{\alpha T s_n}{c_v}\nabla^2\xi \tag{34}$$

On account of the smallness of α, the right-hand sides of (33) and (34) are very small, and will be temporarily neglected. Hence in this approximation the equation in ξ and η appear separated. Equation (33) is now identical with the ordinary hydrodynamic equation (22). Equations (23) - (25) follow as before. The Euler equations for the internal convection are

$$\frac{d\dot{\eta}}{dt} = \frac{\partial\dot{\eta}}{\partial t} + \dot{\eta}\cdot\nabla\dot{\eta} = -\frac{\rho_s}{\rho}\frac{dP_n}{d\rho_n}\frac{1}{\rho_n}\nabla\rho_n = -\frac{\rho_s}{\rho\rho_n}\nabla P_n \tag{35}$$

In case the quadratic terms in the velocity $\dot{\eta}$ can be neglected, (34) reduces to the wave equation:

$$\frac{\partial^2\eta}{\partial t^2} - \frac{\rho_s}{\rho}\frac{dP_n}{d\rho_n}\nabla^2\eta = 0. \tag{36}$$

The corresponding velocity of propagation is

$$c_2 = \left(\frac{dP_n}{d\rho_n}\frac{\rho_s}{\rho}\right)^{\frac{1}{2}}. \tag{37}$$

By using (28) again, an alternate form is obtained:

$$c_2 = \left[-\frac{dT}{d(1/s_n)}\frac{\rho_s}{\rho_n}\right]^{\frac{1}{2}}. \tag{37a}$$

The physical meaning of the ξ and η waves is apparent from the continuity relations (18), (30) and from (9) and (14). It is seen that the ξ waves are accompanied by large fluctuations of the density ρ and pressure P, while the η waves are associated with fluctuations in s_n, ρ_n, P_n and T. Accordingly they have been called by the author pressure and temperature waves, respectively, while Landau called them first and second sound. The designation of temperature waves is suggestive of the simplest way of generating these waves by periodical heating. This has been actually carried out by Peshkov[7], whose results have been verified by Lane and collaborators[9] using a different method suggested by Onsager. In this method a temperature wave (second sound) generated thermally in the liquid reaches the liquid-vapor interface, where the temperature fluctuations cause periodical vaporizations and condensations. Thus ordinary sound is generated in the vapor and observed with a microphone. The boundary condition at the liquid-vapor interface is

$$Q\,\dot{\xi}_\perp = Ts\,\dot{\eta}_\perp \tag{38}$$

where Q is the heat of vaporization per unit mass.

The very existence of the temperature waves bears out the fundamental assumptions of the theory.[*] A more quantitative check of the theories is obtained by comparison of the theoretical expressions of $c_2(T)$ with the observed values. Equation (37a) together with (14) or (14a) enables us to determine this function uniquely from the knowledge of the function $s_n(T)$. Using the approximate relation (11) one obtains[**]

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[*] Recently E. G. Richardson[20] expressed the view that the temperature waves might be explained within the ordinary theory of heat conduction simply by a **large value of the coefficient of heat conductivity** χ, **and without assuming any** wave equation for the temperature. He pointed out that also the parabolic equation of heat conductivity possesses wave solutions. Actually these wave solutions are strongly damped (independently of the value of χ) and show a strong dispersion, their velocity of propagation being proportional to $\omega^{\frac{1}{2}}$ (ω is the frequency). This criterion for distinguishing between the wave solutions of a parabolic and a hyperbolic differential equation has been pointed out previously, (l.c.). The above-mentioned experiments decide the question without ambiguity in favor of the hyperbolic equation. In particular Peshkov failed to observe any dispersion in the range from 30 to 10000 c.p.s. It may be also mentioned that the true heat conductivity in helium II is proportional to the viscosity and is of the same order of magnitude in helium II as in helium I. Its presence is completely masked by the presence of the internal convection.

[**] It may be noted that formula (37) was first given by the author[2]. The explicit temperature dependence was given then in the form (39) except that a constant of the order of unity remained undetermined. A formula similar to (37a) was derived by Landau[6] who obtained $s = s_n + s_{phon}$ in place of s_n. The identity of (37) and (37a) was first noticed by F. London, (paper to be published in Proc. Phys. Soc.) who also obtained (39). The rigorous proof of the identity of (37) and (37a) seems to have been first given in this paper. The author is indebted to Dr. London for communicating his paper before publication.

$$c_2 = 26\left[\frac{T}{T_0}\left(1 - (T/T_0)^{5.5}\right)\right]^{\frac{1}{2}} \text{ m/sec}. \tag{39}$$

The plot of (36) is compared in Fig. 4 with Peshkov's observed values, and with those of Lane and his coworkers.[*] The agreement is seen to be very
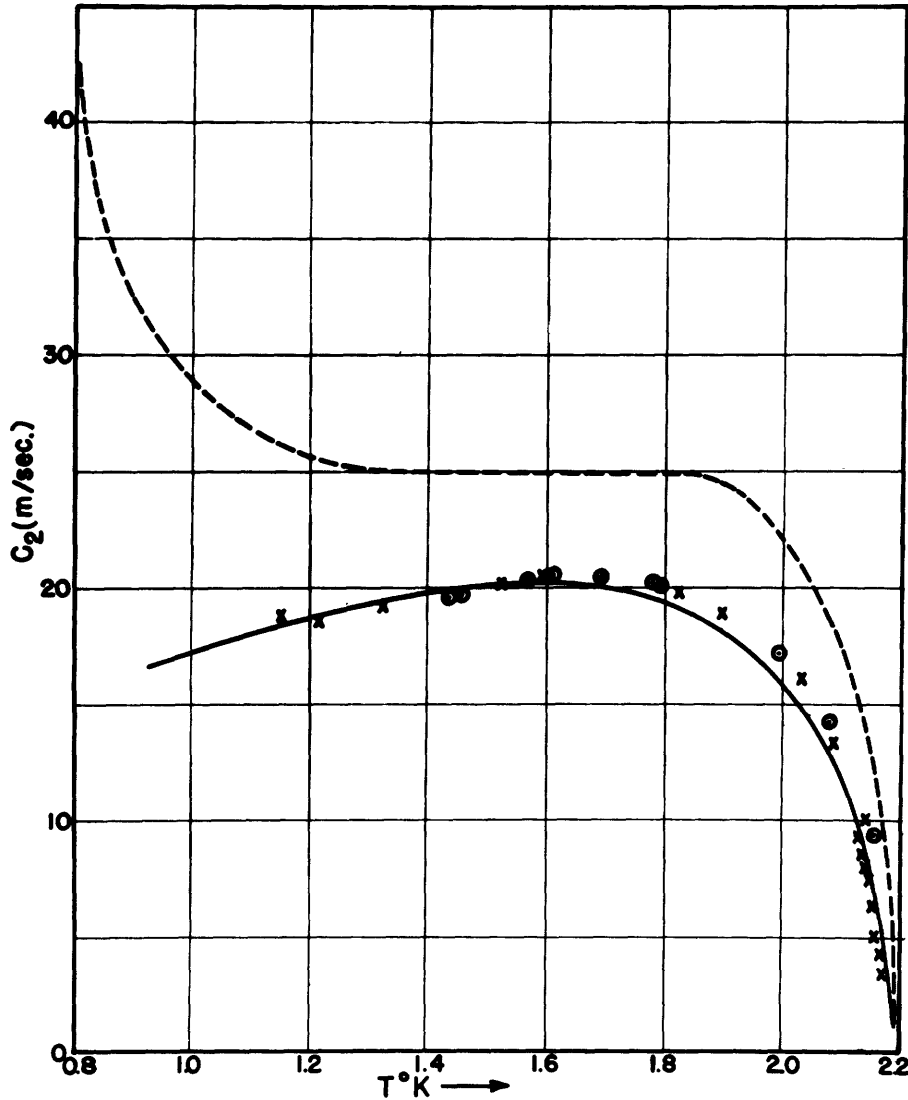

Figure 4. Velocity of second sound. xxx Measurements of Peshkov[7]; oo Measurements of C. T. Lane and collaborators ; ———— Formula (39) ------ Calculated by Lifchitz from Landau's theory[8].

satisfactory. It should be noted that the theoretical expression (39) contains no adjustable constants and is based only on the empirical entropy function $s_n(T)$ which had been measured prior to the discovery of the temperature waves.

The figure also shows the function $c_2(T)$ computed by Lifshitz[8] on the basis of Landau's theory. Landau's expression for $c_2(T)$ is a monotonically decreasing function of temperature which reaches a maximum $c_2(0) = \frac{c_1}{\sqrt{3}} \approx 130$ m/sec at absolute zero. This result is in conflict with experiment. The discrepancy stems from the role of phonons in Landau's theory. We shall return to this question in Section 6.

While the agreement between (39) and experiment bears out the fundamental assumptions of the theory, there is a noticeable discrepancy which is of interest.

Equation (38) has been derived only in first approximation,* but it would be premature to conclude that the discrepancies evident in Fig. 4 are due to higher order effects. It seems likely that these discrepancies stem from the use of expression (11) for the entropy. It was pointed out that this expression has no theoretical foundation and is merely a convenient analytic form representing the experiments to a fair degree of approximation (Fig. 3). In particular (11) represents an extrapolation of the measurements to the λ-point (Kapitza's measurements go only up to $2.10°K$). The extrapolated value $s_0 = 0.405$ cal/gm-deg enters through (14a) in the value of $c_2(T)$. It may be remarked that this value of the entropy at the λ-point is in only fair agreement with $s_0 = 0.39$ cal/gm-deg obtained from the specific heat measurements (p.234 of Ref.5). The agreement becomes somewhat worse if the latter value is corrected by deducting the phonon contribution to the entropy. Of course this value of s_0 is also based on an extrapolation (to absolute zero) and is not too reliable.

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* The effects neglected should be briefly listed. The coupling of the ξ and η waves can be easily taken into account. This effect is very small. Dissipation gives rise to absorption and dispersion of both first and second sound. In addition to dissipation due to viscosity and the true heat conductivity, there are effects characteristic of helium II. The first of these is due to non-adiabatic transitions between the two components which lead to deviations from the continuity equation (30). A second effect is due to the Debye phonons and should give rise to an absorption and dispersion of the relaxation type. This can be expected to be of importance for the second sound at low temperature ($T < 1°K$) and should eventually (at sufficiently low temperatures) lead to a complete attenuation of the temperature wave within one wavelength. In case the "molecules" of assumption (iii) should have internal degrees of freedom (vortex models), another relaxation phenomenon should be expected. The absence of dispersion of the temperature wave from 60 to 10000 cps. observed by Peshkov seems to indicate that these effects are small. A quantitative discussion of the dissipation effects is beyond the scope of this thermodynamic theory, but is of great interest from the molecular point of view.

In summing up, it may be said that the small discrepancy between the theoretical and experimental values of $c_2(T)$ is probably due to the incomplete knowledge of the entropy $s_n(T)$. More precise caloric measurements extended from the λ-point to extremely low temperatures would be desirable. The set of thermodynamic relations, in particular (9), (14), (38), seems to form a valuable criterion for the consistency of different measurements and could possibly be used to establish the thermodynamic temperature scale.

The observation of temperature waves is no doubt the most decisive quantitative justification of the fundamental assumptions of the theory. The earlier experiments on heat conductivity and capillary flow are too complicated to admit a quantitative discussion, but they can be explained qualitatively on the basis of the theory. A brief outline of such a qualitative explanation of the experiments will now be given.

Let us first consider the heat conductivity. Two heat-reservoirs of temperatures T_1, T_2 are connected by a capillary completely filled with helium II precluding any net flow of the liquid. Hence $\xi = 0$. The temperature difference $\Delta T = T_1 - T_2$ gives rise to a difference of osmotic pressure $\overline{\Delta P}_n$ and thus to a circulation in the capillary ($\dot{q} \neq 0$), whereby the normal component flows towards the cold reservoir. The current is closed by a transition of the superfluid component into the normal at the warm reservoir while absorbing the heat $s_n T$. The reverse process takes place at the cold reservoir. The efficiency of this heat transport is about $T/\Delta T$ times larger than ordinary convection. This factor may reach very high values of the order of 1000. In the steady state the flow is limited by dissipation effects neglected in the above discussion. If the dissipation were due to the viscosity of the normal component alone, the heat current should be proportional to ΔP_n and hence to ΔT. Actually the heat current increases rather like $(\Delta T)^{1/3}$ which makes it likely that the dissipation is partly of a turbulent character. This is supported by a series of experiments carried out by Kapitza[21].

The situation is even more complicated in the case of capillary flow. In the limiting case of very thin capillaries only the superfluid component can flow under the influence of a pressure head, say due to gravity. Thus a temperature difference is set up between the two ends of the capillary, the outflowing liquid being colder than the one staying behind.* The flow should thus be stopped by the arising osmotic pressure difference. Actually the increased temperature will lead to vaporization of the liquid with a corresponding cooling and transition of the normal component to the superfluid with a continuation of the flow. In this case, the assumption of adiabatic conditions is no longer justified, and in particular, the equation of continuity (30) loses its validity. In somewhat wider capillaries the normal flow

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* This was one of the first conclusions of the theory which was readily verified by experiment. J. G. Daunt, K. Mendelssohn, Nature 143, 719 (1939). The outflowing liquid is never at absolute zero as it still contains the phonon entropy.

velocity also becomes appreciable and both flows are complicated by turbulence. Consequently, the thermomechanical pressure will be smaller than that expected from Eq.(9) which was derived under the assumption of strictly semipermeable capillaries. In summing up, it is clear that the complexity of the results obtained with all but the narrowest capillaries is not surprising, and the disentanglement of the various effects from these measurements is hardly feasible.

It would seem advisable to study the dissipative effects by measuring the absorption of first and second sound whereby the complications of turbulence are avoided. On the other hand, the study of turbulence could be best carried out at low temperatures, where the concentration of the normal component it negligible and the flow is no more complicated by the thermomechanical effect and the viscosity of the normal component.

5. <u>Outline of a Microscopic Theory of Liquid Helium</u>

The conclusions drawn from the postulates (i) - (iv) are in good agreement with experiment; practically all the peculiar kinetic effects finding either a quantitative, or at least a qualitative interpretation. Thus the task of a microscopic theory is reduced to providing a quantum-mechanical foundation for these postulates. There is at present no question of deriving the postulates from first principles, but we hope to show that such postulates are not in conflict with quantum mechanics.

No special discussion is required for (i) since the presence of Van der Waals attractive and repulsive forces always assures its validity. In contrast to the universal nature of (i), the validity of (ii) is most exceptional. Systems satisfying both postulates have been called quantum liquids in Section II.

It would be desirable in this connection to answer the question: What are the requirements upon the interatomic forces which guarantee that the macroscopic system built of these atoms will be a quantum liquid? At present it does not seem possible to answer this question in such generality. One can, however, point out a peculiarity of the repulsive forces in liquid helium which seem to be at the root of this question. In order to have a finite volume at equilibrium, as postulated in (i), one requires both attractive and repulsive forces between the atoms. In an ordinary (classical) system both of these are of the Van der Waals type. In helium, however, the repulsive forces have a different origin and stem from quantum mechanical zero-point energy. The ordinary repulsive forces are of course also present, but it appears that the quantum mechanical repulsion has a longer range and that the average distance of the atoms is greater than the atomic diameter obtained from the kinetic theory.

The importance of the quantum mechanical zero-point energy in liquid helium was first recognized by F. Simon[22], who came to this conclusion through

essentially **thermodynamic reasoning.** He pointed out that in solid helium existing under a pressure of at least 25 atmospheres, the binding energy of 10 calories per mole results as a small difference between the potential energy due to the Van der Waals forces of ~ 70 calories/mole diminished by a zero-point kinetic energy of ~ 60 calories/mole. If the pressure is released, helium melts with a great increase in volume, whereby both contributions to the energy decrease to give a more favorable net binding energy of about 14 calories per mole (extrapolated to absolute zero). The zero-point energy is very sensitive to compression and hence acts as an apparent repulsive potential. It blows up the volume of the liquid to about three times the volume it would have if the Van der Waals forces were acting alone.

These ideas have been further developed by F. London [23,1]. Interpolating between limiting cases London established an expression for the zero-point energy and showed that the effect to be expected from quantum mechanics is of the right order of magnitude. For further details we refer to London's papers.

The inference from these considerations would be that the absence of microscopic rigidity postulated in (ii) is due to the quantum mechanical nature of the repulsive forces: while in an ordinary liquid the potential barriers regarding a shearing motion (isochoric rearrangement) of the molecules break down under the impact of thermal agitation, in helium this happens as a result of the zero-point motion. No satisfactory proof of this statement has been given so far. However, the following remark may prove relevant:

Consider a particle in a cubic box of volume V. The box is slightly deformed into a rectangular parallelepiped of volume $V + \Delta V$. The zero-point energy appears here as the lowest characteristic energy value E_0 of the particle. It can be easily shown that, neglecting quadratic terms in the deformation, one has

$$\frac{\Delta E_0}{E_0} = -\frac{\Delta V}{V} . \tag{40}$$

Hence, for a shearing deformation ($\Delta V = 0$), the energy is in first approximation unaffected. More generally, (40) holds for any energy level of the cubic enclosure with a characteristic function invariant under the cubic rotation $x \rightarrow y \rightarrow z$, but it does not hold if this condition is not satisfied.

Although this case is altogether too simple to be applicable to the many-body problem of liquid helium, it is hard to suppress the feeling that this selective behavior of the zero-point energy with respect to shear and compression might admit generalization to that enormously more complicated case.

We turn now to the discussion of the postulates (iii) and (iv) which is complicated by the fact that there are at present two molecular models under consideration: the Bose-Einstein and the vortex model. The latter is closely

connected with Landau's theory and will be discussed in the next section.

The Bose-Einstein model was first advanced by London[1] . He suggested that individual helium atoms can exist in liquid helium in excited translational states associated with a definite energy and momentum, in much the same way as the electrons in metals according to Bloch's theory. London has shown also that the Bose-Einstein statistics implies a "condensation in momentum space"; hence postulate (iv) appears as a theorem on the basis of this model.

The incorporation of London's hypothesis regards only the shear modes of motion and otherwise the system has the liquid characteristics as discussed above. Such a system can be conveniently called a Bose-Einstein liquid.

Its ψ function in a state of weak excitation can be written as

$$\Psi = \psi_o (1 \ldots N) \, \psi_{phon} \, \psi_{gas}$$

$$\psi_{phon} = \prod_i \psi_i (u_i) \tag{41}$$

$$\psi_{gas} = \sum_P \prod_{j=1}^{N_{exc}} e^{ik_j \cdot r_j}$$

$\psi_o(1 \ldots N)$ is essentially the characteristic function of the lowest state. It depends on the coordinates of all the atoms. The u_i are the longitudinal normal modes of the system. Every u_i involves the coordinates of all the atoms. The ψ_i are the Hermite functions corresponding to the harmonic oscillator. In contrast to this situation ψ_{gas} depends only on the coordinates of the excited atoms. (The assumption of weak excitation implies $N_{exc} < < N$). $\sum_P$ represents the summation over all the permutations of the particles.

The Bose-Einstein theory has been repeatedly criticized. The question whether the mechanism of Bose-Einstein condensation has actually anything to do with the λ-point of helium, could be definitely answered when a rigorous molecular theory has been developed which explicitly takes into account the intermolecular forces. Until such a time it is about just as difficult to disprove as to prove the Bose-Einstein theory. The claim of the critiques was, however, usually to the effect that the theory can be discarded because of lack of internal consistency. Some of this criticism, such as Keesom's[5] regarding the interpretation in l.c. of the viscosity measurements actually concerned only an erroneous application of the theory, not the theory itself. After removal of this error (see this paper, Section III), the viscosity measurements of Keesom and MacWood give a particularly striking support of the theory.

We believe that -- regardless of whether the Bose-Einstein hypothesis should be proved, or disproved at some future time -- its validity cannot be decided on the basis of the a priori arguments advanced in this connection.

The main source of misunderstanding seems to lie in the failure to appreciate the difference between a real Bose-Einstein gas and a Bose-Einstein liquid. Several attempts have been made to replace the ideal Bose-Einstein by a real Bose-Einstein gas by means of introducing a self-consistent potential. This method fails to account for the correlations between the states of the individual particles, which are all important in the case of a liquid. They are taken into account to a large extent by our postulates: the gaseous nature of the system is restricted to the shear modes of motion (isopycnic rearrangements), its compressibility is that of a liquid. This remark disposes also of the numerous attempts which tended to prove or disprove the theory by means of the "critical opalescence" arising from the large density fluctuations in a condensed Bose-Einstein gas.[24] In a Bose-Einstein liquid the density fluctuations are determined by the liquid-type compressibility (observed e.g. in the first sound), which shows no significant anomalies around the λ-point.

The fact that the Bose-Einstein liquid allows one to take into account the correlations between the excited atomic states to a satisfactory degree is due to the artifice of formulating these correlations macroscopically rather than in molecular, quantum-mechanical terms. In other words, the difficulties of the quantum-mechanical, many-body problem are not solved, but bypassed in the present theory.

Another point may be made. In the Bose-Einstein liquid one has to distinguish "collisions" of the excited atoms from "interactions" of the atoms in general.

All atoms are strongly interacting through the mechanism of the zero-point energy precluding large fluctuations of density. In other words, the factor ψ_0 in (41) is not separable in the coordinates of the particles. In contrast to this interaction one speaks of a "collision" if two or more atoms in definite translational states interact to make a transition into different states. In such processes translational energy and momentum are conserved. Obviously double collisions between excited and non-excited atoms are excluded by these conservation principles which assure an apparent independence of the two components. In particular this is the basis for the assumption, $\rho_n \bar{l} \approx$ constant, advanced in Section III regarding the mean free path.

This apparent independence of the components is no longer true if multiple collisions are considered. These supply the mechanism for the establishment of thermal equilibrium required by Bose-Einstein statistics and tend to maintain the value ρ_n/ρ appropriate to the temperature.

In the absence of a satisfactory a priori justification, it is of importance that the Bose-Einstein theory does find an a posteriori justification by furnishing a molecular model for the postulates (i) - (iv), the consequences of which are in excellent agreement with experiment. It is, of course of great importance to know whether there is another molecular model with the same properties. This will be discussed in the next section.

6. Discussion of Landau's Theory

No attempt will be made here to analyze the logical connection between Landau's microscopic quantum hydrodynamics on the one hand and his macroscopic relations on the other. These relations will be rather considered from the point of view of our quasi-thermodynamic theory and the question to be answered is: what changes and additions have to be applied to the postulates (i) - (iv) in order to obtain Landau's results.

Postulates (i) and (ii) are made tacitly by Landau. In particular the validity of (ii) is essential if the Hamiltonian of the system should depend on the density alone as implied by formula (1.10) of reference 6.

According to Landau the elementary modes of excitations are phonons for the compressional modes, and "rotons" for the shear modes of motion. Due to the general wording of postulate (iii) this is compatible with the present theory. From the molecular point of view it means that the excited atoms of the Bose-Einstein theory are replaced by "rotons". A discrepancy with the present theory arises only in connection with postulate (iv). Again there is agreement inasmuch as helium II has to be considered as the mixture of a normal and a superfluid component, but - in contrast to the present theory - Landau defines the normal component as a "gas of phonons and rotons", leaving the superfliud component with rigorously vanishing entropy.

It is seen that the theories agree sufficiently closely to account for the similarity of the results. The difference in the role attributed to the phonons, however, leads to several observable differences.

The first of these concerns the low-temperature behavior of the thermomechanical effect. Whereas, according to the present theory this effect should cease somewhat below 1°K, where s_{phon} becomes of the same order of magnitude as s_n, Landau concludes that the effect should continue down to absolute zero. Consequently Kapitza [10] suggested a cooling method based on this effect. He states that " . . . this method for obtaining low temperatures as distinguished from the magnetic method will a priori permit us to approach infinitely near absolute zero". Unfortunately, if the present theory is right, this method should not lead much below 1°K. Although we are not aware of any report of an attempt to observe this effect below 1°K, the fact that Kuerti and Simon [25] found a normal and small heat conductivity in the region of 0.2°- 0.5°K, seems to support the author's point of view.

In the temperature region above 1°K, the phonon contribution to the entropy is negligible and one might at first think that the two theories lead to identical results. Although this is true most of the time, a definite difference appears in the expression (39a) for the velocity of the temperature waves. It is seen that c_2 depends critically on the manner ρ_n tends to zero with the temperature.

Landau concludes that so far as phonons are concerned, $\rho_n \sim u_n \sim T^4$ where u_{phon} is the

phonon energy per unit mass, while the entropy is $s_n \sim T^3$. This is a violation of

the relation (14). It is easy to show that (14) always implies $c_2 \sim T^2$ near abso-

lute zero. Landau concludes for the same limit $c_2 \to \frac{c_1}{3^{1/2}} \approx 130$ m/sec. This is the

origin of the discrepancy between Landau's results and the experiments evident in

Fig. 4. A finite value of c_2 at absolute zero is also in contradiction with

Nernst's law, since the temperature waves are due to the thermomechanical effect

which according to (9) should vanish with entropy.

It is interesting to consider briefly the reasoning by which Landau

came to these conclusions. We believe the difficulty to stem from Landau's fail-

ure to notice that - - in contrast to phonons - -, vortex elements possess invari-

ant masses. Such an invariant mass is necessary for the definition of the mass

fraction ρ_n/ρ. - Landau introduces this quantity by the device of the rotating

vessel. This argument is not convincing as it tends to obtain an information on a

kinetic coefficient (viscosity) from equilibrium considerations. Whether or not the

phonons contribute to the viscosity, depends on the relaxation time for the establish-

ment of equilibruim. This question was not investigated by Landau.

We believe that Landau's treatment of the phonons has to be rejected

because of experimental evidence and on the basis of internal consistency. On the

other hand the interpretation of postulate (iii) in terms of a "roton gas" is com-

patible with the thermodynamic theory and needs further consideration. The merits

of this interpretation can be evaluated only on the basis of results which go beyond

those of the thermodynamic theory. Landau's theory contains two specific results

of this kind. According to the first $\rho_n/\rho \sim e^{-\Delta/KT}$ where Δ is an energy gap in the

vortex spectrum. The second concerns the so-called critical velocity which marks

the beginning of dissipation in the superfluid flow. According to Landau this is

of the order of 10^4 cm/sec. Both results are in conflict with experiment. It is

seen from Fig. 3. that $\rho_n/\rho \sim T^{5.5}$ is in good agreement with Kapitza's entropy measure-

ments [10]. It does not seem possible to fit the measurements with Landau's formula.

The critical velocity has also been measured by Kapitza[21]. He found values between

1 and 100 cm/sec depending mainly on the geometrical properties of the flow in a

manner which reminds one somewhat of the critical Reynolds number in the case of

ordinary turbulence.

These disagreements should not be considered, however, as a definite dis-

proof of Landau's theory. The two results mentioned are direct consequences of

Landau's assumption of a gap in the vortex spectrum of the liquid. This assumption

has no theoretical foundation and is obviously an oversimplification. The funda-

mental idea of a quantum hydrodynamical approach may well be an important contribu-

tion to the question of helium and presents also interesting theoretical problems.

To conclude this discussion, we will briefly outline a sequence of ideas

which --if further developed -- might lead to a consistent quantum hydrodynamics.

As indicated above, a standard method of dealing with the many-body problem is the method of self-consistent fields or independent particles. A second alternative is the continuum or field theoretical approach where the positions and velocities of a large number of particles are replaced by a density and a velocity field. It is well known that this method is very successful in the case of the theory of elasticity and hydrodynamics. These theories are sufficiently " coarse grained" so that the molecular structure of matter and quantum effects can be ignored with no serious error. With some limitation, however, the field theoretical method can be used also in a "fine grained" sense. The best known example is the Debye theory of specific heat. The atomic structure is taken into account in a crude but efficient way by "cutting" the short wavelength end of the spectrum. Quantum theory is introduced in the well-known fashion. The method of Bose-Einstein liquid discussed in the preceding section presents a blend of the two methods: the quantum field theoretical method is used for the compressional modes (phonons) and the independent particle method for the shear modes (isopycnic rearrangements). According to a modified Landau theory (briefly referred to as the vortex model), the field theoretical method is extended also to the shear modes. In view of the vanishing microscopic rigidity, the proper approach would be the hydrodynamics of the perfect liquid, rather than the theory of elasticity; the shear modes of motion being vortices. According to well-known theorems of Kelvin and Helmholtz, the vortex motion can be built up from vortex tubes starting and ending on the surface of the liquid or closing upon themselves. These classical results were obtained under the assumption that the velocity field is everywhere continuous. In view of the vanishing rigidity (dynamic viscosity) of a quantum liquid, this seems somewhat too special and surfaces of discontinuity in the velocity might be admitted. Such surfaces can be considered also as surface vorticities in contrast to the volume vorticities of the classical theory. If such surfaces are admitted, the variety of possible vortex motions increases: one may have e.g. rotating spheroids. If the assumption is made that the results remain valid down to rotating groups consisting only of a small number of particles, quantum theoretical methods will have to be introduced. The angular momentum will be quantized in probably much the same way as in the case of rotating polyatomic molecules. Consequently such rotating groups of atoms in quantized rotational states would play the role of the molecules of postulate (iii). It is seen that the connection with the Bose-Einstein theory is surprisingly close. The monatomic molecules of this theory have to be replaced by rotating polyatomic molecules. The condensation postulated in (iv) would not be the consequence of Bose-Einstein statistics, but would rather arise from the Boltzmann factor connected with the internal rotational energy of the molecules.

It should be emphasized that this method is a priori no more rigorous than that based on the Bose-Einstein liquid. While the independent particle approach ignores the correlations between particles, the field theoretical method overestimated them.

(We recall that the compressional modes are treated in both theories according to field theory and the divergence arises only for the shear modes.) In view of the open structure of liquid helium (the blown-up atomic volume), it is not obvious that the laws of hydrodynamics should be valid down to atomic dimensions, and the field theoretical method certainly has its limitations. In the case of the Debye theory one has to "cut" the spectrum in the well-known manner. The corresponding procedure for the vortex spectrum has still to be established.

In summing up, it may be stated that neither the Bose-Einstein liquid nor the vortex model (modified Landau theory) can be excluded as possible molecular models compatible with the present thermodynamic theory. A satisfactory development of both theories is hampered by formidable difficulties.

It would be, of course, of great interest if an experimental method could be devised of distinguishing between the theories. At present this seems to be also rather difficult. The first-order hydrodynamical effects like the propagation of second sound are independent of the nature of the "molecules" of the gas and thus cannot contribute to the decision. It seems probable that the careful theoretical and experimental analysis of second-order effects, like the absorption and dispersion of first and second sound, might decide this issue. For instance, a distinguishing feature of the vortex elements compared with the excited atoms of the Bose-Einstein theory is the existence of an internal rotational energy, which should manifest itself in an absorption and dispersion of the relaxation type. Peshkov's measurements up to 10^4 cps failed to show any such effects. The extension of the frequency range would be of interest.

Interesting crucial experiments could be carried out if the H^3 isotope obeying Fermi statistics could be sufficiently enriched to make flow experiments possible. Such experiments should show conclusively whether the Bose-Einstein statistics is of any fundamental importance for the phenomenon of superfluidity. If liquid He^3 should not be superfluid, this fact could be actually used for the enrichment of this isotope, as has been recently suggested by J. Franck.[26]

7. Conclusions

In the discussion of the properties of liquid helium, it is useful to distinguish three ranges of temperature in which the liquid shows essentially different characteristics. In addition to the well-known modifications helium I and II, a further subdivision is made within helium II. In contrast to the sharp λ-point separating the modifications I and II, the transition between the low-temperature and high-temperature domain in helium II is gradual. The transition region is somewhat below 1°K, but its location needs further experimental study. In the low-temperature region, the entropy is mainly due to the elastic phonons, whereas in the high-temperature region, the phonon contribution is negligible compared with that of the shear modes. This contribution is responsible for the anomaly in the specific heat and has a gaseous nature.

The main properties of the liquid are as follows:

<u>Helium I</u> - From the macroscopic point of view, this modification shows a "normal" hydrodynamic behavior; its coefficient of viscosity is, however, rather remarkable. In ordinary liquids, the viscosity has a negative temperature coefficient due to the "microscopic rigidity." This type of viscosity can be called <u>dynamic viscosity</u>, to be distinguished from the <u>kinetic viscosity</u> which is observed in gases and which has a positive temperature coefficient. The qualitative understanding of the properties of liquids is greatly facilitated by the concept of <u>microscopic rigidity</u>. However, the difficulties of giving to this notion a quantitative formulation seem to be responsible in no small degree for the lack of a satisfactory kinetic theory of liquids. Hence, it seems to be of interest that helium I has a kinetic and <u>no</u> dynamic viscosity, revealing the complete absence of microscopic rigidity. The comparison of ordinary liquids with helium I should be useful for the disentanglement of the gas-like and solid-like properties of liquids.

<u>Helium II, Low-Temperature Region</u> ($0 \leqslant T \leqslant 1°K$)

The main characteristic of the liquid in this temperature range is its superfluidity which implies that the liquid can slip along solid walls. This is demonstrated in spectacular creeping phenomena. Beyond a certain critical velocity, the superfluid flow may be associated with dissipation, apparently of a turbulent character.

<u>Helium II, High-Temperature Region</u> ($1°\leqslant T<2.19°K$)

In this "anomalous" region, the liquid is a mixture of a normal component (like helium I) and a superfluid component (like the low-temperature form of helium II). The main object of the present paper was to develop the macroscopic thermo-hydrodynamic properties of such a complex system. The results are in excellent agreement with experiment. Minor discrepancies may well be due to small inaccuracies in the caloric measurements, although the theory will have to be refined to include small effects neglected at present, like dissipation and the dependence of ρ_n/ρ on pressure.

The formalism developed seems to furnish valuable criteria for the consistency of measurements and might possibly be used to establish the thermodynamic temperature scale.

The principal problems for further experimental and theoretical research seem to be as follows:

1) The derivation of the postulates of the quasi-thermodynamic theory from first principles; in particular, establishing the correct molecular model which would lead to these postulates. At present, the choice seems to be between the Bose-Einstein liquid and the vortex model (modified Landau theory.) The experimental approach to this problem has been briefly discussed at the end of Section 6.

2) <u>The clarification of the nature of the pure superfluid liquid (helium II near absolute zero.</u>) This problem has not been discussed in the present paper. According to Landau, the superfluid state is characterized by the condition curl $\mathbf{v}_s$ = 0 The question has been further discussed by F. London[17] and Onsager[27]. So far, superfluidity (capillary flow and the creeping phenomenon) has been studied

experimentally only above 1°K where the effects have been greatly complicated by the thermomechanical effect and the viscosity of the normal component. Experiments below 1°K where these effects vanish would be greatly desirable. Of particular interest is the investigation of the dissipation arising above the critical velocity. This seems to be of turbulent character; hence, a unique opportunity is offered to study turbulence in the absence of viscosity (no boundary layer). On the other hand, this turbulence will be influenced by quantum effects.

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