

# PROGRESS IN LOW TEMPERATURE PHYSICS

edited by

C. J. GORTER

Professor of Experimental Physics  
Director of the Kamerlingh Onnes Laboratory, Leiden

VOLUME I

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## CHAPTER I

THE TWO FLUID MODEL FOR SUPERCONDUCTORS  
AND HELIUM II

BY

C. J. GORTER

KAMERLINGH ONNES LABORATORIUM, LEIDEN

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## 1. Introductory Remarks

Though the experimental and theoretical investigation of caloric, magnetic, electric and mechanical properties of matter at low temperatures has led to many interesting and important conclusions, it cannot be denied that the most exciting discoveries in this field have been that of superconductivity by Kamerlingh Onnes in 1911<sup>1</sup> and that of liquid helium II by Keesom 16 years later<sup>2</sup>. The superconductors and helium II confront us with many complicated phenomena as well as with fundamentally unsolved problems. The two fluid model has allowed us to create some order among the phenomena while the unsolved problems may be considered to concern largely the interpretation of that model.

## 2. Some Properties of Superconductors

Superconductivity is the frictionless motion of electrons in certain metals. It was not until 1933 when it was recognized<sup>3</sup> that superconductors have a second fundamental property, namely that frictionless surface currents tend to screen off external magnetic fields in such an effective way that the magnetic induction vanishes in a massive fragment of superconductive metal.

In the simplest geometrical arrangement - a thick needle shaped sample in a longitudinal external magnetic field  $H$  - this second

fundamental property leads to a magnetic term  $H^2V/8\pi$  in the free energy, where  $V$  is the volume of the sample. As soon as the sum of the free energy of the superconductor and this magnetic term becomes larger than the free energy of the normal phase at the same temperature, the latter will become thermodynamically stable and superconductivity is disturbed <sup>4</sup>.

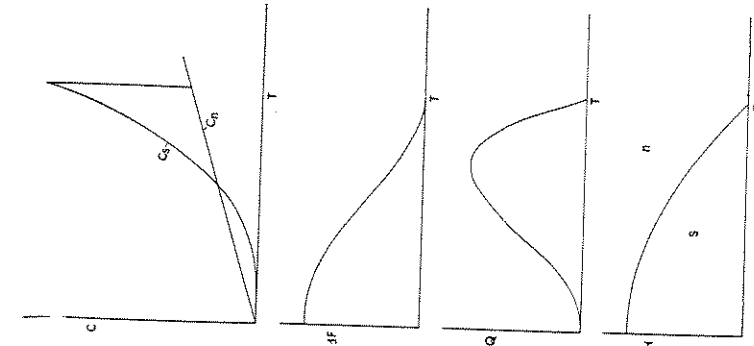


Fig. 1. Specific heats, difference in free energy, heat of transition and threshold curve

The electronic specific heat of a normal metal is given by

$$c_n = \gamma T, \tag{1}$$

where  $T$  denotes the temperature and  $\gamma$  a constant characteristic of the metal (cf. Ch. XI).

With a reasonable accuracy the specific heat of a superconductive metal - duly corrected for the lattice specific heat - is found to be given by

$$c_s \approx 3\gamma T^3/T_c^2. \tag{2}$$

This leads to the following approximate expression for the difference in free energy of the two phases <sup>5</sup>

$$\Delta F = F_n - F_s \approx \Delta U_0 - \frac{1}{2}\gamma T^2 + \frac{1}{4}\gamma T^4/T_c^2, \tag{3}$$

where  $\Delta U_0$  indicates the energy difference between the two phases at zero temperature. The condition  $\Delta F = 0$  for the critical temperature  $T_c$  gives <sup>6</sup>

$$T_c \approx 2 \left( \frac{\Delta U_0}{\gamma} \right)^{\frac{1}{2}}. \tag{4}$$

$$\Delta F = H_{thr}^2 V / 8\pi \tag{5}$$

Now

gives a parabolic magnetic threshold curve (See Ch. VII)

with the threshold field at zero temperature

$$H_0 = \left( \frac{8\pi \Delta U_0}{V} \right)^{\frac{1}{2}}$$

All superconductors obey with a reasonable accuracy the equations provided with the approximate sign. They thus obey a sort of law of corresponding states <sup>7</sup> and we may characterize every superconductor by one constant only. It is reasonable to choose  $\Delta U_0$ , the energy difference between the normal and the superconductive phase at zero temperature, to be this constant. The critical temperature is then given by (4), where  $\gamma$  is a constant characteristic of the normal phase.

At  $T_c$  we have an extraordinarily well-defined transition of the second order in Ehrenfest's sense <sup>8</sup> and the remarkable property of such a transition is that not only the free energies, but also the energies and the entropies of the two phases are equal. In presence of an external magnetic field, however, we have a transition of the first order with a heat of transition

$$Q \approx 4\Delta U_0 \frac{(H_0 - H_{thr}) H_{thr}}{H_0^2} \tag{7}$$

Finally one may remark that the specific heats and the entropies concerned are of the order of  $\gamma T_c$ , which amounts to only  $10^{-3}$  R per gram atom.

Among the other properties of superconductors we might mention that the thermoelectric effects are zero and that the heat conduction (in pure metals) is smaller than in the normal phase, though at  $T_c$  there is no jump (See Ch. X).

### 3. Some Properties of Liquid Helium II

Liquid helium has a jump in the specific heat <sup>9</sup> at its so-called  $\lambda$ -temperature  $T_\lambda$ . This change is of the order of  $R$  and thus much higher than that in superconductors. Its magnitude is also less well-defined and a distinct tail extends above the  $\lambda$ -temperature.

The almost frictionless motion of the liquid below the  $\lambda$ -temperature - the so-called helium II - immediately suggests an analogy with

the motion of electrons in a superconductor. There is no apparent analogy to the action of an external magnetic field on a superconductor, so the complications connected with different shapes are lacking while alloying is only possible with the isotope  $^3\text{He}$  (See Ch. VI).

But in other respects helium II exhibits more varied and curious properties than superconductors do. When almost frictionless motion

occurs through a narrow slit, heat is produced in the vessel left by the liquid while an equal amount of cold is developed in the vessel into which the liquid flows<sup>10</sup>. This is the mechano-caloric effect and the heat or cold per unit mass of liquid passing is called the heat of transport  $Q^*$ . If we introduce a temperature gradient along the slit a very high heat flow is found. At the same time a flow of matter takes place towards the high temperature side, which may lead to a helium fountain of considerable height<sup>11</sup>. This fountain height or rather the pressure gradient which is able to counterbalance the flow of matter does not depend on the dimensions of a very narrow slit.

With the aid of irreversible thermodynamics<sup>12</sup> De Groot has proved<sup>13</sup> that

$$\text{grad } \dot{p} = \frac{\rho Q^*}{T} \text{ grad } T, \quad (8)$$

where  $\rho$  is the density, as long as the transports of matter and heat are linear in the gradients of temperature and pressure.

The most remarkable property of liquid helium II is, however, its capability to propagate heat waves and heat pulses with only a slight attenuation<sup>14</sup>. This so-called "second sound" is generated by an alternating or pulsed supply of heat. The velocity of propagation may be measured with great accuracy and is found to be between  $18\frac{1}{2}$  and  $20\frac{1}{2}$  meters per second between  $1.0^\circ\text{K}$  and  $1.9^\circ\text{K}$  while the velocity of normal sound is more than ten times higher.

#### 4. Behaviour near Zero Temperature

Near zero temperature superconductors and liquid helium have passed into an extraordinary state of matter which is characterized by the possibility of frictionless motion of electrons and helium atoms respectively. It is remarkable that the entropy of this state vanishes with a high power of  $T$ . The energies associated with this decrease of entropy are many times larger in helium than in superconductors. When the frictionless motion is set up the energy increases, the entropy remaining zero. The non-reversible transition to zero motion is extremely slow. Frictional processes due to scattering of electrons by impurities and friction between the helium and the walls have ceased to occur. Apparently we are not concerned with individual motion of electrons or helium atoms, but with moving systems composed of many individuals<sup>15</sup>. No convincing explanation has been offered on the nature of these systems and the conditions under which they may occur.

#### 5. The Services Rendered by the Two Fluid Model

The so-called two fluid model is useful in the description of the phenomena occurring between the absolute zero and the transition temperatures  $T_0$  and  $T_\lambda$ . In this model an internal parameter is introduced which may vary between 0 and 1 only. In one of the two current versions<sup>16</sup> this parameter is called  $x$  and it is supposed to be zero at zero temperature and one at the transition temperature. In the other version<sup>17</sup> the parameter  $\omega = 1 - x$  varies from one to zero in the same interval. The existence is assumed of two interpenetrating fluids, each possessing its own velocity field and its own inertia. There is no or at most very little exchange of momentum between the fluids. One of the fluids, the superfluid, bears the frictionless motion; its last trace disappears at the transition temperature when  $x = 1 - x = 0$ . The other fluid, the normal fluid, behaves more or less as a normal electron gas or a normal liquid: it is scattered by irregularities and heat waves or it has a normal viscosity and adheres to a wall. It rapidly disappears with decreasing temperature when  $x = 1 - \omega \rightarrow 0$ . The relative amounts of the fluids, whatever this may mean, are taken to be  $\omega = 1 - x$  and  $x = 1 - \omega$ .

Without any further definition of quantitative elaboration this two fluid model can offer a simple description of many of the exceptional properties of superconductors and helium II<sup>18</sup>.

In the frictionless motion of helium through a narrow slit it is clearly the superfluid which moves. In the vessel left by the superfluid its relative concentration  $1-x$  decreases while it increases in the vessel on the other side of the slit. Keeping the relative concentration constant requires a supply of heat and cold respectively (mechano-caloric effect).

Introduction of a temperature gradient introduces a gradient of the internal parameter  $x$  which leads to an internal force, sometimes

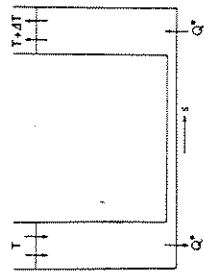


Fig. 3. Mechano-caloric effect

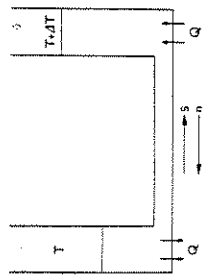


Fig. 5. Heat conduction by convection of the two fluids

called the diffusion force, driving the normal fluid towards the low temperature side and the superfluid to the high temperature side. The resulting convection of the fluids accounts for the heat conduction. In narrow slits the motion of the normal fluid is impeded by its adherence to the wall and its viscosity. The fountain effect then clearly demonstrates the diffusion force acting on the superfluid.

It is found that in relatively wide channels and high temperature gradients the heat flow as well as the fountain pressure increase con-

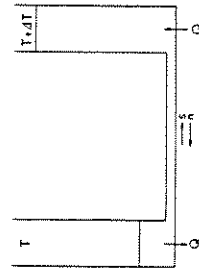


Fig. 4. Fountain effect

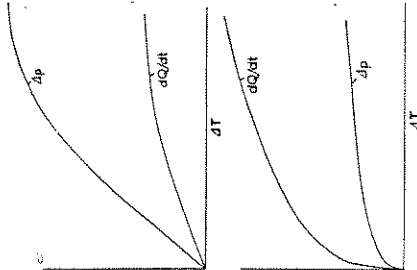


Fig. 6. Heat flow  $dQ/dt$  and fountain pressure in a narrow slit (above) and a wide slit (below)

siderably slower than if they were proportional to the temperature gradient. Under high temperature gradients the heat flow and the fountain effect<sup>19</sup> are approximately proportional to  $(\text{grad } T)^{1/2}$ , while the transition from this cube root dependence to the linear dependence at very small temperature gradients is exactly similar for the two effects. (Allen and Reekie's<sup>20</sup> rule). This behaviour may be accounted for by assuming a mutual friction between the two fluids which is approximately proportional to the cube of the relative velocity of the two fluids<sup>21</sup>. The existence of such a mutual friction is corroborated by the occurrence of a pressure gradient proportional to the cube of the superfluid velocity through a slit of a few microns width.

The very simple equations of motion for the superfluid and the normal fluid:

$$-(1-x) \text{ grad } P + x(1-x) S^* e \text{ grad } T - Ax(1-x) \rho^2 |v_s - v_n|^2 \\ (v_s - v_n) = \rho(1-x) d\mathbf{v}_s/dt \quad (9)$$

$$-x \text{ grad } P - x(1-x) S^* e \text{ grad } T + Ax(1-x) \rho^2 |v_s - v_n|^2 (v_s - v_n) \\ + \eta_n (4\mathbf{v}_n + (1/3) \text{ grad div } \mathbf{v}_n) = \rho x d\mathbf{v}_n/dt \quad (10)$$

have often been used in the discussion of the behaviour of helium II. In these equations  $v_s$  and  $v_n$  are the velocities of superfluid and normal fluid,  $P$  is the pressure, while  $S^*$  varies only slowly as a function of temperature. It has the dimensions of entropy per unit mass and characterizes the diffusion force (cf. §§ 6 and 7). The domain of the usefulness of (9) and (10) is limited. They account satisfactorily for the transport phenomena in slits and capillaries of widths from 5 microns upwards, at not too low velocities. There are even indications that at very low velocities, particularly in very narrow slits, no mutual friction would occur below a critical velocity<sup>22</sup>. Experiments with slits of the order of 1 micron suggest also a considerable decrease in the normal viscosity  $\eta_n$ , while on the other hand investigations with oscillating discs and rotating cylinders (See Ch. IV) reveal the existence of more forces<sup>23</sup> than are present in (9) and (10). We should like to point out that as a consequence of (9) the superfluid velocity  $v_s$  should in general have a curl, while usually the possibility of turbulence is not considered. The condition — perhaps not implausible on theoretical grounds<sup>24</sup> (See Ch. II) — that  $v_s$  should be rotation-free cannot very easily be introduced in an equation of the type (9). The most remarkable success of the two fluid model was the prediction<sup>25</sup> of

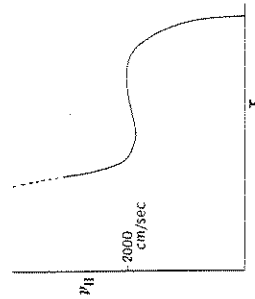
second sound which is interpreted as a wave in the internal parameter  $x$ , the normal and the superfluid oscillating in respect to each other, while the total density  $\rho$  remains constant. From (9) and (10) we get for the velocity of propagation  $v_{II}$ , if we suppose that no periodic transition from superfluid to normal fluid and vice versa occurs,

$$v_{II}^2 = \frac{x(1-x)S^*}{d\kappa/dT}, \tag{11}$$

while the supposition that at any moment the value of  $x$  is adjusted to the momentary value of  $T$  gives

$$v_{II}^2 = \frac{x(1-x)S^{*2}T}{c}, \tag{12}$$

Fig. 7. Velocity of second sound  $v_{II}$  as a function of  $T$ .



where  $c$  is the specific heat per unit mass. At low velocities the normal fluid only exchanges momentum with the walls. This gives the possibility of eliminating friction by choosing a wide channel and to observe in this way second sound and the high heat conduction. On the other hand the normal fluid may be immobilized by choosing a very narrow channel and thus the fountain effect and the mechano-caloric effect may be observed.

In the case of superconductors we do not have the possibility to vary the friction just mentioned, since the normal electrons do not exchange momentum so much with the walls as with irregularities and the heat waves of the crystalline lattice. This leads to a volume friction which would damp down second sound within a fraction of a wavelength and would also impede an energy flow by convection of the fluids. Also several other effects similar to the exceptional phenomena in helium must be so small in superconductors that they are very difficult to observe. Thus the two fluid model renders less brilliant services for superconductivity than for liquid helium II.

However, the losses occurring in the surface screening layer under the influence of high frequency fields confirm the presence of normal electrons in the superconductive phase particularly at temperatures not too much below  $T_c$ . According to London's formulae<sup>27</sup> the thickness of the layer must be inversely proportional to the square root of the density of the superconductive electrons. It is in agreement

with the expectations from the two fluid model that this thickness is found to increase rapidly when  $T_c$  is approached<sup>28</sup>.

6. The Free Energy as a Function of the Internal Parameter

The thermodynamician is inclined to consider the free energy as a function of the internal parameter  $x$  and to state that the value found for this parameter at each temperature must be determined by the minimum of the free energy as a function of the parameter at that temperature. The prototype of such a consideration is the case of a one component - two phase system at constant volume, e.g. of vapour and liquid enclosed in a box. The free energy of this system is the sum of the free energies of the two phases. That of the liquid (supposed to be incompressible) is proportional to its mass  $1 - x$ , that of the vapour contains a term pro-

portional to  $x \ln x$ . At any temperature  $x$  will adjust itself according to the minimum of the non linear total free energy and consequently  $x$  will, with rising temperature, gradually increase from zero to one, which value is reached where  $(\partial F/\partial x)_{x=1} = 0$ . At that temperature the system as a whole exhibits a second order transition.

In 1934 the following expression was proposed<sup>29</sup> for the electronic free energy of a superconductor:

$$F = xAU_0 - \frac{1}{2}x^\alpha\gamma T^2, \tag{13}$$

which gives the electronic free energy for the normal metal if  $x=1$ , and where the factor the  $x^\alpha$  provides for the required non-linearity in  $x$ . If  $\alpha = \frac{1}{2}$  the minimum of this free energy leads to the equations (3), (4) and (6), which approximately describe the experimental data. We also find

$$x = (T/T_0)^\alpha, \tag{14}$$

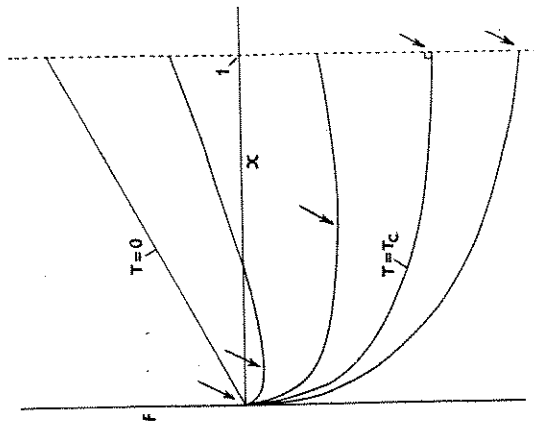


Fig. 8. Free energy of a superconductor as a function of  $x$  according to (13) for different values of  $T$ .

7. The Formule of H. London and Tisza for Helium II

Quite another line of thought is followed by others, who advocate that one should derive the value of  $x$  and the magnitude of the heat of transport etc. directly from a statistical or quantum mechanical model or theory without having recourse to non equilibrium values of  $x$  and the process of minimizing the free energy.

As long as a satisfactory theory is lacking one might then not only ascribe separate velocity fields and inertia to the two fluids, but also separate thermodynamical quantities and in particular give the superfluid the entropy zero. The superfluid entering a vessel with zero entropy needs to be lifted to the entropy  $S$  appropriate to the temperature of the vessel. If this happens reversibly, we obtain (as does H. London<sup>36</sup>)

$$Q^* = TS \tag{19}$$

which by (8) determines the fountain effect while the corresponding expression for the diffusion force has  $S/x$  for  $S^*$  in (9) and (10).

A somewhat more specialized description has been proposed by Tisza<sup>38</sup> who supposes that neither the normal nor the superfluid has a specific heat, so that the entropy below  $T_\lambda$  is entirely due to the temperature independent specific entropy of the normal fluid. Thus

$$S = xS_\lambda \tag{20}$$

Between 1.1°K and  $T_\lambda$  both assumptions are, with a reasonable degree of accuracy, verified by experiments on the fountain effect, the mécano-caloric effect, the velocity of second sound<sup>37</sup> and experiments with stacks of oscillating discs<sup>38</sup> (See Ch. IV).

The expression (15) has the advantage that in connection with (18) it leads to equations (19) and (20) which have in general been confirmed by experimental results.

8. Theoretical Background in the Case of Helium II

The theoretical background of the two fluid model is still obscure. For the case of helium there are two very different proposals advanced by F. London and Tisza<sup>39</sup>, and by Landau<sup>40</sup>, respectively.

London and Tisza describe the separation into two fluids as a Bose-Einstein condensation while Landau considers the normal fluid to consist of excitations, of which he distinguishes two kinds: quantized

an expression which later was found to be in good agreement<sup>39</sup> with the dependence of the thickness of the surface screening layer which presumably should be proportional to  $(1-x)^{-1/2}$ . Encouraged by this success a somewhat similar proposal<sup>31</sup> was made for the approximate Gibbs free energy  $G$  of liquid helium

$$G = x^{1/2} AU_0 - xS_\lambda T, \tag{15}$$

which leads to

$$x = S/S_\lambda = (T/T_\lambda)^6 \tag{16}$$

Though this expression excels because of its simplicity and has been used for several calculations aimed at obtaining a rough interpretation of data, it is unsatisfactory in that the specific heat of the normal state ( $x = 1$ ), which is quite important above  $T_\lambda$ , has been suppressed. Therefore the somewhat more complicated expression

$$G = x^{1/2} AU_0 - \frac{1}{2} \frac{x^{1/2} S_\lambda T^2}{T_\lambda} \tag{17}$$

has sometimes been used<sup>32</sup> and also leads to (16). The quantitative differences in the conclusions derived from (15) and (17) are remarkably small.

Following this thermodynamic line of thought the following expression has been proposed<sup>33</sup> for the heat of transport:

$$Q^* = Tx \left( \frac{\partial S}{\partial x} \right)_T = -Tx \left( \frac{\partial^2 G}{\partial x \partial T} \right), \tag{18}$$

which by (8) determines the fountain effect. The corresponding expression for the diffusion force has  $(\partial S/\partial x)_T$  for  $S^*$  in (9) and (10). However, the two fluid model is apparently not sufficiently sharply defined, so the validity of (18) is still a controversial matter<sup>34</sup>.

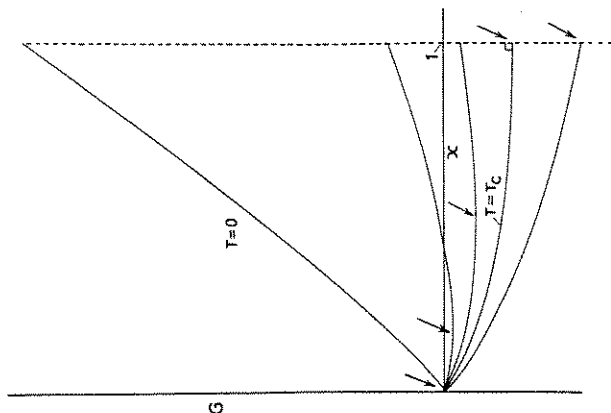


Fig. 9. Gibbs free energy of helium II as a function of  $x$  according to (15) for different temperatures

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compressional waves (phonons), and excitations with a minimum energy, called rotons (See Ch. II).

In the neighbourhood of the  $\lambda$ -temperature both views lead to difficulties. For an ideal gas the Bose-Einstein picture predicts a jump here in the derivative of the specific heat rather than in the specific heat itself and more or less artificial additional assumptions<sup>41</sup> are required to adjust this discrepancy. On the other hand it is difficult to see how, in the phonon-roton picture, an assembly of those excitations can gradually approach the whole normal liquid at the  $\lambda$ -temperature.

Landau predicted a pronounced change in the properties of the liquid at about 1°K, below which the rotons would rapidly vanish. The confirmation of this change by the steep increase of the velocity of propagation of heat pulses in helium II was a triumph for his interpretation<sup>42</sup>. The formulation of his views, however, was rather vague in so far as the phonons and rotons are concerned. Kramers' and Kronig's analysis<sup>43</sup> would give a sound basis of the two fluid model also confirming H. London's formula (19), but unfortunately it is not sure whether their treatment is applicable at the higher temperatures too. At the lower temperatures, where the phonons apparently are dominant, there is little use for the two fluid model since direct description by means of phonons of long mean free paths seems more appropriate<sup>44</sup>. If one could experiment with cubic kilometers of helium instead of with cubic centimeters, this might be different. Feynman's recent analysis<sup>45</sup> of the rotons, however, gives hope that the Landau treatment may be worked out and confirmed for the temperatures above 1°K too (See Ch. II).

### 9. Theoretical Background in the Case of Superconductivity

Heisenberg and Koppe<sup>46</sup> have proposed a statistical background for the two fluid model for superconductors. They assume that the free electron levels just above the Fermi limit may combine into a condensed state which has the properties of the superfluid. Condensation into this state is accompanied by a decrease of energy. Supposing

that electrons may occupy free electron levels as well as enter the condensed state and that the electrons in this latter state contribute zero to the entropy, the total free energy may be obtained as a function of  $T$  and of the number of condensed electrons  $\omega = 1 - x$ . Not too far below  $T_c$  there is little difference with (13), but at low temperatures there are deviations which lead to lower  $x$ -values than given by (16). This latter behaviour seems to be confirmed by data on Sn, Nb and a few other superconductors<sup>47</sup>. Heisenberg and Koppe originally suggested that electrostatic interaction could account for the condensation phenomenon, but the isotope effect (cf. Ch. VII) made it plausible that, in agreement with views expressed by Fröhlich and Bardeen, the condensation is due to interaction through the intermediary of lattice waves.

Marcus and Maxwell<sup>48</sup> have recently analyzed the criteria which the statistical background must fulfill in order to lead to the two fluid model, but they were not able to establish a satisfactory connection with the theoretical treatment of Fröhlich and Bardeen.

### 10. Final Remarks

Several years ago Daunt, Mendelssohn and F. London<sup>49</sup> discussed the analogies between superconductors and liquid helium. Aside from making many remarks repeated in the present article, they stressed the analogy between the maximum transport rate occurring when a helium II film creeps on the surface of a solid and the maximum transport rate of the surface screening currents on a superconductor.

In the first case this transport is found to be of the order of  $10^{-5}$  gr/cm sec while in the second one it amounts to  $mcH_{\text{lim}}/4\pi e$ . In both cases it decreases linearly to zero when the temperature approaches the transition temperature. A connection of these quantities with quantum theory was suggested by observing that the transport rates are of the order of magnitude of Planck's constant  $h$  divided by the volume per helium atom or per superconductive electron respectively.

As attractive as this suggestion was at first sight, it must be stated that no recent advance has been made in its elaboration. On the contrary, it is not easy to see why the energy difference  $\Delta U_0$ , on which  $H_0$  and the transport rate of superconductors depend (cf. (5)), could be connected with  $h$  in a rather complicated way. Moreover, experimentally the values of  $H_0$  seem not to be simply connected with the number of superconductive electrons.



The connection often sought between the superfluid properties in helium II and the Bose-Einstein condensation has incidentally led to the suspicion that in superconductors one might also be concerned with even units *viz.* with electron pairs. This suggestion has likewise had little success so far.

In the present contribution it has been stressed that many apparently conflicting and incoherent data find an easy interpretation or at least a description with the help of the two fluid model. This model has moreover rendered remarkable results in the prediction of the mechano-caloric effect and of second sound. On the other hand it must be admitted that it suffers from vagueness and ambiguity which is, for instance, reflected in the uncertainty about the equations of motion of the fluids in helium II. This vagueness will possibly remain until the theoretical foundation of the two fluid model has acquired solidity.

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## CHAPTER II

## APPLICATION OF QUANTUM MECHANICS TO LIQUID HELIUM

BY

R. P. FEYNMAN

CALIFORNIA INSTITUTE OF TECHNOLOGY, PASADENA, CALIFORNIA

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## 1. Introduction

Liquid helium exhibits quantum mechanical properties on a large scale in a manner somewhat differently than do other substances. No other substance remains liquid to a temperature low enough to exhibit the effects. These effects have long been a puzzle. It is supposed that they can all be ultimately understood in terms of the properties of Schrödinger's equation. We cannot expect a rigorous exposition of how these properties arise. That could only come from complete solutions of the Schrödinger equation for the  $10^{23}$  atoms in a sample of liquid. For helium, as for any other substance today we must be satisfied with some approximate understanding of how, in principle, that equation could lead to solutions which indicate behavior similar to that observed.

Since the discovery of liquid helium considerable progress has been made in understanding its behavior from first principles. Some of the properties are more easily understood than others. The most difficult of these concern the resistance to flow above critical velocity. If we permit some conjectures of Onsager<sup>1</sup>, however, perhaps a start has been made in understanding even these. The aim of this article is to describe those physical ideas which have been suggested to explain the behavior of helium which can most easily be related to properties of the Schrödinger equation.

We shall omit references to the phenomena involved in the Rollin