Some measurements in liquid helium four

J. M. Goldschvartz

# SOME MEASUREMENTS IN LIQUID HELIUM FOUR



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Grateful acknowledgements are made to all those of the Department of Applied Physics of the Delft University of Technology who made possible the accomplishment of this thesis.

To my wife and to my daughter.

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8	§ 2.2	The fountain effect, the two fluid model.	The fountain effect and the two fluid model.
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42	Fig. 2.1	The fluid model	The two fluid model
44	Equation	$g(P_A, T_A) = g(P_B, T_B) + g$	$g(P_A,T_A)=g(P_B,T_B)=g$
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57	Refe- rence (4)	Concon, E.O.	Condon, E.O.
64	Refe- rence (3)	Smit, J.	Smidt, J.
77	26	Thus must be inves- tigated and experi- ments which may or may not be signifi- cant should be re- ported.	Thus one must do and report experiments which may or may not be significant.
78	last	this particular one will be	this particular will be
166	16	an extension to	of
172	25	After the six equations.	The curve obtained was not an exponen- tial but the same one as mentioned above, $E_0 = Cd^a$ .

#### PREFACE

Liquid helium is a very peculiar and conspicuous substance. We could say that it represents a dramatic manifestation of nature revealed by science. Particularly this is true for the phase of liquid helium four (<sup>4</sup>He) called "liquid helium two" (liquid <sup>4</sup>He II) with its superfluid component which has no entropy, no viscosity, presents no friction and exhibits a notable zero point energy. A fundamental feature of the superfluid liquid helium four is that it is a quantum liquid, i.e., has quantum properties. Liquid helium three is a quantum liquid too although it does not present the phenomenon of superfluidity.

A part of the Low Temperature Group of the Department of Applied Physics of the Delft University of Technology is doing research on liquid helium four.

This thesis is the result of this research. That is, it gathers the set of papers published in the last five years. In chronological order these papers are:

J.M. Goldschvartz and B.S. Blaisse, A new test-cell for the electrical breakdowns of liquid helium, Cryogenics 6 (1965) 169.

J.M. Goldschvartz and B.S. Blaisse, Electrical breakdown of liquid helium, British Journal of Applied Physics 17 (1966) 1083.

J.M. Goldschvartz, Modified helium dewar, Cryogenics 8 (1968) 108.

J.M. Goldschvartz, F. Landheer and B.S. Blaisse, Destruction of electrodes in the electrical breakdown of liquid helium, Appl. Sci. Res. <u>19</u> (1968) 14.

J.M. Goldschvartz and B.S. Blaisse, A silicon carbide superleak, Proc. 2nd. ICEC, Brighton, U.K. (1968) 304.

J.M. Goldschvartz and B.S. Blaisse, Parameters involved in the electrical breakdown of liquid helium, Inst. Intern. du Froid; Commission I - London (1969) Annexe 1969-1, 367.

B.S. Blaisse, J.M. Goldschvartz and P.C. Slagter, A method of measuring ionic mobilities in high electric fields in isolating liquids, Cryogenics 10 (1970) 163.

J.M. Goldschvartz, E. Martin and B.S. Blaisse, Onset point of a new natural superleak, Cryogenics 10 (1970) 160.

J.M. Goldschvartz, A.C. Ouwerkerk and B.S. Blaisse, On the electrical breakdown of liquid helium, I.E.E. (London) Conference Publication no. 67 (1970) 218.

J.M. Goldschvartz, E. Martin and B.S. Blaisse, New Superleaks, Proc. 3rd. ICEC Berlin, (1970) 496.

B.S. Blaisse, J.M. Goldschvartz and M. Naeye, Mobility of negative ions in liquid helium I and helium II, Read at the 12th International L.T. Conference held at Kyoto, September 1970. In press.

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J.M. Goldschvartz and B.S. Blaisse, New Materials for superleaks, Inst. Inter. du Froid, Commission I, Tokyo (1970) Annexe 1970-2, 231.

The papers are presented "in extenso" and completed with the necessary information in order to obtain a good approximation of an organic, complete, comprehensible and consistent whole.

Finally, I would like to point out that in the following text the expression "liquid helium" means liquid helium four. Otherwise, it will be clearly indicated.

## SOME MEASUREMENTS IN LIQUID HELIUM FOUR

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CHAPTER I

#### LIQUID HELIUM

#### 1.1. Introduction.

In the year 1908 Kamerlingh Onnes liquified the gas helium and three years later discovered the phenomenon of superconductivity. These two facts and dates are a remote beginning of what is called physics of the very low temperatures. The boiling point of liquid helium four establishes the upper limit of this temperature region. The physics of the very low temperatures gave birth to presentday technology with its sophisticated techniques.

Thanks to the development of the simple and compact modern helium liquifiers, liquid helium is now available in a great number of physics laboratories all over the world where the physics of very low temperatures is studied. Following Mendelssohn (1961) one can say that the low temperature physics and techniques are divided in two periods:"before and after the advent of the commercial liquifiers".

The boiling points of liquid <sup>4</sup>He and liquid <sup>3</sup>He are 4,2 K and 3,2 K, respectively. One can obtain lower temperatures by reducing the vapour pressure which pumps out the vapours. Then the lowest temperature obtainable in this way is 0,35 K

for liquid <sup>3</sup>He. These limits are established by a mechanical reason: the speed of the pumping systems and in the case of <sup>4</sup>He, by the so-called film creep too. Below a given temperature a thin film of liquid helium creeps up quickly on the surface of any solid in contact with the liquid. When this film reaches a region of sufficiently high temperatures it evaporates and at the same time introduces in the system a big heat leak due to the high thermal conductivity of the liquid at that temperature. In turn, this heat leak increases the evaporation of the bulk of the liquid helium.

In Figure 1.1 are plotted the vapour pressure temperature curves for liquid helium three and four. Observing these curves one sees why the temperature range of the helium three is lower than the one of the helium four: the vapour pressure of liquid helium three at any given temperature is higher than the vapour tension of liquid helium four at the same temperature.

Among the four isotopes of the element helium, helium three and helium four are the two stable isotopes and helium four is the most abundant in nature. The abundance ratio between this and the lighter isotope <sup>3</sup>He is of the order of 10<sup>7</sup>. Because of the small amount of <sup>3</sup>He found in nature it can not be gathered economically. However, enough <sup>3</sup>He for liquefaction purposes is obtained by an artificial nuclear reaction, namely:



Figure 1.1. Vapour pressure temperature curves for liquid <sup>3</sup>He and liquid <sup>4</sup>He.

As will be seen further on, liquid helium four does not solidify under its own vapour pressure; neither does liquid helium three, i.e. helium does not have a triple point gas-liquid-solid. Here we must mention again a very important difference between these two liquids. Below a certain temperature called the lambda point liquid helium four presents very peculiar features which make it a superfluid. As far as the experiments

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up to this date indicate liquid helium three does not.

One can obtain lower temperatures by means of other methods and devices, namely with a  ${}^{3}\text{He}{}^{-4}\text{He}$ dilution refrigerator<sup>(1)</sup> with adiabatic demagnetization  $\alpha$ ) a paramagnetic salt (electron cooling)<sup>(2,3)</sup> and  $\beta$ ) a nuclear spin system of, for instance, the Zte erratum certain metals or the protons of the crystalwater of certain salts (nuclear cooling)<sup>(4,5,6)</sup>. In table 1.1 the different cooling methods and the final temperatures are shown.

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Method	Final temperature (K)
Boiling point of liquid <sup>4</sup> He Boiling point of liquid <sup>3</sup> He Reducing vapour pressure of liquid <sup>4</sup> He	4,2 3,2 0,72
Reducing vapour pressure of liquid <sup>3</sup> He	0,35
<sup>3</sup> He- <sup>4</sup> He dilution refrigerator Electron spin cooling Nuclear spin cooling	0,01 10 <sup>-3</sup> 10 <sup>-6</sup>

## 1.2. The diagram of state of <sup>4</sup>He.

In figure 1.2 a diagram of state of <sup>4</sup>He below its critical point at 5,20 K is shown. The so called  $\lambda$ -line separates two regions which relate to two different phases of the liquid helium which have notable dissimilar characteristics as will be explained in the next paragraphs.



Figure 1.2. The diagram of state of <sup>4</sup>He at very low temperature.

In figure 1.3 the phase diagram of <sup>4</sup>He is extended to higher temperatures and pressures. As has already been stated in 1.1. the triple point gas-liquid-solid exists neither for <sup>4</sup>He nor for <sup>3</sup>He. Therefore, an external pressure is necessary in order to obtain solid helium. In this latter diagram the different regions of the solid phases of <sup>4</sup>He are shown. The distinct allotropic forms in the solid phase are:

(α) hexagonal close-packed structure phase(hcp) below 15 K;



Figure 1.3. The diagram of state of <sup>4</sup>He extended to high pressures and temperatures showing the three different allotropic form regions.

- (β) face centered cubic structure phase (fcc) above 15 K; and
- (y) body centered cubic structure phase (bcc), which takes a very small region between 1,45 and 1,78 K.

1.3. Some physical properties of liquid helium.

In this paragraph some physical properties of liquid helium will briefly be reviewed. The curves indicating the specific heat, density and viscosity versus temperature can be seen in figures 1.4, 1.5 and 1.6, respectively.



Figure 1.4. Specific heat of liquid helium as a function of the temperature. The  $\lambda$ -point was named after the shape of this curve.

It must be pointed out that all these curves show a discontinuity in the property indicated or in one of the derivatives at 2,17 K. This temperature is called the lambda temperature  $(\lambda$ -point) after the shape of the curve of the

#### specific heat of liquid helium.



Figure 1.5. Density of liquid helium.

However, some features about the viscosity must be emphasized because the measurements of this parameter brought consequences of capital importance for the knowledge of liquid helium at temperatures between 2K and 1K. In fact, when measuring the viscosity with an oscillating disc or a very narrow capillary tube, the values obtained were different: the dotted line and the solid line of figure 1.6, respectively. While the method with the oscillating disc gave values of normal behaviour of a normal liquid, the method using very narrow capillaries gave values which were not normal at all.



Figure 1.6. The viscosity of liquid helium.

## 1.4. The lambda transition.

When the temperature of liquid helium is lowered by pumping out the vapour over its free surface and when a pressure of 38 tor is reached, a peculiar and visible change takes place in the bulk of the liquid. In spite of the fact that the liquid is still rapidly evaporating, any vestige of the boiling disappears. The temperature which corresponds to this change is also 2,17 K. The two phases of the liquid helium, above and below the lambda point, are called liquid helium I and liquid helium II, respectively. The regions of these phases in the phase diagram are indicated in figure 1.2.

The above mentioned disappearance of the bubbles in boiling liquid helium is a consequence of the abnormally high heat conduction of He II at temperatures not too far below the lambda point. At a temperature of 1,8 K it is approximately  $10^3$  that of copper or silver at room temperature, while the thermal conductivity of Helium I is that of an ordinary liquid. Because of this high conduction any reasonable heat influx in the liquid distributes itself too quickly to allow the formation of bubbles; the liquid evaporates at the surface (and at the film; see § 1.5.).

Furthermore, the entropy presents anomalies; below the  $\lambda$ -point it drops rapidly with the temperature. But no discontinuity takes place at the lambda transition because this transition is not accompanied by a latent heat. This means that under equilibrium conditions helium I and helium II cannot co-exist. The compressibility and the thermal expansion are not normal, that is, they present discontinuities as well. These anomalies and discontinuities are typical characteristics of a second order transition. This means a transition which involves discontinuities in the second order derivatives of Gibb's function or thermodynamical potential G but not in the first order ones.

G = U - TS + pV

where U is the internal energy, T the absolute temperature, S the entropy, p the pressure and V the volume. Then,

 $\frac{1}{V} \cdot \left(\frac{\partial^2 G}{\partial T \cdot \partial p}\right) = \alpha \quad (\text{thermal expansion coefficient})$  $T \cdot \left(\frac{\partial^2 G}{\partial T^2}\right) = -C_p \quad (\text{specific heat})$ 

and

$$\frac{1}{V} \cdot \left(\frac{\partial^2 G}{\partial p^2}\right) = -K \quad (compressibility).$$

So that, the lambda transition is a second order one as it is the transition of a superconductor from the superconductive to the normal state in zero magnetic field.

Finally, it is interesting to make the following remark. In (diluted) mixtures of  ${}^{3}\text{He}$  in  ${}^{4}\text{He}$  the lambda point decreases with increasing concentration of  ${}^{3}\text{He}$ . As far as the experiments up till now indicate, pure liquid  ${}^{3}\text{He}$  does not show a lambda transition.

## 1.5. Superfluidity.

When Kamerlingh Onnes liquefied helium he failed to discover a property of this material as odd and as important as superconductivity. In fact, pumping out the vapour, a temperature below 0,8 was not possible. However big the improvements of the vacuum technology have been since, not much lower temperatures have been reached with liquid helium four using the same method. This is due to the fact that when liquid helium undergoes the  $\lambda$ -transition, see figure 1.2, the new phase of liquid helium, He II, becomes a superfluid and the thin film of liquid helium starts to creep up fast on the walls of the container, as was anticipated in 1.1. This is one of the consequences of the superfluid state which was discovered almost simultaneously by Kapitza<sup>(7)</sup> and by Allen and Misener<sup>(8)</sup>. The viscosity of liquid helium II has been measured with different methods which produced systematically different values, and hence different consequences, as was pointed out in paragraph 1.3.

Those authors observed that the flow of this liquid in long and narrow capillaries appeared to be non-classical; it was not proportional to the pressure difference between both ends of the capillary. For instance, in a tube of a diameter of about  $10^{-5}$  cm a "critical" velocity of approximately 13 cm/sec was obtained which was quite independent of this pressure difference. The existence of this critical velocity leads to an explanation in terms of a fluid having zero viscosity. Indeed, these two features, the zero viscosity and the independence of the pressure, are those that define a superfluid; it behaves in a way different from that of a classical fluid.

An immediate result of superfluidity is that when there is any kind of very small leak in the container or apparatus the liquid helium flows through it, also when it is very difficult to detect as a vacuum leak. This is called a lambda leak. Here it must be pointed out that a lambda leak must not be confused with a superleak. The former is an accident while the latter is made on purpose. Later on the superleaks will be discussed. A remark must be added. Superfluidity is not a property of liquid helium four only. Superfluidity is also found in a system of electrons in metals which at low temperatures can form pairs which behave as Bose particles. The macroscopic manifestation of this formation is superconductivity. Moreover, as recently has been suggested by Leggett <sup>(9)</sup>, certain insulating solids may be in a sufficiently similar quantum state to that of helium to show superfluidity. Leggett suggests solid helium as the most adequate solid to prove this hypothesis. But experiments have not yet been made.

Three fundamental experimental facts are a consequence of the existence of superfluid liquid helium, namely: the thermo-mechanical effect, the mechano-caloric effect and the helium film. If in an arrangement as the one of figure 1.7 a small amount of thermal energy reaches the upper part of the tube T producing a very small temperature difference, which can be as small as  $10^{-3}$  K, the superfluid starts to penetrate the tube T through the emery powder plug with very small pores. If the heat input is not too small an emergent jet of liquid helium is produced.

The thermo-mechanical effect is a thermodynamically reversible process. Figure 1.8 shows



Figure 1.7. Arrangement to produce the fountain or thermo-mechanical effect. Figure 1.8. Arrangement to produce the mechanocaloric effect.

how it is possible to produce the inverse effect or mechano-caloric effect. A thermometer indicates the temperature of the liquid He in the small Dewar D which has been filled by immersing it in liquid He II. When the Dewar is raised from the liquid helium bath and the superfluid falls through the small pores in the emery powder, the temperature of the rest of the liquid helium rises. Here it would be useful to note that the emery powder plugs act as a superleak in both cases. They are so mounted as to allow the superfluid to go through the

narrow channels on purpose.

The third fact is the following. As it was stated in 1.1 and the beginning of this paragraph, a thin film of liquid creeps up on any solid in contact with it. In figure 1.9 it is possible to see how a beaker can be filled or emptied, introducing an empty beaker in the liquid helium II or rising the filled beaker, respectively.





(b)



Figure 1.9. The liquid helium II film. The double beaker experiment indicated in figure 1.9 c demonstrates that the flow of the superfluid does not require an observable pressure gradient.

In Table 1.2 are given the thicknesses in Åu of the superfluid helium film at different heights above the free surface of the liquid. The determination of the thickness of liquid helium film was made by light interference and polarized light methods (10,11)

TABLE 1.2

Height above the liquid <sup>4</sup> He surface	Thickness (Åu)
(cm)	2,05 K 1,55 K
0,4	480 408
1,0	315 280   255 246

As will be explained later on, it is possible to introduce a definition, namely: A superfluid is a condensed degenerate gas obeying Bose-Einstein statistics in its lowest energy state. Consequently, superfluidity is of quantum mechanical origin because at very low temperatures a significant part of the atoms are in the lowest energy state permitted by quantum mechanics.

## 1.6. The two fluid model for liquid helium II.

In order to understand the startling properties of liquid helium II an abstract model was conceived by Tisza<sup>(12)</sup> in 1938. With this model, sometimes called theory, it is possible to explain phenomenologically most of the facts related to the behaviour of this liquid. Nevertheless, some authors insist on remarking that the two fluid image of liquid helium two is only a model which helps to explain the facts and that it is not a physical theory. a) liquid helium II consists of two mutually interpenetrating fluids (components), the "superfluid" of density  $\rho_s$  and the "normal" fluid of density  $\rho_n$ , so that at any point in space the density  $\rho$  can be written as

 $\rho = \rho_s + \rho_n$ .

Figure 1.10 represents the densities of the two fluids based on measurements of Andronikasbvili <sup>(13)</sup>.





- b) The <u>superfluid</u> corresponds to the condensed phase of a Bose-Einstein degenerate gas in its lowest energy state. But phase is not in this case a phase in the sense of the Gibbs' phase rule, because the two liquids do not separate in two phases.
- c) The <u>normal fluid</u> is supposed to be the carrier of all thermal excitations of the liquid. Therefore, the normal fluid is the one which possesses the total entropy.

With the above described hypotheses the observed phenomenon that at a given temperature the viscosity has two different values as we saw in 1.3 and 1.5, can be explained: the oscillating disc gave account of the viscosity of the normal fluid and the narrow capillary method of the interaction of the superfluid component with the wall of the capillary and with the normal component. The fluid which goes through the pores of the emery powder in the thermo-mechanical and mechano-caloric effect and, finally, the fluid which creeps up quickly on any solid surface in contact with the liquid (see figures 1.7 , 1.8 and 1.9 ) is the superfluid component of the liquid helium II.

Moreover, the small amount of the light isotope <sup>3</sup>He present is not carried with the superfluid component. This is so not only because liquid <sup>3</sup>He does not show superfluidity but also because <sup>3</sup>He atoms are fermions obeying Fermi-Dirac statistics while <sup>4</sup>He atoms are bosons following Bose-Einstein statistics.

# 1.7. Propagation of thermal and mechanical perturbations in liquid helium II.

The propagation of sound in liquid helium II has been studied at length and is called first sound. Another longitudinal wave motion appears in this fluid and was called second sound. The first sound in liquid He II is ordinary sound and the second sound is a thermal wave originated when the concentration of normal and superfluid fluid changes at a given point because of an increase or diminishment of the temperature at that point. These thermal waves, which have been predicted by Tisza and Landau<sup>(14,15)</sup> were first observed by Peshkov<sup>(16)</sup>, using a "hot" wire carrying an alternating current as a source and resistance thermometers as detectors. Above 1,2 K there is very good agreement between the measured values of the second sound velocity u and Landau's expression

$$u_2^2 = \frac{\rho_s}{\rho_n} \frac{TS^2}{C} \quad .$$

Here the densities  $\rho_s$  and  $\rho_n$  are determined from Andronikashvili's (1.6) experiment while S and C are the entropy and the specific heat respectively. However, below 1,5 K the values of  $\rho_s$  and  $\rho_n$  from Andronikashvili's experiment are not accurate enough and then  $\rho_n$  is calculated from the observed velocities of second sound.

In figure 1.11 the speed of propagation  $u_1$ of the first sound and  $u_2$  of the second sound as a function of temperature are shown.





Furthermore, another oscillators motion has been predicted by Atkins<sup>(17)</sup> and observed in helium films. This is called third sound and is an oscillation which occurs in the superfluid component while the normal component remains locked to the wall. This wave exists only in a superfluid since a classical wave would be immediately damped in

a thin film by the viscosity. The waves were observed as follows. A narrow strip of a He II film was irradiated with periodic pulses of infrared light. This caused a periodic heating (and evaporation). The periodic variation of the film thickness was measured in another region of the film. This effect is the propagation of a thermal wave in a thin film while the second sound is the same kind of propagation which occurs in the bulk of the He II.

Atkins<sup>(17)</sup> also predicted a fourth sound which is a combination of first and second sound. It exists only in narrow channels. In these channels the normal component is locked to the walls. Therefore, the density fluctuations can originate from motion of the superfluid only. With the fourth sound the wave length of the excitations becomes comparable to or greater than the diameter of the small channel.

These fourth sound oscillations produce density fluctuations together with the temperature fluctuations of second sound. Therefore, as Khalatnikof (1965) says: in this case it is possible to hear sound propagation in the superfluid component.

Finally, the so called zero sound should be mentioned as well. This new mode of sound

propagation was predicted by Landau<sup>(18)</sup>. The name zero sound is due to the fact that it is a phenomenon which occurs only at very low temperatures. It is the normal mode of propagation of ordinary sound in a system obeying Fermi-Dirac statistics. Therefore it can occur in liquid <sup>3</sup>He or in another Fermi liquid.

## 1.8. Zero point energy.

As has been said previously, below the lambda point liquid helium exhibits the property of superfluidity and also one is dealing with a quantum liquid. It was pointed out that the appearance of superfluidity is accompanied by a very rapid drop in the entropy which is null at zero temperature. Moreover, no discontinuity in the energy takes place when the lambda point is passed.

As is well known, in classical physics the motion of heat as a form of motion relates the absolute temperature of the substance to the kinetic energy of the particles (atoms and molecules). So that, if v is the average velocity of the particles, one can write

 $T \stackrel{\sim}{=} v^2$ .

Thus, when T = 0, v = 0; therefore, at zero temperature everything is at rest.

However, experiments carried out at low temperatures show results which classical theory cannot explain. That is, experiments in which the particles of the substance evidently remain in movement at the absolute zero of temperature (i.e., at temperatures as close to it as the technology can achieve).

Quantum mechanics explains very clearly why movement must remain. In fact, considering Heisenberg's uncertainty relation

 $\triangle q$  .  $\triangle p > h$ 

where the  $\Delta q$  is the uncertainty in the position of a particle,  $\Delta p$  the simultaneous uncertainty in its momentum and h is Planck's constant. If the velocity of a particle is fixed at any value, p is fixed. So is  $\Delta p = 0$  and  $\Delta q$  becomes infinite. This means that a particle of a fixed and definite velocity including zero, cannot be located at all. Nevertheless, experiment shows that it remains located also at absolute zero. Therefore, it must have a velocity as well.

When we consider an oscillatory system of, for instance, phonons, the energy of each mode is

 $E = (n + \frac{1}{2}) h v$ 

where n is a positive integer (including zero) and v is the frequency of the oscillation. This expression indicates that the lowest energy obtainable is not zero but

$$\frac{h v}{2}$$

This energy cannot be removed, not even by lowering the temperature to the absolute zero. At very low temperatures  $\frac{1}{2}$  hv is a significant part of the total energy left but at higher temperatures is negligible.

From another point of view this zero point energy is so high that interatomic forces are too weak to bind the helium atoms into the crystalline state. That is why helium does not solidify under its own vapour pressure. Furthermore there is the fact that at temperatures of the order of 1 or 2 K the de Broglie wavelength of helium atoms is comparable to the interatomic distances.

# 1.9. <u>The microscopic theory of liquid helium: Landau and</u> <u>Feynman.</u>

However succesful the two fluid model based on the Bose-Einstein condensation was in explaining the features which had already been discovered in liquid helium II, Landau<sup>(15)</sup>(translated into english in Khalatnikov book, 1965) thought that liquid helium II had to be treated as an entity rather than as divided into two fluids. This new point of view considers the liquid helium to be a continuum, or more precisely a quasicontinuum, filled with two types of normal modes namely longitudinal modes and rotational modes. The quanta of these two excitations are the phonon and the roton, respectively. A similar model is, for example, a precinct or enclosure filled only with electromagnetic radiation which is considered as a continuum filled with photons. That is to say, that Landau regarded liquid helium II as a kind of background for an assembly of phonons and rotons which should be treated in the same way as photons, namely as particles interacting with the surroundings. These excitations provide liquid helium II with normal properties, forming the normal component, to use the terminology of the two fluid model.

> The energy of a phonon is given by  $E = \frac{h v_s}{\lambda} = p v_s$

where h is Planck's constant,  $\lambda$  and p are the wavelength and momentum, respectively, of the phonon and v is the velocity of sound. On the other hand, the energy of a roton is given by the expression

$$E_{rot} = \frac{(p-p_o)^2}{2\mu} + \Delta$$

where  $p_0$  is a constant or the value of the momentum at which the energy of the roton has a minimum equal to the constant  $\Delta$ ; the quantity  $\mu$  has the dimensions of mass and is called the effective mass of the roton. In figure 1.12, the energy spectrum of liquid helium II is represented according to Landau's theory. Landau determined



Figure 1.12. The energy spectrum of liquid helium II. the values of the parameters  $\Delta$ , p<sub>o</sub> and  $\mu$  by adapting experimental caloric data and obtained the following, values :

$$\Delta/k = 9 \text{ K}; \text{ p}_0/\hbar = 2 \text{ Å}^{-1}; \mu = 0,2 \text{ m}_{\text{He}}$$

where k is Boltzmann's constant,  $\hbar = h/2\pi$  and  $m_{He}$ the mass of one helium atom. In experiments suggested by Cohen and Feynman in 1957 the curve of the energy spectrum was measured by means of inelastic neutron scattering. Monochromatic neutrons emit or absorb elementary excitations in liquid helium, which energy was measured and the whole spectrum was determined. In this experimental way the parameters obtained are:
$$\Delta/k = (8,65 \pm 0,04)K$$
;  $p_0/\hbar = (1,92 \pm 0,01)A^{-1}$ ;  
 $\mu = (0,16 \pm 0,01)m_{He}$ 

which are in good agreement with those obtained by Landau. Moreover, when the effective density of the rotons plus the phonons is equal to the density of the liquid the  $\lambda$ -point appears. Landau estimated this would take place at 2,3 K, a vlaue which is very near to the observed value of 2,17 K. These results confirmed Landau's equation for the energy of the rotons and, consequently, his hypotheses.

An important contribution to the theory of liquid helium was given by Feynman in 1953 - 4 (see Gorter, volume I, 1955) considering the form of the wave function in an assembly of bosons. He obtained an energy spectrum similar to that given by Landau. In this way both series, the one based on Bose-Einstein statistics leading to the two fluid theory and Landau's theory based on the quanta of excitations or quasiparticles are unified.

As a consequence of the value of  $\Delta \approx 9$  k in the expression of  $E_{rot}$  the density of the rotons must decrease exponentially with decreasing temperature below about 0,6 K.

According to Mendelssohn<sup>(19)</sup>, Landau himself, when asked about the bosons and the lambda transi-

tion, said his opinion was that "bosons are required to produce superfluidity but that he was equally convinced that the lambda point of helium is not a feature of Bose-Einstein condensation."

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### CHAPTER II

## SUPERLEAKS

## 2.1. Introduction.

In paragraph 1.5 the fountain effect or thermomechanical effect <sup>(1)</sup> was described as an experimental fact which occurs in liquid helium II. Also the superleaks have been mentioned and defined in the same paragraph as a "voluntary lambda leak". A superleak could be used with different purposes, for example to produce the fountain effect in order to fill an experimental cell with liquid helium II or to empty it. This effect can be explained qualitatively making use of the two fluid model of liquid helium II or quantitatively by means of the thermodynamics of the effect.

# 2.2. The fountain effect and the two fluid model.

With the arrangement of figure 2.1 it is possible to produce the thermo-mechanical effect. The two containers A and B are filled to the same level with liquid helium II and connected by a fine capillary C. An amount of heat Q is supplied to one of them, for instance to B, where the temperature rises by  $\Delta T$ . Then the liquid flows in the direction of the highest temperature producing a difference of pressure  $\Delta P$ . This is an experimental effect which can be explained by means of the two fluid model. In effect, at any temperature between



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Figure 2.1. The fluid model explanation for the thermo-mechanical effect.

2,17 and 0 K the behaviour of liquid helium II is determined by either the relation  $\rho_n/\rho$  or  $\rho_s/\rho$ . When the temperature rises, superfluid is excited into the normal state and the concentration of the normal component increases, see figure 1.10, which is compensated by the flow of more superfluid through the superleak C in the direction of the higher temperature. Moreover, the normal component in B cannot counterflow through the superleak into A. Hence the capillary acts as a form of semi-permeable membrane.

# 2.3. Thermodynamics of the thermo-mechanical effect.

It was said (1.5) that the passage of liquid helium II through the porous plugs in the mechanocaloric and the thermo-mechanical effects is a reversible process. Therefore, both cases can be treated thermodynamically <sup>(2)</sup>. This treatment leads to a quantative picture of the fountain effect. If the volumes of the two containers A and B of figure 2.2 are constant, if they are joined by means of a superleak, and filled with liquid helium II and moreover thermally isolated from each other and from the outside, the equilibrium condition is given by the minimum of the total energy.



Figure 2.2. Constant volumen containers joined by a superleak.

That is,

 $\delta U = \sum_{k=A,B} \delta(M_k \cdot u_k) = 0 = \sum_{k=A,B} (u_k \cdot \delta M_k \cdot M_k \cdot \delta u_k)$ 

and since

$$u = u(s,v),$$
  

$$\delta u_{k} = \left(\frac{\partial u_{k}}{\partial s_{k}}\right) v_{k} \cdot \delta s_{k} + \left(\frac{\partial u_{k}}{\partial v_{k}}\right) s_{k} \cdot \delta v_{k},$$

so that

$$\sum_{k=A,B} \left\{ u_k \delta M_k + M_k \left[ \left( \frac{\partial u_k}{\partial s_k} \right) v_k \cdot \delta s_k + \left( \frac{\partial u_k}{\partial v_k} \right) s_k \cdot \delta v_k \right] \right\} = 0$$

where  $u_k$ ,  $s_k$  and  $v_k$  are the specific internal energies, entropies and volumes, respectively and M the mass of the liquid.

For each container S = Ms and V = Mv are constants, therefore

 $\delta S_{k} = M_{k} \delta s_{k} + s_{k} \delta M_{k} = 0$  $\delta V_{k} = M_{k} \delta v_{k} + v_{k} \delta M_{k} = 0.$ 

Bearing in mind that  $\left(\frac{\partial u}{\partial s}\right)_{v} = T$  and  $\left(\frac{\partial u}{\partial v}\right)_{s} = -p$ and combining these equations one gets

$$\sum_{A,B} (u_k - T_k s_k + p_k v_k) \delta M_k = 0$$

where the quantity within the parenthesis is the specific thermodynamical potential or specific Gibbs' function  $g_{\mu}$ . If we put  $\delta M_{A} = -\delta M_{B} = \delta M$ , then

 $(g_{A} - g_{B}) \delta M = 0$  $\sum_{a} e_{a} (P_{A}, T_{A}) = g(P_{B}, T_{B}) + g.$ 

Therefore, for small  $\Delta T = T_A - T_B$  and  $\Delta P = P_A - P_B$ 

$$\left(\frac{\partial g}{\partial T}\right)_{P} \Delta T + \left(\frac{\partial g}{\partial P}\right)_{T} \Delta P = 0.$$

As  $\left(\frac{\partial g}{\partial P}\right)_T = \frac{1}{\rho}$  where  $\rho$  is the density and  $\left(\frac{\partial g}{\partial T}\right)_P = -s$ , this formula is equivalent to

 $\Delta P = \rho s \Delta T$ .

This equation, called London's formula, has been checked experimentally and found to be consistent. Naturally, the measure of the fountain effect,  $\Delta P/\Delta T$ , is maximal when the entropy of the liquid helium II is greatest, which occurs just below the lambda point. The lower the temperature the smaller the thermo-mechanical effect because the proportion of superfluid with zero entropy increases, Figure 1.10.

## 2.4. The onset point of a superleak.

Since the experiments performed by Atkins et al <sup>(3)</sup> it is known that in the thermo-mechanical effect the superfluid starts to flow in a narrow channel of a superleak not as soon as the lambda point is passed but at lower temperatures. This is a phenomenon very similar to the one obtained with the unsaturated helium films in which the onset of the flow is below the normal lambda point as well and which was studied some years before by Bowers et al <sup>(4)</sup> and by Long and Meyer <sup>(5,6)</sup>. These temperatures are called the "onset temperatures" and the fact that they are below the normal lambda point is called "depression of the onset of superflow". There is a theoretical relation, which is a solution of the

Ginzburg-Pitaevskii <sup>(12)</sup> equation for helium, between the depression of the transition temperature and the width 2 d (in cm) of a parallel-sided channel

$$\Delta T \simeq \frac{2 \times 10^{-14}}{d^2} (K).$$

The smaller the pores the lower the onset temperatures. The channel connections which run between both sides of a superleak are not uniform and are at random, interconnecting each other. If we call  $\phi_{\min}$  the minimum diameter along a connection path, then the largest  $\phi_{\min}$  of all possible paths determines the onset temperature; because in that one the onset of the movement of the superfluid will take place first, i.e., at the highest temperature below the lambda point.

# 2.5. Determination of the onset point.

Atkins et al.<sup>(3)</sup> defined the onset temperature of a superleak as the temperature at which the level of the liquid helium inside a cell moved at about 0,01 cm sec<sup>-1</sup>. This method depends upon the optical perception of the movement of a liquid column and upon the measurement of times. It seems that the exactness of a method independent of human observation would be better. Nonetheless, all methods have an inherent uncertainty due to the fact that, as Hammel and Schuch <sup>(7)</sup> pointed out, there is a flow of saturated vapour of <sup>4</sup>He and a capillary condensation above as well as below the lambda point.

Considering a superleak as a device used in low temperature technology to fill or to empty a cell inside a liquid helium bath, the method<sup>(8)</sup> described in 2.8, figure 1, seems to be accurate enough since the resistor R indicates automatically the presence of liquid helium II inside the cell as soon as it appears. This apparatus can also be used with Atkins' method since it is provided with a calibrated capillary tube in the upper part. It would be possible to improve the accuracy of this method by painting a resistor of colloidal carbon or by evaporating a metallic superconductor resistor on the inner surface of the superleak. The superconductor material could be an alloy with a transition temperature as near as possible to the  $\lambda$ -point. The transition temperature from the normal to the superconducting state can be adjusted by an external magnetic field.

# 2.6. Applications of superleaks.

a) As a filter.

To fill a cell with very pure liquid helium free of macroscopical impurities. For instance, for the experimental study of the electrical breakdown of liquid helium in the way indicated in paragraph 3.3, figure 1.

 b) As an isotope separator. This is obvious because, as was said in paragraph 1.6,the light isotope <sup>3</sup>He is not carried by the superfluid component.

- c) <u>As a separator in the <sup>3</sup>He-<sup>4</sup>He dilution</u> <u>refrigerator</u>. Strictly speaking, this application proposed by H. London<sup>(9)</sup>, is the same as the former but used in the construction of an <sup>3</sup>He-<sup>4</sup>He dilution refrigerator in order to separate the liquid <sup>3</sup>He from the liquid superfluid
- d) <u>In a vortex refrigerator</u>.

Staas and Severijns<sup>(10)</sup> have designed a refrigerator based on the interaction between the superfluid and the normal fluid flow through a capillary; the superleak is used to "separate" the normal from the superfluid liquid.

e) <u>In a servo-valve for liquid helium II. The</u> <u>Cocatron.</u>

This one is a simultaneous application of both superfluidity and superconductivity <sup>(11)</sup>. In effect, one might need it to keep a constant liquid helium II level in a cell C when the cell is immersed in a liquid helium bath as in the case described in paragraph 3.3, figure 1. For that purpose a device, figure 2.3, can be made by means of a "superconductor resistor" R at the liquid level and a heater H acting on the external face of a superleak S. When the resistor at the liquid level becomes a superconductor, a large current runs through the circuit, and the temperature

48

<sup>4</sup>He.



Figure 2.3. Outline of a servo-valve for liquid helium II. The Cocatron.

of the superleak is raised above the onset temperature. Thus the flow of liquid helium II through the superleak stops. When the liquid He II in the cell evaporates, the level falls below that of the resistor. This becomes normal, the current drops to a low value and the heater nearly stops warming the superleak. Therefore, the liquid helium II starts again to fill the cell, and so on.

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## 2.7. A SILICON CARBIDE SUPERLEAK

J.M. Goldschvartz and B.S. Blaisse.

Nowadays, the physics of very low temperatures has already arrived at the stage of technological applications. In view of the fact that superconductivity provided an enormous advance in some techniques, doubtless superfluidity will as well. Therefore, devices and materials must be available for applications of this fundamental phenomenon. For instance, in some of our experiments in which the use of very pure liquid helium was required, the best way to obtain it was through a superleak. For that purpose we used Vycor glass.

At present, the most useful superleaks are: a tube filled with very fine jeweller's rouge made by Bots and Gorter <sup>(1)</sup> and the above-mentioned Vycor glass <sup>(2)</sup> developped by Norberg <sup>(3)</sup> and first used as a superleak by Atkins, Seki and Condon <sup>(4,5)</sup> and Champeney <sup>(6)</sup>. But however ingenious those superleaks are, they show various disadvantages: jeweller's rouge must be renewed each time it is used and the Vycor glass is very difficult to machine, breaks easily due to its brittleness, lacks mechanical strength, often breaks when a vacuum is made in the cryostat, probably due to the freezing of the occluded water vapour and to the sudden release of a large amount of occluded gases and cannot be easily sealed to other glasses.

In our search for another material to be used instead as a reliable and fine superleak, we found that a certain kind of a dense form of reaction sintered silicon carbide<sup>(7)</sup> did behave as the desired superleak. This form of silicon carbide, as well as others obtained by following different techniques, made by Zee erratum sublimation or by pyrolisis of silicon and carbon containing gaseous compounds <sup>(8)</sup>, can be produced in almost any desired porosity. Sintered silicon carbide porous plugs have already been used by Christian and Leck <sup>(9)</sup> as an element for the controlling and measuring the gas flow into a vacuum system.

> This paper is a preliminary and rather qualitative report of our first results and is concerned only with that particular case of a dense form of reaction sintered silicon carbide obtained by sintering technicalgrade silicon carbide grains smaller than  $10\mu$ , impregnated by a carbonaceous substance, e.g. colloidal graphite, and heated in SiO vapour for several hours at 2.000 <sup>o</sup>C in a hydrogen atmosphere. A definite porosity can be obtained by regulating the temperature, the reaction time and the repetition of the impregnation. Moreover, some conspicuous technical features make the

silicon carbide particularly suitable for our purpose, namely: it is machinable, very strong, chemically inert, thermically stable, beakable, can be directly sealed to Pyrex glass and can be cleaned and degassed at high temperatures. The samples used for our experiments are porous plugs of approximately 8-10 mm diameter and 10 mm height, sealed in a Pyrex glass tube, vacuum tested and with a porosity of about 3% and 8%.

We had to probe this material to see if it was capable of producing the fountain effect and, particularly as a superleak, the "dropping" of liquid helium II through the pores under definite conditions. In other words; we had to detect the shifting of the  $\lambda$ -transition to a lower temperature.

For this purpose we used the simple glass devices of Figs.la and lb. In Fig. 1a the silicon carbide sample B sealed to the Pyrex glass tube t was immersed in liquid helium below the  $\lambda$ -point. The temperature difference between the cell and the bath, despite their good thermal contact, was regulated mainly by the pumping systems but also with the help of the small 0,24 - W electric lamp L approximately 5 mm in diameter, 20 mm long. This lamp can supply either heat pulses or can heat steadily.

Of course, the contribution of the heat leak along the tube t is not negligible since the cell



Figure 1. Simple devices used for testing the sintered SiC as a superleak.

constitutes an almost independent system inside the cryostat, i.e., it has its own manometer and can be filled with liquid helium and pumped out without interfering with the cryostat system. This simple device could be used for both the fountain effect and the dropping but we used it particularly to control the dropping.

Once the Dewar was filled with liquid helium and tested during a reasonable time to see if the tested material showed any leak to liquid helium I, the cell t was then filled with liquid helium. When the cell was filled by means of the fountain effect to a sufficiently high level, we then reduced the level of the liquid helium in the Dewar to a position below the bottom of the superleak. Hereafter the pressure in the Dewar was allowed to rise and, after a time, the dropping began. When the cell t was filled by means of a syphon, then we pumped out some helium in the Dewar in order to obtain a level below the bottom of the superleak and then we pumped out the inside of the cell in order to reach the  $\lambda$ -point and to obtain, after a time, the dropping from the superleak. In both cases it was also possible to raise the cell in the Dewar over the level of the liquid in it. In any case the successful samples did keep the liquid helium II inside the cell till the onset point was reached and the dropping began. This dropping could be regulated as desired: started, fastened and stopped.

On the other hand, the device shown in Fig. 1b was used particularly to control the filling of the cell by the fountain effect, due to the fact that the vacuum chamber a, evacuated to  $10^{-6}$  torr, made possible a better thermal insulation and consequently a better control on the inside pressure. In Fig. 2 different stages of liquid helium II drops can be seen. These photographs were taken with the sample B8 at a

shutter speed of  $10^{-3}$  sec and the direction of the movement of the curtain shutter was parallel and opposite to the direction and movement of the drop.



Figure 2. Photographs of liquid helium II drops at different stages.

TAT	DT	F	т
TH	TD.	L.	1

	Mean value of onset dropping,	Mean value of Dewar temperature,	Poro- sity,	Porous dia- meter,
Sample	°K	°K	%	Å
B1	1,62	1,75	3	< 50
B8	1,79	1,91	8	< 70
Porous Vycor glass	1,64 [4] 2,0 - 2,1 [10]	-	28	50 70
Jeweller rouge	's 1,8[1]	-	-	100

The values shown in Table I were obtained by means of the procedures described above. These values are consistent with those of the porosities. The values for Vycor glass and jeweller's rouge are included as reference material. As far as the diameters of the pores are concerned we are not yet able to say anything, except that they must be smaller than 50 Å for sample B1 and smaller than 70 Å for sample B8, as compared with the values given by Atkins et al. (4) and by Brewer, Champeney and Mendelssohn (10).

However, these results must be taken only as being provisional. Just now we are engaged on more precise experiments with this and other forms of silicon carbide, applying more sensitive methods to determine the onset point. Nevertheless it is well to remember, that these sintered silicon carbide samples behaved like very good superleaks and that, at the same time, they offer all the above mentioned technical advantages.

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2.8. ONSET POINT OF A NEW NATURAL SUPERLEAK

J.M. Goldschvartz, E. Martin and B.S. Blaisse.

In a previous paper <sup>(1)</sup> we described a silicon carbide superleak which has some technical advantages over those hitherto in use. Nevertheless, we continued our investigations to discover new superleaks which are even more convenient. This paper reports the results obtained with a natural material called 'wonderstone' <sup>(2)</sup> which is a very fine-grained sedimentary rock of volcanic origin found in South Africa, in the Western Transvaal. Apart from the electrical, electronic, and chemical applications of wonderstone, it is also used, after undergoing different thermal treatments, to make cavities for electron spin resonance spectrometers <sup>(3)</sup>.

Wonderstone possesses some conspicuous characteristics. In the first place, after heat treatment its dimensions are enlarged. In Table I the features of this material can be seen before and after thermal processing. In the second place, before heating it, wonderstone can be cut very easily, it can be drilled, a thread can be cut in it, it can be turned on a lathe, and polished. Because it can be shaped as desired this material could be very suitable as a superleak

Sample	Orig dime	ginal ensions	Fina dime	l nsions	Var	iation	Total time of thermal	Maximum tempe-	Final colour
	φ, mm	Thickness, mm	φ, mm	Thickness,	φ, %	Thickness %	process, h	rature, C	
Natural	35	5	35	5	-	_	-	_	grey
Half-hard	35	5	35,9	-	2,5	-	24	750	grey- pink
Hard	35	5	36,2	5,1	3,4	2	72	1200	pink

TABLE I



Figure 1. Bottom of the cell used for the determination of the onset point of the wonderstone superleaks.

in many applications.

The device shown in Figure 1 is the bottom of the cell in which the onset points were measured, and which is suspended inside the helium dewar. The small Allen-Bradley resistor R, (type BB,  $\frac{1}{8}$  W, 56  $\Omega$  at room temperature) lies on the internal face of the superleak S. Both, the superleak and the terminals T through the glass are closed with indium wire in the conventional way. The procedure of the experiment was the following. The dewar was filled with the liquid helium at boiling point, while the cell was kept evacuated. Then the temperature of the bath was decreased and the onset point was reached, at which the helium passed the superleak and reached the resistor R. The resistance R experienced a sudden change which was recorded. This discontinuity is due to the discontinuous change in the heat conductivity of the liquid helium when it reaches the  $\lambda$ -point and because the superfluid penetrates into the resistor. By this procedure and with the device used, the onset points and also the  $\lambda$ -point could be determined very accurately.

In figure 2 the calibration of the resistor R in the region below the  $\lambda$ -point is given. The power dissipated by the resistor was of the order of  $10^{-3}$  W.

In Table 2 the mean values of the onset points of these superleaks are shown. The third sample, the so-called 'hard', was leaking He I, which means that the pores were enlarged too much. However, once the helium was below the  $\lambda$ -point, this sample was also sensitive enough to the action of radiation to produce the fountain effect, that is, to fill the cell shown in figure 1 and to keep the He II inside the cell.



Figure 2. Calibration of the Allen-Bradley resistor type BB in the temperature regions of the onset points of the wonderstone superleaks.

	IADLE 2
Sample	Onset point K
Natural	2,05
Half-hard	2,12
Hard	-

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## 2.9. NEW SUPERLEAKS

J.M. Goldschvartz, E. Martin and B.S. Blaisse.

As is well known, a very fine jeweller's rouge <sup>(1)</sup> and porous Vycor glass <sup>(2)</sup> are used as devices through which the superfluid component of liquid helium-II can flow. The temperature at which this flow starts depends upon the diameter of the pores; the smaller they are the lower that temperature is. This is called the onset temperature. For the two superleaks mentioned above, these onset temperatures were 1,8 and 1,64 K respectively. Nevertheless, for the porous Vycor glass it was later found that the onset temperature <sup>(3)</sup> lies between 2,0 and 2,1 K.

In 1968 we found <sup>(4)</sup> that silicon carbide plugs sealed to Pyrex glass tubes were also good superleaks with an onset point of 1,62 and 1,79 K for porosity percentages of 3 and 8 respectively. Moreover, if these SiC plugs are available, in some aspects they are rather preferable as superleaks to those previously mentioned.

However, we continued our search for new materials to be used as superleaks. Particularly,

we looked for a natural one. In fact, we found that a mineral called wonderstone <sup>(5)</sup>, an aluminium silicate from South Africa, with a porosity between 5 and 5,6% works as a good superleak. The chemical composition of wonderstone is:

Silica	57,19%	Titanium oxide	2,08%
Aluminium	32,78%	Lime	0,40%
Ferric oxide	0,72%	Magnesia	0,36%

Verualing : 6,47%

This mineral wonderstone possesses a conspicuous feature: on heat-treatment its dimensions are permanently enlarged depending on the time and temperature.

For the measurements with the wonderstonde we used three kinds of samples, namely: a natural one, one heated to 750  $^{\circ}$ C for 24 hr, which we call 'half-hard', and a third one which was heated to 1200  $^{\circ}$ C for 72 hr, and which we call 'hard'. The pores of the 'hard' one were so large that the sample was leaking helium-I. The onset points of the natural and the half-hard samples were 2,05 and 2,12 K, respectively.

On continuing our search for more natural materials to be used as superleaks, we found another mineral (6), from Rabenwald, Austria - a talc-stone (7), which is a magnesium silicate, and works as a very good one. Its chemical com-

## position is:

Silica	51,6%	Aluminia	8,8%
Magnesia	30,9%	Ferric oxide	1,0%
Water	4,7%	Calcium oxide	0,3%
Carbon dioxide	2,7%		

In the Table the onset temperatures of these two new minerals able to work as superleaks are shown.

Both the natural wonderstone and the natural talc-stone have a crystalline structure and a greasy touch. Both are soft and can be easily machined and shaped as desired.

All the samples used for the measurements were discs of 40 mm diameter and 5 to 10 mm thickness and were cut at random from natural blocks.

A disc was attached to a Pyrex glass cell by means of aluminium flanges and the vacuum and lambda tightness obtained with an indium wire seal.

In order to detect the onset points we used carbon resistor thermometers for the wonderstone and visual detection of the variation of the liquid helium level inside the cell for the talc-stone. This cell was suspended in the liquid helium and a moderate vacuum was made inside. In both cases we have made use of the thermo-mechanical effect to fill the cell through the superleak.

Material	Onset temperature, K	Diameter of the pores, Å
Wonderstone: natural half-hard hard	2,05 2,12 	> 70  
Talc-stone: natural	1,69	< 70

TABLE

We think that the new materials described above would be able to help to symplify some problems in the technological applications of superfluid helium, particularly in the construction of refrigerators to obtain temperatures well below 1 K.

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## 2.10. NEW MATERIALS FOR SUPERLEAKS

J.M. Goldschvartz and B.S. Blaisse.

### SUMMARY

Last year we found that the African mineral wonderstone, an aluminium silicate, is a good material for the construction of superleaks. In search for other materials we found that natural talc-stone from Rabenwald, Austria, behaves as a good superleak as well. The onset point, that is the temperature at which the superfluid component starts to flow through the pores, is 1,69 K in the sample used. This means that the largest pores have a diameter less than 70 Å.

Two years ago we found and described <sup>(1)</sup> a new kind of superleaks made of silicon carbide. These superleaks present some advantages over the ones conventionally used until then, i.e., the very fine jeweller's rouge pressed into stainless steel capillaries <sup>(2)</sup> and the porous Vycor glass <sup>(3)</sup>. For these SiC superleaks we found that the onset points for two different samples were 1,62 K and 1,79 K for a porosity
percentage of 3 and 8 respectively. The most remarkable features of these superleaks are that it is possible to seal them to a Pyrex glass tube and that the porosity percentage can be fixed "a priori".

Recently we have reported the results (4,5) obtained with a natural material of South African origin, used as a superleak. This material is an aluminium silicate named wonderstone and has the following components percentages <sup>(6)</sup>: Silica 57,19; Alumina 32,78; Ferric Oxide 0,72; Titanium Oxide 2,08; Lime 0,40 and Magnesia 0,36. We have checked three different samples of this material, namely: a natural one, a "half-hard" and a "hard" one. The half-hard and the hard were heat treated at temperatures of 750 C and 1200 C respectively. A consequence of this heat treatment is a permanent increase of the dimensions, enlarging the diameters of the pores. The onset points of the natural and half-hard samples were 2,05 K and 2,12 K respectively. The pores of the hard sample were so large that it was leaking He I.

Finally, we must remark that the natural wonderstone can be easily machined, shaped as desired and has a crystalline structure. All the samples used for our measurements were cut at random from a block.

This paper deals with the description of a new natural material  $^{(5)}$  which is a good superleak as well. It is talc-stone, a magnesium silicate from Rabenwald in Austria. Its chemical composition percentage  $^{(7)}$  is: SiO  $_2$  - 51,6; MgO - 30,9; H O -  $_2^2$  4,7; CO  $_2$  - 2,7; Al O  $_2^-$  8,8; FeO - 1,0 and CaO - 0,3.

It has a crystalline structure, its colour is a light-grey, almost white, it has a notable greasy touch and it is found in blocks.

This talc-stone can also be machined easily and shaped as desired. However it is not as strong as wonderstone. The sample used for our experiment to determine the onset temperature, was a disc of 40 mm diameter and 10 mm thickness. It was also cut at random from a natural block. The disc was fixed to a Pyrex glass cell by means of aluminium flanges; the lambda tightness was obtained with indium wire covered with some silicon vacuum grease <sup>(8)</sup>.

For this talc-stone superleak we have found an onset point of 1,69 K. This temperature is determined by the largest diameter of the channels in parallel which is the smallest diameter of this largest channel considered as channels in series. The onset temperature of 1,69 K means that the diameters of those pores are between 50 and 70 Å. In the table we can see the features of the superleaks described.

The method used to determine the onset point in the case of wonderstone was already described in the paper mentioned above <sup>(4)</sup>, in which we used two calibrate Allen Bradley carbon resistance thermometers. One was placed below the superleak in the liquid helium and the second one inside the cell and lying on the internal surface of the superleak. When the superfluid component penetrates the cell due mainly to the thermomechanical effect and reaches the internal carbon thermometer, its resistance undergoes a sudden change. This is recorded and at the same time the temperature of the helium is read.

For the case of the talc-stone superleak we made use of the thermo-mechanical effect as well but with visual control of the movements of the helium level inside the cell and then reading the temperature.

We think that the application of these natural materials for the fabrication of superleaks could eventually implicate a significant advance in the construction of new technological devices for the obtainment of temperatures well below 1 K.

Туре	Onset Tempe- rature (K)	Pores dia- meter (Å)	Porosity %
Jeweller's Rouge	1,8	100	
Porous Vycor glass	1,64 2.0 - 2.1	50	28
Sic	1 79	> 70	8
SiC	1,62	< 50	3
Wonderstone Half-hard Natural	2,12	> 70	5 - 5,6
Talc-stone	1,69	< 70	

TABLE

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#### CHAPTER III

# THE ELECTRICAL BREAKDOWN OF LIQUID HELIUM

# 3.1. Introduction.

The electrical breakdown or dielectric strength of an insulator is the maximum electric field strength that a material can withstand without rupture. All kinds of insulators used for scientific and technological applications have been studied. Nowadays, there are engineering standards for any insulator material. But the liquified gases, as insulators, represent a special type. Particularly liquid helium, which from the technological point of view is still rather an exotic one. Due to the rapid advance of the applications of superconductivity and its perspective for the near future, liquid helium acquired a great importance. It plays a double rôle, namely: it is the cooling agent to obtain low temperatures in order to transform normal conductors into superconductors and it is the insulator. Therefore, the knowledge of values for the dielectric strength of liquid helium became technically important. On the other hand, from the scientific point of view, in the study of electrical breakdown in general, and in the case of that of liquid helium in particular, one does not know which factors are relevant or not. Thus must be investigated Zie employe and experiments which may or may not be significant should be reported. Moreover, all over the world only

a few groups are doing research on the electrical breakdown of liquid helium.

It is necessary to look for a better and better reproducibility by means of the fixation of all the parameters involved, which is what has been done during the last six years. This signifies the realization and repetition of very naïve experiments with a rather complicated instrumentation.

The resistivity of liquid helium within the temperature range from 4,22 to 1,28 K including the  $\lambda$ -point was estimated by Wolfke and Keesom (Keesom, 1942) of the order of magnitude of 10<sup>15</sup> ohm.cm. Nowadays, the resistivity of liquid helium is estimated to be of the order of magnitude of 10<sup>18</sup> ohm.cm. in accordance with our observations. It is notable that in these estimations the purity of the liquid helium is not mentioned. The purity is a parameter of great importance and therefore it is considered in the studies of the electrical breakdown. Although, paradoxically enough, in technological applications the liquid helium is not and never will be pure. However, it is noteworthy that there are no apparent reasons for not considering the existence of an intrinsic electrical breakdown, at least as a theoretical limit. Later on, enumerating the theories of the electrical breakdown, this particular one will be mentioned Zle erratum again.

# 3.2. Apparatus.

All the experiments made and the results obtained presented in the following paragraphs have been accomplished using the same type of electrodes. The test cell described in the paragraph 3.4 permits a better control of many parameters involved in the phenomenon. Further modifications in the test cell itself, as the ones shown in paragraph 3.7, are only technical improvements but the principle is kept exactly the same. As a result of these last improvements both inside the test cell and in the external instrumentation, a reasonable reproducibility was obtained. This means, that the good control of other parameters is as important as the good control of those that intuitively could appear as more important, namely, the gap distance and the purity of the helium.

It is now worthwhile to make some remarks: in the first place, that with the present equipment one is not able to make a fine discrimination between the phenomenon of electrical breakdown and pre-breakdown. The breakdowns are recorded by our automatic equipment while the pre-breakdowns are not. Perhaps it is due to a lack of sensitivity of the external instrumentation.

Secondly, in spite of all the care taken the purity of the liquid helium in the test cell is far from satisfactory, and in this respect no

solution is in sight. If it were possible to build a very small cell containing only the electrodes and the electrical contacts, it is very probable that the purity of the liquid could be improved. But then another problem arises which is very difficult to solve. That is, to avoid the increasing of the helium vapour pressure inside such a small cell.

# 3.3. Models and theories of the electrical breakdown.

There is no completely satisfactory theory of the phenomenon of the electrical breakdown, neither for solids nor for liquids nor for gases. There are, however, some models for mechanisms which are the bases of physical theories; some examples will be given later on. Some so-called theories are only explanations ad hoc of what happens in organic liquids, which were and are the most used in electrotechnics . Between these explanations and a theory of the electrical breakdown of a liquefied simple gas (4He, H , N etc.) there is a very big gap. Moreover, in physics one very seldom makes a theory for complicated molecules which afterwards is applied to simple ones. A discussion of these theories is given in Appendix II whereas some basic ideas are given here.

Kok<sup>(1)</sup> developed a theory for the electrical breakdown of insulating oils which is only a

typical explanation. A non-uniform field impels impurities of high dielectric constant present in the liquid to migrate into the region of the highest field, were they form a bridge along which the breakdown takes place. Furthermore, the defects on the surface of the electrodes reinforce the non-uniformity of the field. It is obvious that this cannot be used to explain the electrical breakdown of pure liquified gases.

Sharbough and Watson <sup>(2)</sup> developed a thermal theory especially for n-hexane. This hydrocarbon can be prepared with a high grade of purity. According to the authors the breakdown occurs through a bubble formed by the local energy input. Here, once again, intrinsic electrical breakdown will not occur. Even in the case where this model could explain all the experimental results obtained from the measurements so far found including those of liquid He I, it would fail for the case of liquid He II. The existence of a bubble in liquid He II is only possible in a very particular case (3,4). This is when one uses as a cathode an emitter which is a very thin wolfram pin with a very sharp tip. At the tip the intensity of the field is so high and the power so great that the sharp tip of the cathode glows, and this is only possible in a gas atmosphere, i.e., in a bubble formed around the tip.

There is, however, a physical theory developed by Swan  $^{(5)}$  for liquid dielectrics which might be applicable to liquid He as well. Swan assumes electron emission and collision ionization in the liquid and that the cummulative effects of the applied field and the space charge of the positive ions produce a continuously increasing electron current at the cathode due to Fowler-Nordheim cold emission. A feature for this theory is that Swan considered the ionization in the liquid to be very small, even at breakdown. The following relation between breakdown field strength ( $E_0$ ) and the gap distance (d) between the electrodes was derived

$$d = \left[\frac{\varepsilon_{o}\varepsilon_{r}\mu_{+} (\sqrt{1+8\varepsilon_{o}/B-1})}{AC (\sqrt{1+8\varepsilon_{o}/B+1})}\right]^{\frac{1}{2}} \exp\left(\frac{D}{2\varepsilon_{o}^{\frac{1}{2}}} + \frac{B(5-\sqrt{1+8\varepsilon_{o}/B})}{8\varepsilon_{o}}\right)$$

where

ε<sub>o</sub> is the permittivity of vacuum,
ε<sub>r</sub> the dielectric constant of the liquid,
μ<sub>+</sub> the ionic mobility of positive ions,
A and B are constants depending upon the
material of the cathode, and
C and D constants depending upton the
nature of the liquid.

The validity of Swan's theory for liquid He was checked with the fifteen experimental curves <sup>(6)</sup> of figure 1a, paragraph 3.8.

Finally, as far as the development of a theory for the electrical breakdown of liquid helium is concerned as a particular case of a theory of liquified gases, it is only possible to affirm what is generally admitted: that for the so-called mechanism of the electrical breakdown in general three conditions are necessary but not sufficient, namely:

 a) The production of free electrons from the cathode by cold electron emission according to the Fowler-Nordheim law

$$j_e = A\mu^2 E_c^2 \exp(-\frac{B}{\mu E_c})$$

where

j is the electron current density,

E the electric field at the cathode,

- A and B are constants depending on the cathode surface, and
- µ is a constant called local field enhancement factor which depends upon the polish of the cathode surface;
- b) The acceleration of these electrons by a field and a loss of energy through collisions or scattering with the liquid and the impurities;
- c) The ionization which, with a great probability, is responsable for the instability and the erratic results, that is, the presence of a conductive state between the electrodes.

As a matter of fact, as far as the electric breakdown of liquefied gases is concerned the following questions are still open, namely: is it a macroscopical phenomenon or is it a macroscopical manifestation of a microscopical phenomenon?

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3.4. A NEW TEST-CELL FOR THE ELECTRICAL BREAKDOWN OF LIQUID HELIUM

J.M. Goldschvartz and B.S. Blaisse.

During the search for new experimental facts and more coherent results in the electrical breakdown in liquid helium a new apparatus has been evolved. This is shown in figure 1; this shows how the requirements for the apparatus were met and how it functions.

The gap g between the electrodes can be adjusted precisely from the outside by means of the stainless steel cap m with ball bearings b and the bellows B. By turning the cap the length of the bellows is changed so changing the width of the gap g. With the mechanism indicated in figure 1 it is possible to avoid variations in the bellows length as a consequence of the evacuation of the apparatus or of increase of pressure.

The measurement of the electrode gap is performed in the following way. An ohmmeter is connected to both electrodes. By turning m the gap between anode a and cathode c is decreased till they just touch. This is indicated by the ohmmeter. Now the micrometer M is turned till it stops at the removable spherical surfaces at



the top end of the bellows. The reading on the micrometer gives the 'reference point'. Subsequently, the micrometer is rotated in the opposite direction, the cap m is turned, and the travel of S, which is equal to the gap between the electrodes, is measured with the micrometer.

The apparatus has been constructed in such a way that during a series of measurements different parts of the cathode can be used. This has been achieved in the following way. The anode a is mounted eccentrically on the stainless steel tube t. This tube is connected to the upper part with the bellows B by the precision ball-bearings r and q. Now, rotation of this tube t, and thus the displacement of the anode a over the cathode c, is made possible by attaching a soft iron piece i clamped to t, the axis of i being perpendicular to that of t, and by mounting an external permanent ferrite magnet F, mounted on a gear device. Now a rotation of F achieves a rotation of t.

In order to replace the electrodes quickly, the test-cell must be easy to dismantle. Another requirement is tightness for vacuum, and for superfluid helium. This has been achieved by a construction in four parts. The connections between these parts have metallic and also standard Pyrex flanges and, as indicated by Brewer, Champeney and Mendelssohn <sup>(2)</sup>, gaskets of indium at the flanges B and B', or of aluminium in the neighbourhood of the bellows B. Gaskets of indium and aluminium are indicated by In and Al, respectively, in figure 1.

A vibration of the tube t results in a displacement of the anode with respect to the cathode. Then, the end of the anode will remain on a sphere with a radius of curvature roughly equal to the length of the tube t. As a result of the vibrations the gap between the electrodes may change periodically. In order to avoid the uncertainty in the gap distance, two measures have to be taken.

- (1) The vibration of the anode support is reduced as far as possible by means of three phosphorbronze spiral springs d mounted on a small ball-bearing on the tube t and leaning against the internally polished wall of the copperglass seal n.
- (2) The cathode is mounted perpendicularly to the axis of the tube t. Moreover, the cathode is saucershaped with a radius of curvature equal to the effective radius of the above sphere. When using the six small cathodes as indicated in the drawing (at A<sub>4</sub>) the surfaces of these cathodes are tangential to the sphere.

The control of the correct position of the electrodes is performed in the following way. At the top of the mechanism A and beneath the

reference piece S a glass window sealed with an indium gasket has been mounted. Thus it is possible to look through tube t to the cathode by taking away the spherical metal part S. In the centre of the cathode a cross has been engraven; at the bottom end of tube t a cross-wire has been mounted. When the operator turns the control screws of A and A the centres of both crosses are made to 3 coincide. This can be checked by looking through the window.

Next, the 'reference point' in the measurement of the gap is measured with an ohmmeter (see above). Then the tube t is rotated and the reference point is measured again. This point should coincide with the first one. Using the screws A and A, this can be achieved by iteration, whereas at the same time the centres of both crosses must coincide.

The tube t has some holes along its length in order to avoid a difference in the level of the liquid helium inside and outside it. The hollowmachined pieces of Perspex P and P to which the parts of the tube t are fastened act as a highvoltage and thermal insulators. The main tube of the apparatus is made of Pyrex, which gives a good and reliable electrical insulation and a low heat conduction. It is connected to the upper metal part and to the metal flange B' by means of electrolytic oxygen-free copper-glass seals n and 1

The electrical lead from the power supply is connected to the anode a through the terminal L (see section B-B) at which is fixed the support n with a glass tube p. Inside this glass tube is the 0,2 mm diameter tungsten wire connection k, from the accessible screw w to the anode. The tungsten wire is covered by a flexible glassfibre tube o. The terminal L is made tight to superfluid and vacuum by an indium gasket.

During the measurements it is important that the level of the liquid helium is higher than that of all metal parts to which high tension has been applied. In the opposite case an electric discharge through the helium gas will originate between this part and the top of the apparatus, which is earthed.

In order to avoid such a discharge from the electric lead connected to the power supply this lead is insulated over its whole length by a polythene tube, welded vacuum-tight to the lead, and the terminal L is as far below the electrodes as possible. The electrical leads inside the helium Dewar are made of 0,3 mm diameter stainless steel wire. The superleak 1 is made of the material developed by Norberg<sup>(4)</sup> and tested by Atkins et al. <sup>(1)</sup>, i.e. a porous Vycor glass plate 3,5 cm in diameter and 5 or 8 mm thick. Thus the space round about the electrodes can be filled with very pure helium four

because the helium three is carried by the normal component in accordance with the two-fluid model of helium II.

The low temperature arrangement described permits measurements of the breakdown not only in liquid helium but in any kind of liquefied gas. In this case the Vycor plate has to be replaced by an ordinary one. The gas is condensed inside the apparatus through the side tube T. Helium can also be condensed in this way in the apparatus; then the helium evaporating from a liquid helium container is led into the apparatus through a charcoal trap at liquid nitrogen temperature. In order to obtain good measurements the helium must be very pure.

Filling procedures both without and with the superleak will now be described. Without the superleak a vacuum of  $10^{-8}$  torr, as indicated by a high sensitivity Penning ionization gauge (Philips tube W 4.303.64), is obtained by means of an oil diffusion pump and with a liquid nitrogen trap (Edwards F 203 and NTM 2 A, respectively). The high-vacuum part of the system is isolated from the moderate vacuum part by an all-metal valve (Edwards MC V.4). Subsequently the electrodes are outgassed by induction heating (coil b). Then the apparatus is rinsed with pure helium evaporated from a liquid helium container after passing through a charcoal trap at liquid nitrogen tempe-

rature, as indicated above, and filled with helium at 1 atm. After cooling to helium temperature, the pure helium is condensed in the apparatus.

With the superleak, the best vacuum obtained is now only 10<sup>-5</sup> torr without cooling the trap and  $10^{-6}$  torr with the trap at liquid nitrogen temperature, because the vacuum in the Dewar surrounding the apparatus cannot be made high and the Vycor allows the passage of some gas. Therefore, the Dewar is first rinsed and filled with pure helium at room temperature. After that, it is evacuated and filled with liquid helium from the container. Then the temperature is reduced well below the  $\lambda$ -point by pumping the Dewar vessel. As a result the cell is filled with liquid helium through the superleak. In the vacuum system a number of valves (Edwards Quarter Swing Valves BQSB 1) have been mounted permitting the circuit to be connected for the reactivation of the charcoal.

For measurements at  $4,2^{\circ}$  K the apparatus can be connected directly to a gas-holder. In order to obtain temperatures down to  $1,1^{\circ}$  K the apparatus can be connected to the moderate vacuum system. Moreover, a separate vacuum system with a capacity of 60 m<sup>3</sup>/hr (Galileo pump V.2d) serves to reduce the pressure in the helium Dewar.

A standard d.c. power supply with an output of 30 kV and 1 mA was used after a slight modification due to the application of an automatic remote control. In fact, to avoid the human factor in the voltage control a very simple electromechanical device was added. This consists of a multi-turn potentiometer of 10 turns with a dial scale of 100 divisions per turn, driven by a 250 rev/min synchronized motor through a set of reduction gears with which two angular constant speeds can be obtained: 1,2 and 6 sec/turn. In this way, and because there are two scales in the power supply with the total length of the potentiometer in both cases, four 'electrical speeds' are obtained: With the 30 kV scale, 2,5 kV per second and 0,5 kV per second; with the 8 kV scale, 0,66 kV per second and 0,13 kV per second.

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#### 3.5. ELECTRICAL BREAKDOWN OF LIQUID HELIUM

J.M. Goldschvartz and B.S. Blaisse.

# ABSTRACT

An interim report of the measurements on the electrical breakdown of liquid helium at steady voltages as a function of the electrode gap distance and the temperature is given. The influence of the rate of rise of voltage has also been studied as well as the damage of the electrodes by the formation of craters. Special attention has been paid to the purification of the helium, since small particles of solid air or dust, for example, have a strong influence on the results obtained.

#### I. INTRODUCTION

As an extension of the experiments on the electrical breakdown of liquefied gases performed by Kronig and Van de Vooren (1942) with argon, nitrogen and oxygen, one of us (Blaisse et al. 1958) made the same kind of measurements with liquid helium. Since that time a great number of experiments have been carried out, but this paper only deals with some of the experimental results on the electrical breakdown of liquid helium obtained since September 1962.

In all these measurements a spherical tungsten anode of 5 mm radius and a flat stainless steel cathode were used. The stainless steel used was the AISI-316, American Code, C-0,05; Si-0,5; Mn-1,5; Cr-17,5; Ni-12 and Mo-2,4 and the tungsten was pure forged wolfram for industrial applications.

The cleaning of the apparatus and the helium is very important. As pointed out by Kok (1961) and by Arkhipov and Shal'nikov (1959) among others, contamination greatly influences the results obtained. During the first experiments mentioned below the glass tube (test-cell) containing the electrodes was first rinsed with degreased cotton and pure aceton and afterwards with alcohol, and left in a desiccator for one night. The next day the electrode set was cleaned again and mounted in the clean test-cell which was pumped out and left for one night in a moderate vacuum and then pumped out again and filled with helium gas for some hours, as a contact atmosphere in order to cool the test-cell. Afterwards this gas was pumped out and the space was filled with liquid helium. In the later experiments it was not possible to rinse the tube with hydrofluoric acid solution; instead it was cleaned in the same way as the electrodes.

In the bottom of the tube containing the electrodes either a combination of two filters (bacteria asbestos, pores 0,7 µm, followed by sintered glass, pores 5 µm, to retain detached asbestos fibres) or a superleak filter (porous Vycor glass, pores between 40 and 100 Å) were mounted. The latter was much the better and was used in later experiments. Well below the  $\lambda$ -point only the superfluid can pass through the Vycor glass - all impurities are removed, even the isotope <sup>3</sup>He.

# 2. EXPERIMENTAL RESULTS

In figure 1 curves are drawn through points indicating the highest field strengths, each point being based on ten breakdowns. These measurements were performed in one day with the same batch of helium and are typical of several series done on other days during the autumn of 1962. Figure 1 shows a jump in the electrical breakdown value for any fixed gap distance when the temperature passes the  $\lambda$ -point. Moreover, one can see a dependence of the electrical breakdown on the gap distance and on temperature for gaps larger than 20 µm.

Figure 2 gives a graphical comparison between some of our results of 1962 and those obtained by Blank and Edwards (1960), who used larger gaps than ours and used stainless steel electrodes which were both spherical (9,2 mm diameter). At





Figure 1. Electrical breakdown as a function of gap (1962).

Figure 2. Electrical breakdown as a function of temperature. 1000, 500 and 150 µm, Blank and Edwards (1960); 80 and 40 µm, present measurements (1962).

distances of 1000 and 500 µm there is a rather small dependence on temperature and none at all at 150 µm, a distance bigger than our largest gap. Our points correspond to gaps of 80 and 40 µm. Obviously, due to the narrower gaps, our values of the electrical breakdown are higer.

In figure 3 results are given of measurement made in October 1963. At each gap distance and at each temperature thirty breakdowns were performed.





Each of the figures gives three curves, one with the highest values obtained at each gap distance, the second with the mean values of the ten highest breakdown values and the third with the mean values of the thirty. The root mean square deviations are respectively 12% and 20% for a gap of 10  $\mu$ m; 25% and 19% for a gap of 100  $\mu$ m, at 1,32 and 4,2° K respectively. We should like to remark that reproducibility of breakdown voltages with an accuracy of 20-30% is considered fair, as was pointed out by Ionov (1960).

All these measurements were performed without switching off the high tension immediately after a breakdown. This resulted in rather pronounced craters, especially in the stainless steel cathode, as can be seen in figure 4 (plate \*). This picture also gives a comparison between the behaviour above and below the  $\lambda$ -point. At the left-hand side are seen the craters resulting from breakdowns at the boiling point and at the right those of the same number of breakdowns below the  $\lambda$ -point at 1,7° K.

This particular distribution of the craters, in which one observes a greater spread of spots on the cathode below the  $\lambda$ -point than above, could be due to a larger mean free path of the ions in liquid He II. Another difference between breakdowns above and below the  $\lambda$ -point is that the noise of the breakdown is much more pronounced below the  $\lambda$ -point and increases as the temperature is lowered.

When a crater is formed, the gap distance is diminished at certain spots; the local electric field is increased both by this effect and by the curvature of the high spots. Thus, the electron emission from the cathode tends to increase. On the other hand, the electrode material is heated and melted and is thereby cleaned; this

\* Plates at the end of this paper.

results in an increased work function and thus in a decrease of the electron emission. Figure 4 indicates that breakdowns do not take place preferentially from the craters produced previously, as the scatter is very pronounced. This means that the increase of the work function produced the greater effect.

In order to distinguish the two effects, we examined the electrode surface after the formation of the craters by breakdowns in liquid helium and by breakdowns in vacuum of about 10<sup>-8</sup> torr at room temperature. All these breakdowns were performed in the same apparatus. The measurements of the surface roughness were recorded with a Talysurf (model 3 from Taylor, Taylor and Hobson Ltd.). We found that there are hills formed with the molten material of the electrodes and that the height of such hills is sometimes of the order of 5-6 µm.

In figure 5 we can see two of the enlarged profiles obtained in this way. The profile (a) corresponds to a cathode with breakdowns in liquid helium below the  $\lambda$ -point. Profile (b) corresponds to a cathode at room temperature in high vacuum. The formation of hills also takes place on the tungsten anode surface; the damage is less than on the cathode. Using resistors of 60, 30 and 15 MΩ between the cathode and earth, we produced electrical breakdowns, but the profiles we obtained were not appreciably

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Figure 5. Profiles on the cathode surface. Profile (a) at liquid helium temperature; profile (b) at room temperature in high vacuum.

different from those obtained without resistors in series with the cathode.

When producing single breakdowns we found, as could be expected from experiments of Maitland (1961), that these breakdowns were by no means simple. The cut-off was effected with an electromechanical relay. The relay time was about 0,03 sec. A 'single' breakdown is in fact composed of a lot of holes or a colony of craters. Figure 6 (plate) shows an enlargement of one of them. Another phenomenon we obtained was that of the so called pre-breakdowns. This was easily detectable in the dark by the naked eye as a small spark between the electrodes. The current in this spark was so small that the power supply was not cut off automatically. However, this could then be done by hand. These pre-breakdowns have also been found by most of the workers in the field in vacuum as well as in liquefied gases. We found that the spot produced by a single pre-breakdown was a colony of groups of small spots.

Although the influence of the rate of rise of voltage on the breakdown voltage has seldom been mentioned in the literature on the electrical breakdown of liquefied gases, it is a well-known phenomenon in engineering since all the standards of testing the electrical strength of insulating materials (A.S.T.M., D.I.N. and British Standards) prescribe a definite rate for each particular case. We made a slight modification to our power supply by adding a simple electromechanical device to keep the rate of rise of voltage constant. Figure 7 shows the voltage of sixty breakdowns for a fixed gap of 30 µm in liquid helium at boiling point.

The first 20 breakdowns were made by increasing the voltage manually. This series is indicated by A







Figure 8. Breakdown voltages at a fixed gap produced by hand and with automatic control. A, C, automatic at 2,7 kV sec<sup>-1</sup>; B, by hand waiting 1 minute (15th October 1964).

in figure 7. Subsequently 20 breakdowns were performed with an automatic constant voltage rise of 2,7 kV sec<sup>-1</sup> (series B). Finally series C was a repetition of series A. The speed of rise of voltage of the hand control was much less than 2,7 kV sec<sup>-1</sup> and was moreover irregular. The breakdown voltages made with constant rise of voltage are higher than those made by the slower hand control.

In figure 8 one sees the influence of pre-stressing. Series A was performed with constant rise of voltage of 2,7 kV sec<sup>-1</sup>. Each point of series B was obtained by rising the voltage to a value near the breakdown value, waiting for one minute and then raising the voltage manually to produce the breakdown. Here too the mean value of the points obtained with constant speed is higher.

The measurements of figures 7 and 8 were made with an apparatus (Goldschvartz and Blaisse 1965) especially designed for obtaining a high vacuum and for introducing helium into the electrode space, either by condensation or by filtration through a superleak of porous Vycor glass. The experiments were carried out with condensed helium.

Finally figures 9 (a), (b), (c), (d), (e) correspond to experiments we made obtaining the helium by filtering through a superleak of porous Vycor glass. The sequence was (a), (b), (c), etc. 9(a) and 9(b) have lower breakdown voltages than 9(c) and 9(d) respectively, illustrating a time effect which we often observed. By taking the averages of 9(a) and 9(e) and also 9(b) and 9(d) one finds here higher breakdowns at a higher temperature. This behaviour is different from that of figure 1. However, in the measurements of



Figure 9. Electrical breakdown of helium which had passed through a superleak (29th November 1963). O, highest values; X, average of the 10 highest values; C, average of 20 values.

figure 1 special attention was given to breakdowns just above and just below the  $\lambda$ -point.

# 3. CONCLUSIONS

The reproducibility of the measurements made on different days leaves much to be desired. The main factor is probably the different degrees of purity of the helium. Differences in the condition of the electrodes are also important, as observed in the measurements on liquid argon by Swan and Lewis (1960, 1961), but in our measurements we always had the electrodes in the saturated state of oxidation.

Over the years we increased the purity of the helium; probably as a consequence the breakdown voltages increased too as can be seen in figure 10, where the highest breakdown strengths are plotted against gap width for different temperatures and different years. We hope to improve the purity further in order to obtain intrinsic breakdown values. At the moment we do not know if we are approaching these values or if many more experiments have to be performed.

# ACKNOWLEDGEMENTS

The authors wish to thank Ir. H. Nienhuis, Ir. M. van Asperen, Ir. K.J. Schell and Mr. F. Landheer, who performed the measurements and discussed the subject with us.


Figure 10. Comparison between some of the curves of the electrical breakdown obtained in the last years. A, 31st October 1963; B, 19th March 1962; C, 19th September 1962; D, 18th May 1961; E, 1957.

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Figure 4. Stainless steel flat cathode (magnification 8x before reduction of photograph by  $\frac{1}{2}$ ) with distribution of craters at 4,2 K (left) and 1,7 K (right).

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Figure 6. A colony of craters and holes on the cathode surface due to a 'single' breakdown (x65 before reduction of photograph by  $\frac{1}{2}$ ).



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3.6. DESTRUCTION OF ELECTRODES IN THE ELECTRICAL BREAKDOWN OF LIQUID HELIUM

J.M. Goldschvartz, F. Landheer and B.S. Blaisse.

In the search for the reproducibility of the values of the electrical breakdown of liquid helium, the need to study the damage of the electrodes surfaces has become apparent, partly to see how the formation of craters and hills on the surface affects the values of the breakdown voltages and partly because the factors relevant to the characteristics of a breakdown are not known exactly.

Earlier results obtained <sup>(1)</sup> are: A greater spread of spots or craters on the cathode surface occurs below the lambda point than above, a "single breakdown" produces a spot which is not just one hole but a colony of craters, and, finally, the spot produced by a "single prebreakdown" is also a colony of groups of small holes. This letter deals whith these single breakdowns between electrodes of the kind used since this investigation was started <sup>(2)</sup>, i.e. a spherical tungsten anode of 5 mm radius and a stainless steel cathode both contained in the test cell <sup>(3)</sup>.

In order to see the extent to which those prebreakdowns and main breakdowns - and consequently

the destruction of the electrode surfaces - are dependent upon the sensitivity of the external instrumentation, an electronic relay was designed following (4). Producing only single breakdowns and using resistors in series in the electrode circuit we could detect time dependent differences in the damage of the cathode surfaces. In shortening the duration of the cutoff time of the applied voltage we tried to get rid of the prebreakdowns as well. These differences are shown in Fig. 1, while Table 1 contains all features of these photographs.

Unfortunately, it was not possible to obtain a single breakdown with the electromechanical relay of the power supply using some reasonable resistor in series, due to the fact that the current during the breakdown was never sufficient to energize the relay. These breakdowns were done just above the lambda point. The test cell was filled with liquid helium through a Vycor glass superleak.

Fig. 1b and 1c are particularly interesting because both were obtained at the same temperature, without any resistor in series, and with electronic and mechanical relay, respectively. The latter shows by far the greater amount of damage.

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Fig.	1	Type of relay *)	Resistor (MΩ)	Magni- fication (x)	Gap (µm)	Т ( К)	E_ **) (MV/cm)	Cutoff time (s)	Influence	Maximum extension of damage (µm)
a) b)		E	50	500 500	96 99	2,21	0,41 0,42	$20 \times 10^{-6}$ $20 \times 10^{-6}$	resistor cutoff time	16 (one big crater) 70 (colony of craters)
c)		М	_	500	94,5	2,18	0,57	30 x 10 <sup>-3</sup>		30 (the bigger crater) and 120 (colony of craters)

### TABLE 2

Fig.	2	Type of relay *)	Resistor (MΩ)	Magni- fication (x)	Gap (µm)	Т ( К)	E <sub>o</sub> **) (MV/cm)	Cutoff time (s)	Influence	Maximum extension of damage (µm)
a)		М	-	500	50	4,2	0,66	$30 \times 10^{-3}$	cutoff	58
b)		Е	-	500	50	4,2	0,49	20 x 10 <sup>-6</sup>	time	29 (the bigger crater)

\*) E electronic relay

\*\*)  $E_{o}$  electrical breakdown value

M electro-mechanical relay



Figure 1. Craters on the surface of stainless steel cathodes produced by 'single' electrical breakdowns in liquid helium just above the  $\lambda$ -point; 500x; a) and b) cutoff time regulated by an electronic relay and c) by an electromechanical relay.



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- Fig. 2. Craters on the surface of stainless steel cathodes produced by single electrical breakdowns in liquid helium at boiling point; 500 x; a) cutoff time regulated by an electromechanical relay and b) by an electronic relay.

Perhaps the most demonstrative pictures are Fig. 2a and 2b representing single breakdowns just at boiling point. The test cell was filled with liquid helium by condensation. Table 2 gives the corresponding data.

We think that from the above mentioned examples, chosen from a lot of experiments, the influence on the destruction of the electrodes due to the duration of the breakdown is clear and at least as important as the influence of a big resistor in series.

Besides, it is also clear that these results are consistent with <sup>(1)</sup>. Nevertheless, the problem of the nature and the total elimination of the prebreakdowns remains. In this respect the present experiments are not conclusive. Could the prebreakdowns be totally eliminated increasing the sensitivity of the external instrumentation and diminishing even more the cutoff time?

We thank J.R. de Haas for the design of the electronic relay and the Delfts Hogeschool Fonds for financial support of one of us (F.L.).

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# 3.7. PARAMETERS INVOLVED IN THE ELECTRICAL BREAKDOWN OF LIQUID HELIUM

J.M. Goldschvartz and B.S. Blaisse.

#### INTRODUCTION

The present and future applications of superconductivity, particularly the future possibilities of the transmission of power by means of superconductors, mean that the electrical breakdown of liquid helium has become a matter of great importance. In fact, liquid helium can act both as an insulator and as a means to obtain the low temperature necessary to reach the transition temperature of most of the metals to become a superconductor. Nevertheless, as far as the literature reveals, hitherto little attention was paid to this subject. Only a few groups are working on the electrical breakdown of liquid helium, namely: Mathes (1) in the United States of America, Fallou and Galand <sup>(2,3)</sup> in France, and ourselves <sup>(4,5,6,7)</sup> in the Netherlands, although Blank and Edwards <sup>(8)</sup> published a paper on this matter in 1960.

The aim of this paper is by no means to present either final results or to be an up-todate résumé. Yet, in view of the current importance attached to the knowledge of the electrical breakdown of liquid helium, this paper is meant to indicate our progress in this matter and to criticize it, considering the different parameters involved, how these parameters could be commanded and moreover the fact that the results obtained were always rather disconcerting. Those parameters are:

- Purity of the liquid helium;
- Gap distance between the electrodes;
- Temperature;
- Speed of rise of the voltage applied;
- Time lag between breakdowns;
- Duration of a breakdown and destruction of the electrodes;
- Prestressing effects.

#### PURITY OF THE LIQUID HELIUM

We finished a previous paper <sup>(6)</sup> on the electrical breakdown of liquid helium showing the situation with the representation of the experiments made till that time (the end of 1963). Figure 1 shows the curves taken at boiling point (4,2 K) and at different temperatures below the  $\lambda$ -point of the breakdown strength as a function of the gap distance. The curves were plotted with the highest values obtained for each gap distance and the rise of the voltage was controlled by hand. This procedure of taking the highest values was based on the fact that we



Figure 1.

considered these values to be the nearest approximation to the values of the intrinsic breakdown strength. Observing these curves we concluded that the more we improved the purity of the liquid helium, the higher the values for the breakdown voltage.

In figure 2 the curves are drawn which were obtained with the equipment which has been considerably improved upon in the last few years. Each point represents the mean value of fifty values of the electrical breakdown for each gap distance. For the purpose of comparison with the curves of figure 1, the curves corresponding to the highest values of the breakdowns are also drawn and the scale is the same in both figures. For symplifying things and for practical reasons, only one batch of liquid helium was used in the Dewar during a whole one day experiment, and the measurements of figure 2 were performed only at boiling point.

It is evident that curves of figure 2 are lying in a lower region than those of figure 1, even those corresponding to the highest values. Here must be remarked that for <u>all</u> experiments of figure 2 we used a Vycor glass superleak as a filter to keep the liquid helium free of macroscopic impurities, whilst a superleak was not used in the experiments of figure 1. Here must be also pointed out that, in the case of figure 2e, we had a crack in the Vycor glass



Figure 2	Date	Symbol of the points	Mean val. stand. dev. (MV/cm)	Mean val. stand. dev. (%)
a b c d e	5/VI/68 5/VI/68 7/XI/68 7/XI/68 19/XII/68	+ <b>A</b> 0 <b>Q</b>	0,025 0,022 0,023 6,018 0,040	2,6 2,6 2,7 2,2 2,9

superleak. Probably, we had less pure helium. Nevertheless, the curves of figure 2e are shifted to a higher position. This fact would indicate a contradiction: the liquid helium is less pure and the values of the breakdown voltages are higher than those of the curves of figure 2a, b, c, d. Features of the curves of figure 2 are shown in the preceding table.

As a matter of fact, the control of the purity of the liquid helium is very difficult if not impossible, and one can never know exactly how clean the cell and the helium are. Before each whole day experiment the cell was chemically cleaned and then, after being mounted, it was evacuated. But in the cases of figure 1 and figure 2 there are some important differences. In the former the apparatus was pumped during one night before filling it with liquid helium; after this only a moderate vacuum of order of 10<sup>-1</sup> torr was obtained. On the other hand, in figure 2, apart from the fact that all the experiments were made with the improved equipment <sup>(5)</sup> plus all the changes described here, the dismountable bottom part of the cell which contains the electrodes, figure 3 and 4, was always cleaned in an ultrasonic system. Then this part was mounted and a vacuum of the order of  $10^{-5}$  torr was maintained for about two days. Even so the curves are kept in a lower region.

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#### THE GAP DISTANCE

The gap distance is a relevant parameter in the study of the electrical breakdown and one should be able to control it very carefully. In the case of the electrical breakdown of liquid helium the control of this gap distance is not at all simple, for the gap distance must be changed and controlled from the outside of the cryostat, i.e., from the outside of the cell, which is inside the liquid helium Dewar which, in turn, is inside the liquid nitrogen Dewar. Since the new test cell for the electrical breakdown of liquid helium was designed <sup>(5)</sup>. we have introduced numerous changes, some slight and some important for the better performance of the apparatus as a whole. But the principle of operation remained the same.

That which makes things more complicated is that one should be able to turn the support P of the six different cathodes k by pushing the pin p of figures 3 and 4, such that each one of the cathodes can face the anode a. This rotation is commanded from the outside by means of the magnets m fixed on the mechanism M, figure 5. The anode is fixed and faces one of the slit-windows of the Dewars. Thus, one can always see the electrodes from the outside.

To change the gap distance between the electrodes, the support P of the cathodes is





Figure 3 .

Figure 4.

pushed down or up by means of the action of the transmission T and the spring v, figures 3 and 4. This transmission T is commanded by

the head H of the instrument and the support P, figures 3 and 5, is approximately one metre and so is also the transmission T, because the electrodes system must be deep in the liquid helium bath if one wants to make an appreciably long lasting experiment, that is, to obtain a sufficiently large number of breakdowns to give some statistical sense to the whole experiment. The average number of breakdowns we performed in each one of the last experiments, figure 2, is of the order of one thousand.

To measure the gap distance and its changes, we now use the clock micrometer C of figure 5 instead of the normal micrometer in the former equipment. The new micrometer is a very sensitive and accurate instrument (\*): 1 division = 1 µm. It has the advantage that its bottom ruby spherical surface is always pressed by a spring on the head H of the cell which can be moved up and down. This avoids the error introduced each time a contact was made between the micrometer and the head of the cell in the equipments used before. The anode a, figure 3, obviously had to be mounted on an insulation material, in this case a thermoplastic called Delrin. This does not permit the outgassing of the electrodes by means of an induction oven. However, it seems from the reproducibility of the curves of figure 2, that this would not be

(\*) C.E.J. Mikrokators No: 509/4, Eskilstuna, Sweden.



Figure 5.

an important drawback.

With this configuration of the electrodes, and by controlling everything from the outside of the cryostat, we reached a very good constancy of the gap distance, a maximum error of 2,5 µm in all the measurements of the curves of figure 2. This means that 5 µm was the maximum change measured in the gap distance before and after each set of fifty breakdowns.

#### TEMPERATURE

Temperature is another parameter that we controlled in our experiments on the electrical breakdown of liquid helium, although

in the last experiment (those represented in figure 2) we concentrated our attention only on

the measurements at boiling point (4,2 K) due to the uncertainty of our results of previous experiments. However, we can fix any desired temperature between 4,2 K and 1,3 K using a very simple and reliable manostat, as described by Walker <sup>(9)</sup>, but which we improved upon considerably.

#### OTHER PARAMETERS

The electronic equipment permits the direct control of the speed of rise of the applied voltage and the time lag between breakdowns. Moreover it cuts off the breakdowns and thus diminishes the destruction of the electrodes. The total electronic automatic system shown in figure 6 will be described in general. The automatic remote control (4) controls the voltage supply and the speed of rise of the applied voltage to the cell (1) through the electronic relay (3). The cut off time of the electronic relay diminishes the time of the cut off from 0,03 sec, cut off time of the electro-mechanical device of the power supply (2) to 20 x  $10^{-6}$  sec <sup>(7)</sup>. The following table gives the features of this automatic remote control and the power supply.

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DC voltag of the po supply	ge ower	Calibroid of the of the contro	ation dial remote	Gear 5	reduc. : 1	Gear. 25 :	reduc.
8 kV 30 kV		9,15 32,15	V/div. V/div.	0,75 2,7	kV/s kV/s	0,15 0,54	kV/s kV/s



Figure 6.

The number of breakdowns to be accomplished is predetermined in the unit (5) and also the time interval between them in unit (6). The time between one breakdown and the starting point of the rise of voltage for the next one can be set from 10 sec to 9 min 50 sec in steps of 10 sec. The minimum time of 10 sec was chosen because it is somewhat greater than the time necessary for the automatic resettlement of the zero of the potentiometer of unit (4).

Unit (6), which in fact is mounted together with units (5) and (4) in only one unit, delivers the "start" and "stop" signals to the digital voltmeter through unit (7). Unit (9), a binary to decimal code converter, makes the printer (10) deliver the breakdown voltages.

The results hitherto found concerning the destruction of the electrodes, the pre-

breakdowns and the prestressing effects were already described in previous papers (6,7). We did not yet carry out experiments on the time lag.

#### CONCLUSION

On the one hand, the high values of the dielectric strength in figure 1 could be due to the high purity of the helium. They could also be due to the presence of dissolved (possibly metastably) oxygen in the liquid helium which increases the strength of liquid dielectrics, according to Swan and Lewis (10,11). On the other hand, in the case of figure 2, in addition to the fact that these curves were obtained with a highly improved equipment, they could also be a better approximation to the physical reality. They were obtained statistically, taking the mean of fifty measurements for each gap distance, and each of these sets of fifty breakdowns were preceeded and followed by sets of fifty breakdowns at a reference gap distance of 100 um.

This means that, for the time being, almost all parameters involved in the electrical breakdown of liquid helium can be controlled to a reasonable extent and it seems that the values for the dielectric strength of liquid helium at boiling point would be provisionally given by

those curves of figure 2. However, the situation is still uncertain and a considerable number of experiments should be carried out in order to obtain more consistent and conclusive results.

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## 3.8. ON THE ELECTRICAL BREAKDOWN OF LIQUID HELIUM J.M. Goldschvartz, A.C. Ouwerkerk and B.S. Blaisse.

#### INTRODUCTION

In a recent paper <sup>(1)</sup> we have analyzed the different parameters involved in the electrical breakdown of liquid helium and the results hitherto obtained. We showed that not only the purity of the helium and the gap distance are important, but that other parameters are also important in order to obtain a reasonable reproducibility which was obtained with the measurements of the last year.

#### APPARATUS AND PROCEDURE

Since the cell to measure the electrical breakdown of liquid helium presently in use was designed, it has experienced a great number of important improvements. Also the external instrumentation was highly improved and the whole system is automatized. Both the improvements of the cell and those of the external system are described in the paper <sup>(1)</sup> mentioned above.

In order to obtain very pure liquid helium

in the cell it was filled through a wonderstone superleak <sup>(2)</sup>, the onset point of which is below the lambda point. Nevertheless, all the measurements shown here were made at boiling point. Hence, we had to wait a long time during each experiment to reach the boiling point. Moreover, this fact means also a big waste of liquid helium. Consequently, the amount of liquid helium left for an experiment as well as the total time available for breakdown measurements are smaller than in the case of a normal filling at 4,2 K. Nevertheless, the number of measurements used to draw each of the lines shown in figure 1 is 500. This means that each point is the mean value of fifty electrical breakdowns. Furthermore, each set of fifty breakdowns is preceeded and followed by a set of fifty breakdowns at a fixed reference gap distance, in our case 100 µm. In addition, before and after each set of fifty breakdowns, the zero point of the gap distance is checked. This gives an indication of how the surfaces are or were destroyed and also gives, eventually, an idea of the order of magnitude of the error introduced by the damage of the surface of the electrodes.

In all these measurements and since this research was started <sup>(3)</sup> the shape and nature of the electrodes were the same,

namely a spherical tungsten anode of 5 milimeters radius and a flat stainless steel cathode. The stainless steel used was the AISI - 316, American code, C - 0,05; Si - 0,5; Mn - 1,5; Cr - 17,5; Ni - 12 and Mo - 2,4 and the tungsten was pure forged wolfram for industrial applications.

As far as the electrode surfage damages are concerned, by the one hand we have studied the profile of the surfaces (4) with the melten material and by the other hand we have made a light microscipy study of the craters on the surfaces (5).

The conclusions of those two studies are: a. The height of a hill can be as big as 5 - 6 µm; b. a greater spread of spots or crates on the cathode surface occurs below the lambda point than above;

- c. a single breakdown produces a spot which is not just one hole but a colony of craters;
- d. a single "prebreakdown" produces also a
  spot which is a colony of groups of small
  holes;
- e. the damage is time dependent of the duration of the breakdown, i.e., whether we used an electronic relay with a cutoff time of the order of  $10^{-6}$  second or an electromechanical relay with a cutoff time of 3 x  $10^{-2}$  second;
- f. that breakdowns do not take place preferencially from the craters produces previously.

#### RESULTS

The sets of curves shown in figure 1 correspond to the series of the last experiments (lines 5 ... 15) and are drawn together with those included in the previous paper <sup>(1)</sup> (lines 1 ... 4). These curves show the breakdown values  $E_0$  at the temperature of 4,2 K as a function of the gap distance d on a double logarithmic scale. The lines of figure 1 (a) are drawn following the procedure described above while the lines of figure 1 (b) correspond to the highest values of each set of those fifty breakdowns.

The standard deviation of each set of fifty breakdowns is of the order of 2,5%; the mean deviation of the points from the lines corresponds to 0,033 MV/cm.

#### DISCUSSION

In table I are included the values of the parameters a and b for the lines of figure 1, obeying the formula in  $E_0 = a \ln d + c$ , where  $E_0$  is the electrical breakdown in MV/cm and d the gap distance in  $\mu$ m. From these values we conclude that the reproducibility is rather good.

We see in table I that the standard deviations of the a's and c's for the lines of the highest

number of the curve	curves of	the means of 5	0	curves of the highest of 50				
	а	с	mean deviation of the points from the lines in MV/cm	a	с	m <b>e</b> an deviation of the points from lines in MV/cm		
,1	- 0,297	1,01	0,064	- 0,242	1,10	0,081		
2	- 0.343	1,34	0,020	- 0,289	1,38	0,025		
3	- 0,279	0,94	0,028	- 0,275	1,19	0,057		
4	- 0,303	1,14	0,023	- 0,209	1,02	0,014		
5	- 0,161	0,30	0,043	- 0,243	0,95	0,041		
6	- 0,236	0,93	0,044	- 0,250	1,25	0,062		
7	- 0,372	1,50	0,037	- 0,369	1,77	0,045		
8	- 0,200	0,93	0,038	- 0,234	1,33	0,036		
9	- 0,136	0,72	0,024	- 0,176	1,12	0,019		
10	- 0,190	0,88	0,022	- 0,163	1,04	0,034		
11	- 0,218	0,97	0,054	- 0,178	1,05	0,032		
12	- 0,237	1,07	0,007	- 0,218	1,21	0,015		
13	- 0,245	1,30	0,024	- 0,228	1,51	0,053		
14	- 0,345	1,37	0,032	- 0,303	1,41	0,058		
15	- 0,220	1,06	0,008	- 0,254	1,45	0,015		
						141		
mean values	- 0,252	1,03	0,031	- 0,242	1,26	0,039		
standard deviation	7,1%	7,3%	0,12	5,7%	4,6%	0,041		

TABLE I



- Figure 1. (a) Curves drawn with the mean value of fifty breakdowns for each gap distance.
  - (b) Curves drawn with the highest value for each gap distance.

of the fifteen curves of figure 1.

- A: Mean value of the curves of figure 1(a).
- B: Mean value of the curves made with the mean values of the ten highest values of fifty breakdowns for each gap distance (not seen in figure 1).
- C: Mean value of the curves of figure 1 (b).

values of the fifty breakdowns are smaller than the standard deviations for the lines of the mean value of the fifty breakdowns. The reason for this fact could be the following: solid impurities (e.g. solid air) diminish the breakdown strength <sup>(6)</sup>. The amount of these solid impurities will be subject to statistical fluctuations. Thus the standard deviations of a and c of the curves corresponding to the average of fifty breakdowns contains these fluctuations as well as those of the intrinsic breakdown. In this train of thought the breakdowns which correspond to the purest helium in the gap will be high. Thus the highest breakdowns will be near to the intrinsic ones. This argument stands separate from the fact that oxygen may increase the breakdown strength; in the measurements of this paper the oxygen content of the helium in the cell was extremely low due to the filling of the cell through a superleak (1,7,8). In this case the contribution of the fluctuating impurity to the standard deviation of a and c will be less than in the case of fifty breakdowns. For technological application the latter are more important because in practice the liquid helium will never be pure.

#### CONCLUSIONS

Finally in figure 2 we can see the "mean curves" drawn from those of figure 1. Here is included a third curve B which is drawn with
the mean value of the ten highest values of every set of fifty breakdowns for each gap distance. Thus, the curve A of figure 2 could be taken as representing the values for the electrical breakdown of liquid helium at boiling point till beter reproduced values are obtained.

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#### CHAPTER IV

## IONIC MOBILITIES IN LIQUID HELIUM

# 4.1. Introduction

Amongst the different experimental methods for the study of liquid helium, especially at temperatures below the lambda point the use of charged particles has been particularly fruitful. This method has the advantage that the ions can rather easily be controlled and detected. It is a powerful tool which has provided valuable information about the formation of vortices in the superfluid.

If a charged particle moving through the liquid helium under the action of an electric field E acquires a drift velocity u, the definition of the mobility  $\mu$  of such a charged particle is given by the relation

 $u = \mu E$ .

The reason for the importance of this parameter of a charged particle is that it is independent of the field, although in liquid helium it is only "approximately" a constant of proportionality between the drift velocity and the electric field. Experiments show that for weak fields it is rigorously constant while for high fields there are deviations.

According to current views, positive charges in liquid helium form solid spheres with diameter of about 10 Å while negative charges form bubbles of about 20 Å. The positive ion as a (1)solid sphere was studied in detail by Atkins who suggested the action of electrostriction for the formation of the sphere. A molecular ion <sup>4</sup>He<sup>+</sup> is considered. It is very stable, also while drifting through the liquid helium. The electric field of the ion polarizes the neutral atoms in the surroundings. The liquid density increases by electrostriction and the ion drags with it about 100 4He atoms. At a distance of 2 or 3 Å from the ion the pressure due to the electrostriction was found to reach the order of magnitude of 10<sup>3</sup> atmospheres. Therefore, it was necessary buupt to assume that the helium was solid in the vicinity of the ion. Atkins' electrostriction model gives reasonable agreement with experiments.

Negative charges, on the other hand, are considered as electron bubbles. This idea that a negative ion consists of a free electron surrounded by a vacuum bubble was studied in detail by Kuper <sup>(2)</sup>. Now it is commonly agreed, as Du Vall and Celli <sup>(3)</sup> say, that the lowest energy configuration for a negative ion in liquid helium can be described as an electron trapped in a spherical bubble of radius of 10 to 20 Å. The creation of this bubble is a consequence of a short-range repulsion between the electron

and the helium atoms, due to the exclusion principle and by the zero point energy of the electron in the bubble. This bubble model leads to a mobility in good agreement with experiments and explains the mobility and the trapping of ions in rotating helium II. In fact, Careri et.al. <sup>(4)</sup> suggested, as an explanation for the anisotropy they found only for a negative ion current in the direction perpendicular to the axis of rotation, that the negative ions were trapped by the vortex lines. Careri's explanation was confirmed by direct measurements by Douglas <sup>(5)</sup> and by Tanner, Springett and Donnelly<sup>(6)</sup>.

On the one hand, using Van der Waals interatomic attractive forces Kuper estimated the pressure  $P_{v.d.W}$  that the helium atoms exert on the bubble. On the other hand, he also calculated the pressure  $P_e$  due to the polarization attractive forces between the atoms and the free electron. These two pressures have to be equal.

It is remarkable that both Atkins and Kuper found for the effective mass of positive and negative ions approximately the same value, namely, about 100  $M_{4}_{He}$ . As far as the radii of the positive and negative ions are concerned, Kuper found the radius of the bubble to be approximately of 12,1 Å. That is, approximately two times the value of 6,3 Å the radius of the positive ion as found by Atkins. In 1966 Parks and Donnelly <sup>(7)</sup> using a different

method, based on the measured mean lifetimes of ions trapped in vortex rings and lines, have obtained a radius of  $6,44 \pm 0,10$  Å for positive ions and  $14,5 \pm 0,4$  Å for negative ones. The former is in good agreement with Atkin's value but the latter is about 15% greater than the one calculated by Kuper.

Finally, following Careri (1963) it must be pointed out that the motion of ions in liquid helium II under the action of electric fields is quite interesting and mysterious. He calls these ions "hot ions" because of very great discrepancies which appeared, for instance, in the drift velocity measurements made at very low temperatures. Another interesting phenomenon is the already mentioned interaction between ions and vortex lines and vortex rings. Moreover, it seems that discontinuities in the mobilities of ions have appeared below the lambda point (8). These are still open problems which are of capital importance in the microscopic study of liquid helium II in rotation. According to Kapiza's <sup>(9)</sup> statement "... the rotation of <sup>4</sup>He II and the critical velocities remain as the fundamental problem of low temperature physics".

# 4.2. Methods of measuring the ionic mobilities.

The ionic mobilities in liquid helium II mentioned in the foregoing paragraph were all

measured using, in principle, the same method based on the application of an electric field. In general, the ions in liquid helium are scattered by the thermal excitations. The electric field is necessary to maintain a determined drift velocity and produce an ionic current. It is obvious that the lower the temperature the lower the number of thermal excitations. Then one has the possibility of using lower electric fields. High fields, however, can produce interesting results as well, as will be seen later.

The common method of measuring the ionic mobilities is the so-called time-of-flight-technique developed by Careri and his co-workers <sup>(10)</sup>. A set of three electrodes is in the bulk of the liquid helium: a surface near which the ions are produced by a polonium source, a grid at which the accelerating potential is applied and a collector of the ionic current. A square-wave generator provides an alternating field E across the grid-collector distance L. Under these conditions, for certain values of the field and for the square-wave frequency v the ions can reach the collector and are recorded by an electrometer. As v is increased the number of ions able to reach the collector decreases, until at the cut-off frequency  $v_0$  the ions cannot reach the collector and the ion current drops to its background value. Hence the mobility µ

$$\mu = \frac{u}{E} = \frac{2 v_0 L^2}{V}$$

where V is the amplitude of the square-wave voltage applied between the collector and the grid.

Williams <sup>(11)</sup> measured the mobilities of ions in liquid helium applying high fields at the range of temperatures between 4,2 and 1,4 K. His results are mentioned further on in paragraphs 4.3 and 4.4. As far as the bibliography reveals, these were the only measurements in which high fields were used. So far, the ion implantation was made using a radioactive salt of <sup>210</sup>P<sub>o</sub> which is an  $\alpha$  emitter.

In paragraph 4.3, figure 1, a new method (12) of measuring ionic mobilities in liquid helium is described. Its main features are: the use of high voltages and the ion implantation which is made by means of a thin thoriated wolfram cold emitter provided with a very sharp tip at which the field strength is very high. Measuring the ionic current and the variation of the he-lium level  $\Delta$ h inside a narrow glass tube, the ionic mobility is determined.

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# 4.3. A METHOD OF MEASURING IONIC MOBILITIES IN HIGH ELECTRIC FIELDS IN ISOLATING LIQUIDS

B.S. Blaisse, J.M. Goldschvartz and P.C. Slagter.

## PRINCIPLE OF THE METHOD

In figure 1 two electrodes, 1 and 2, are mounted in a glass tube, immersed in a liquid up to the level A. The distance between the electrodes is b. If 1 is the cathode and 2 the anode, a sufficiently high tension will liberate electrons from 1, especially when this cathode has one or more needles on its surface. The negative ions travel to the anode in the electric field E. This field exerts an upward force on the ions which tends to lift the liquid surface in the tube at A. After some time a stationary state is obtained. In this state the difference in the levels inside and outside the tube is Ah. Then the weight of the column of height  $\Delta h$  is equal to the force on the ions. If  $\rho_{m}$  is equal to the specific mass of the liquid, p the charge density, S cross-section of the tube, g gravity constant,  $E_z$  vertical component of E, and  $j_z$ and v are the vertical components of the current density and the velocity of the ions respectively, we have

 $g \rho_{m} \Delta h S = \int_{1}^{2} E_{z} dx dy dz = \int_{1}^{2} \frac{E_{z} j_{z}}{v_{z}} dx dy dz.$ 



1 Cathode 3 Glass tube Immersed in liquid helium 2 Anode A Outside level A' Inside level

Figure 1. Principle of the device used.





The axes x and y are assumed to be horizontal. Now the current I through each cross-section is independent of the height.

$$I = \int j_z \, dx \, dy.$$

If we suppose that the mobility  $\mu = v_z/E_z$  is constant for the different fields existing between the electrodes, we find

 $\mu g \rho_m S \Delta h = I b.$ 

Thus by measuring  $\Delta h$  and I we can determine the mobility  $\mu$ .

## EXPERIMENTS

The method described above was used to measure µ in liquid helium. The internal diamter of the tube was  $\phi_i = 4$  mm. The electrodes were a flat stainless steel anode  $\phi = 3$  mm and in these preliminary experiments a pin-shaped thoriated tungsten cathode (1,5% Th) of  $\phi = 0,25$  mm with a sperical tip of  $r = 1,5 \mu m$  was used. The distance b between both was 10 mm. A tension could be applied up to 30 kV. When the point was negative, we obtained a current. Some results for 4,2 K are shown in figures 2 and 3. In figure 2,  $\Delta h$  is proportional to the current I at different values of the tension V. This indicates that the mobility is independent of the field in the range of electric fields of our measure-\* metns. Figure 3 shows that ∆h depends linearly on V at tensions greater than 8 kV. At lower tensions

Ah and I are probably proportional (1,2) to V<sup>2</sup>. From these results it follows that  $\mu = 0,025 \text{ cm}^2/\text{Vs}$  at field strengths between 2 and 25 kV/cm. This is in good agreement with measurements carried out by Williams (3) who found values of 0,024 and 0,021 cm<sup>2</sup>/Vs for field strengths of 12 kV/cm and 46 kV/cm respectively, and at an electrode distance of 0,44 mm. At shorter distances his values are somewhat higher.

We could obtain neither a current nor a  $\Delta h$  when the tungsten point was positive. This is probably due to the same cause as a phenomenon found by Coelho and Sibillot <sup>(1)</sup> in liquid nitrogen and by McClintock <sup>(2)</sup> in liquid helium. In their experiments on raising V from zero, there was no current until a critical potential V<sub>c</sub> was exceeded. Thereafter it was possible to reduce V considerably - often to  $V_c/10$  - before the current was extinguished. McClintock used a much finer tip (r = 0,05 µm) than we did (r = 1,5 µm). Therefore in his experiments the field strength at the anode could be made higher.

This phenomenon is probably caused by electrostriction  $^{(4)}$ . At the highest fields, of the order of 2 x  $10^8$  V/cm, at the tip surface the pressure by electrostriction amounts to 500 and 5000 atm (x 1,013x  $10^5$ N/m<sup>2</sup>)



Figure 3. Increase of the inside helium level as a function of the applied voltage.

for helium and nitrogen respectively. At these pressures helium and nitrogen solidify at their boiling temperatures as well as at temperatures far beyond this. Therefore when V is raised, a solid sheath of high electric resistance is formed. By cold emission of the tungeden tip in this sheath, positive or negative space charges are built up for positive or negative tip tension. These charges diminish the field strength at the tip surface and reduce the emission current to zero. However, if the tension increases beyond a critical value  $V_c$ , the isolation is broken, the current heats the solid and melts it. Then the current is triggered.

It will be necessary to extend our measurements based on the effect described above to other temperatures, also to below the  $\lambda$ -point and to rotating He II.

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4.4. MOBILITY OF NEGATIVE IONS IN He I AND He II MEASURED WITH A NEW METHOD

B.S. Blaisse, J.M. Goldschvartz and M. Naeye.

#### SUMMARY

When ions move in a liquid they exert a force on this liquid. By measuring this force and the current I one can calculate the mobility  $\mu$ . The current I emitted by a tungsten tip in liquid helium is space charge limited. The mobility  $\mu$ is found to be independent of the field between 7 and 27 kV/cm.

In a former paper a new method for the measurement of ionic mobilities in high electric fields in isolating liquids has been described <sup>(1)</sup>. The principle is as follows. Two electrodes are mounted in a glass tube immersed in the liquid. Both ends of the tube are open. Ions are injected into the liquid by cold emission. The electric field exerts a force on the ions and lifts the liquid column in the tube. By measuring simultaneously the change of the liquid level  $\Delta h$  and the electric current I the mobility  $\mu$  can be calculated. With this method some measurements of the mobility of negative ions in liquid helium at 4,2 K were performed.

Since then, the equipment has been improved. The measurements were extended to temperatures below the lambda point. With the improved apparatus it turned out that  $\Delta h$  is proportional to the square of the applied voltage, indicating that the current is space charge limited. Thus:  $\Delta h = aV^2$ .

Figure 1 shows two series of  $\Delta h$  as a function of V at 4,2 K. Series I was performed at the beginning of a day of experiments, series II at the end. The line drawn is the best parabolic fit to the points. The value of the constant a is a = 0,068 mm/(kV)<sup>2</sup>.

If one calculates a from a linear model of infinite cathode and anode separated by a distance b one finds

$$a = 9 \varepsilon_0 \varepsilon_r / 8 g \rho_m b^2 \tag{1}$$

where g is the gravity constant,  $\varepsilon_0$  is the permittivity of the vacuum,  $\varepsilon_r$  and  $\rho_m$  are the relative dielectric constant and the mass density of the liquid helium respectively. In the derivation of (1) use has been made of the formula derived in reference <sup>(1)</sup>:

$$\mu g \rho_{m} S \Delta h = I b$$
 (2)

Here S is the internal cross-section of the tube in which the electrodes were mounted.

Inserting the numerical values in (1) (b = 10 mm) one finds:  $a = 0,078 \text{ mm/kV}^2$ .

The model used is an extreme one and gives a too high value of a. Another extreme model is that of a spherical cathode with negligible radius mounted in the centre of a spherical anode with radius b. In this case one finds for the quantity a the formula (1) in which the factor 9 has been changed to 3. The geometry of the apparatus is between both extremes but is probably nearer to the first one. Thus, the experimental value found is a satisfactory one.

In figure 2a and 2b typical measurements of I and  $\Delta h$  as a function of T at two tensions V between the electrodes are shown (b = 7,5 mm). From these measurements the mobility  $\mu$  was calculated with equation (2); the results are plotted in fig. 2c.

When increasing the temperature at constant applied voltages from T < T<sub> $\lambda$ </sub> to T > T<sub> $\lambda$ </sub> in most experiments either a breakdown occurred after passing the lambda point or the current I, and thus also  $\Delta$ h, dropped to zero. The last happened at lower voltages. It seems to indicate that a cap of solid helium is more easily formed on the cathode point just above the lambda point than below it.



Figure 1. Variation  $\Delta h$  as a function of V. Series I at the beginning of the day of experiments, II at the end.



Figure 3. All the measurements of the mobility below the lambda point as a function of the temperature.



Figure 2. (a) and (b) typical measurements of the ionic current I and the variation  $\Delta h$  of the liquid helium level as a function of temperature. (c) the mobility as a function of the temperature calculated from (a) and (b) with formula (2).



In figure 3 all measurements of  $\mu$  at temperatures below  $T_{\lambda}$  and two above  $T_{\lambda}$  have been represented in the same way as done by Scaramuzzi<sup>(2)</sup>. The values found are about the same as those of Scaramuzzi despite the fact that the electric fields he used were more than two orders of magnitude lower than ours.

Williams (3) measured the mobility  $\mu$  of helium ions in high fields, ranging from 4 kV/cm to 130 kV/cm at 4,2 K and to 50 kV/cm for temperatures below  $\boldsymbol{T}_{\boldsymbol{\lambda}}$  . He found a decrease of  $\boldsymbol{\mu}$  with increasing field strength E. Our measurements indicate that u is independent of E. It is true that in our experiments E in the cell was not constant, decreasing from a very high value at the tungsten tip to a much lower value at the anode. If µ would depend strongly on E, a change of the voltage between anode and cathode would result in a change of the ratio  $I/\Delta h$ . In a number of experiments the total voltage was changed in steps up to a factor 4. The value of  $I/\Delta h$  proved to be independent of V. For temperatures below  $T_{\lambda}$  our results agree with the values found by Williams at a field strength of 23 kV/cm.

Due to a numerical error in our calculations, we mentioned in our former paper <sup>(1)</sup> the value  $\mu = 0,025 \text{ cm}^2$ :Vs at 4,2 K. This should be  $\mu = 0,018$ .

With the improved apparatus we find now  $\mu = 0,0160 \pm 0,0009$  as an average from seven series of measurements. This is lower than the value of Williams ( $\mu = 0,024 \text{ cm}^2/\text{Vs}$ ) at this temperature and at 23 and at 12 kV/cm.

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## APPENDIX I

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# MODIFIED HELIUM DEWAR

J.M. Goldschvartz.

Mendelssohn (1) remarked about Dewar's invention: '.... his famous vacuum vessel in a form so perfect that it has remained unchanged to this day'. Nevertheless, some slight modifications have been carried out in order to ameliorate or to simplify the use, while increasing the efficiency of a dewar vessel for low temperature techniques. For example the one made by Lonsdale and Smith (2) who introduced a spiral-exit tube in the bottom of the vacuum chamber of a liquid nitrogen or oxygen dewar, and the one in which the upper part of a helium vessel is single walled <sup>(3)</sup>, permitting a better contact with the external cooling bath and reducing the heat leak and the rate of diffusion of helium gas into the vacuum chamber. These two modifications changed the structure of a dewar.

This note deals with a succesful attempt to fill a helium dewar in a more rational way, especially when it contains a large apparatus for taking measurements at liquid helium temperatures.

Figure 1 shows in detail how the dewar is made. The tube A, of the same diameter of the external tube of the transfer siphon, is an



extension of the vacuum chamber of the Dewar, and the inner tube B, of the same diameter of the inner tube of the transfer siphon, leads the liquid helium to the aperture C in the bottom of the helium space of the dewar. This means that the dewar itself is an extension to 20 erratum the transfer siphon. The coupling between the siphon and the dewar is made by means of a protecting Teflon piece D and a normal vacuum coupling tube E with O-rings. The dewar is made of Thermax glass, the space between walls

is a high vacuum chamber and the slit or window on the silvered surface reaches only the point F, which means that the bottom of the dewar is completely covered with the silver shield.

This Dewar was compared with a conventional one used in this laboratory, with the same single walled part, containing the same apparatus, with the same cap and in the same external conditions, but with the transfer siphon through the cap and the inside helium vessel 5 mm smaller in diameter.

The behaviour of this dewar was excellent. The rates of filling in both cases are approximately the same in spite of the heat leak due to the external coupling with the transfer siphon and in spite of its larger volume. Closing the siphon valve, the  $\lambda$ -point was reached without any difficulty. The refilling of the dewar, which as far as the liquid helium consumption is concerned must always be avoided, was found to be better in the conventional type.

Besides the advantages mentioned above, the following points may be noted: (1) the fact that the siphon does not go through the neck of the vessel could eventually allow modifications of the neck and a simplified introduction of radiation baffles - which practice was used by Lyman, Proctor, and Scurlock <sup>(4)</sup> to reduce the evaporation rate of liquid helium, (2) there is more room in the dewar cap for electrical leads and mechanical devices, and the space beneath the cap is free of interferences between them and the transfer siphon, (3) the height at which the dewar can be filled is not limited by the depth of the siphon inside the vessel, (4) in time it is also possible to transfer the liquid helium from the dewar into another container, and (5) even in the case where one wishes to fill the vessel in the conventional way, this system is a very good one for the pre-cooling.

The author is grateful to Professor B.S. Blaisse for encouraging him in this work. Thanks also are due to Mr. H. Verbeek, head of the glass workshop, who materialized the idea of this dewar and to Mr. A. Kollen for his advice and assistance during the experiments.

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  (1966) 331; 'The Quest for Absolute Zero',
  p. 55 (World University Library, Wedenfeld and Nicolson, London, 1966).
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## APPENDIX II

## GENERAL DISCUSSION

# On Chapter I. Liquid Helium.

Obviously, it is a very incomplete survey. It is only designed to supply some fundamental and elementary information which can be a link between the other chapters.

## On Chapter II. Superleaks.

The search for new superleaks is still on our agenda for the near future and we think that the following materials will be interesting to check, namely: porous ceramics, silica gel, zeolites and ferrites.

The methods described for the determination of the onset points of superleaks must be improved in order to obtain a much higher accuracy. This accuracy depends upon the complete automation of the system and on the calibration of the low temperature thermometers. Using the fact we have already mentioned in paragraph 2.8 that the value of the resistance of a carbon thermometer undergoes a sudden change as soon as the superfluid reaches it, a completely automatic method has been designed. With it it will be possible to read the pressure without time delay. This method is also applicable to calibrate the thermometers using the following devices: a bridge, an ohm-converter and an automatic manometer, such as that of Texas Instruments. On Chapter III. Electrical Breakdown of liquid helium.

Measurements of the electrical breakdown which are within a reproducibility of 20 to 30% are considered as good, as was mentioned in paragraph 3.5. Although this is the reproducibility of our last two years of experiments, we think that this reproducibility can and must be improved. This means that a good deal of new experiments should be performed improving the instrumentation and the control upon some parameters, mainly the purity of the liquid helium. Also other conditions should be changed for future experiments, namely: the material and the shape of the electrodes; measurements should be done at bigger values of the gap distance; as a consequence, one should have available a power supply, for higher tensions up to one hundred kilovolts.

Another important improvement would be the recording of the breakdown voltage and currents of what we still call pre-breakdowns. All these changes and improvements will result in better experiments which will not only supply a better knowledge of the value of the electrical breakdown of liquid helium for the technological applications, but will also have an influence on the possibility of elaborating a theory of this phenomenon.

As far as the theories are concerned, here something will be added to what was said in paragraph 3.3. To begin with, Kok's theory is based only on the presence of impurities. That is, it is not a theory of the electrical breakdown of a liquid dielectric. One could say that it is an explanation of the electrical breakdown of any kind of liquid containing impurities.

In the case of the thermal theory developed by Sharbough and Watson it was said that for liquid He II the formation of bubbles is only possible with a very particular type of electrodes and this is not the case for the type of electrodes used to measure the electrical breakdown of liquid helium. Moreover, if Sharbough and Watson are right one could expect a drastic change in the value of the electrical breakdown strength when decreasing the temperature from above to below the  $\lambda$ -point. The heat conductance of liquid helium namely increases by a factor of 10<sup>7</sup> when the temperature of the liquid is decreased from 2,2 to 1,8 K.

Swan's theory seems to be the most plausible as a physical theory of the electrical breakdown of liquid helium. As was stated in paragraph 3.3 we have checked this theory with the results of the fifteen sets of experiments plotted on a double logarithmic scale in figure 1 a,b; see also Table 1, paragraph 3.8. In fact, the lines of this represen-

tation obey the equation  $\ln E_{o} = a \ln d + c$ , i.e., a straight line. On the other hand, Swan's equation is an exponential which represented in the same way gives nearly straight lines in a not too limited region. However, the values of the constants of Swan's equation obtained from our experiments differ very much from Swan's values for liquid Argon. In the derivation of his equations Swan supposed the mobility  $\mu_{\pm}$ of the positive ions to be very much smaller than  $\mu_{-}$  of the negative ions. This applies to liquid Argon but not to liquid helium. For this liquid  $\mu_{\perp}/\mu_{\perp} = 4 + 1$  according to Williams (Chapter IV, reference 10), while our own measurements give 2,4 at boiling point and 1,4 at 2 K (not yet published). Swan's theory should be extended.

Furthermore, we have applied computer techniques to four new sets of experiments made at boiling point. We compared them with those mentioned above <sup>(1)</sup>. Applying regression analysis the computer using a special program searches for a curve which fits best to the experimental values. Six types of curves are tried, namely:

y=A+Bx, y=A exp.(Bx), y=Ax<sup>B</sup>, y=A+ $\frac{B}{x}$ , y= $\frac{1}{A+Bx}$ , y= $\frac{x}{Ax+B}$ . Zie erratum

In figure AII.1 the curve m obeying this equation is the average curve calculated from the four sets of experiments. As compared with the average



Figure AII.1.

curve A (Lancaster, 1970) of figure 2 of paragraph 3.8, one can see that this new average curve lies in the same region. This means that a relatively good reproducibility was obtained. The relative differences in the dielectric strength value for different gap distances are: for 100 µm of the order of 12%, for 55 µm of 10% and for 20 µm of 10%.

Although it was not mentioned in paragraph 3.3

we shall comment here on another theory of the electrical conduction and breakdown of liquid dielectrics by Goodwin and Macfadyen <sup>(2)</sup>. They, as Swan, developed a theory for a system liquid-electrodes, i.e., based on an intrinsic concept of conduction and breakdown, as was done in paragraph 3.1. They considered also the cold emission according to the formula of Fowler and Nordheim, but a bigger ionization than in Swan's case. This theory was analysed by two co-workers (3) of the Low Temperature Group and they found a numerical error in the mathematical development. As a consequence of this error Goodwin and MacFadven gave a wrong sign to a square root in one of their formulas. In reality this solution of their equations without the numerical error has no physical sense. The right solution when adopted to the experiments gives a very large value for the ionization reinforcement of the current ( $\alpha$ -mechanism; factor about 10<sup>9</sup>). This large value is not in agreement with the fact that it has not been possible to measure electric currents in pure helium before breakdown takes place. The sensitivity of these measurements was about  $10^{-12}$  A. Because of these reasons the theory of Goodwin and MacFadyen was not used for comparison with our experimental data.

As it was pointed out in paragraph 3.1 and 3.7, there are only few groups working on the electrical breakdown of liquid helium. This signifies that there would be still time to fix

some kind of international standards in order to be able to talk, when results are compared and discussed, about experiments performed under the same conditions. A proposal was made at the meeting on "Dielectrics at Very Low Temperatures" (London 7th April, 1971) and now we are engaged on working it out.

On Chapter IV. Mobilities of Ions in Liquid Helium.

A new method of measuring ionic mobilities in liquid helium in high electric fields is described. Although rather few experiments were performed by other investigators in the same range of temperatures and tensions to compare with, we have obtained enough evidence that this method is satisfactory and simple. For the time being the results obtained coincide with existing results which have been measured with classical methods. Nevertheless, we still have experimental difficulties, particularly above the  $\lambda$ point with the generation of bubbles in the electrode space. These bubbles introduce a certain uncertainty in the readings of the variation  $\Delta h$  of the helium column level inside the cell. If the mounting of the electrodes is such that the sharp tungsten tip is below the flat electrode, the current of the ions is going upwards as in fig. 1, paragraph 4.3. As a consequence the level inside the electrode tube rises. The level is lifted more if there are bubbles. In this case one measures a too high value of  $\Delta h$ . When the mounting is the reverse (tungsten tip above the flat electrode) the level inside the tube is

depressed. Now the presence of bubbles diminishes the absolute value of  $\Delta h$ . Hence measurements are made with both mountings. It turned out that above the  $\lambda$ -point a difference exists in both values of  $\Delta h$ ; therefore in this case the average is taken. Below the  $\lambda$ -point the two values of  $\Delta h$  coincide. Then there are no bubbles or only very few if the electric current and thus the heat dissipation is not too high. The absence of bubbles in this case is due to the enormously high heat conduction in helium II.

Another important disadvantage is that frequently an electrical breakdown takes place between the electrodes, destroying the sharp tip of the wolfram pin. These breakdowns take place only above the lambda point. The occurrence is independent of whether the temperature is diminishing or increasing. They also occur at low values of the applied voltage; for instance at 5 or 6 kV. These breakdowns never take place below the lambda point save for extremely high fields, e.g. for an applied voltage of 20 kV. As a matter of fact, these low voltage breakdowns cannot be ascribed to impurities because they would also be present below the lambda point. Here Sharbough and Watson's theory of the electrical breakdown (see paragraph 3.3) based on the formation of bubbles applies.

In the near future our main task will be

the measurement of mobilities of both positive and negative ions in the range from 4,2 K to the lowest temperature obtainable with our equipment. The generation of either positive or negative ions is made as follows <sup>(4,5)</sup>: an electric field of the proper sign can draw positive or negative ions out of a microscopic bubble which is formed around the sharp tip of the thin wolfram electrode. In order to make more accurate measurements some improvements in the equipment are being made: first guard-rings around the two electrodes to improve the uniformity of the field, second, a versatile holder at which the thin electrode with the sharp tip will be fixed is being designed; this holder must be able to be used (a) to prepare the tip, (b) to apply it to the electron microscope to observe the tip, and (c) to use it in the equipment for the experiments and, third, the heat leak in the cryogenic system will be diminished to obtain a more steady pressure, i.e., a very definite temperature.

On the other hand, as has been suggested in paragraph 4.3, measurements of the ionic mobilities in rotating helium are planned for the distant future. This is based on the fact that in rotating He II the negative ions are trapped in the vortices (see paragraph 4.1). Moreover, we want to extend the measurements to much lower temperatures (about 0,6 K).

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## SUMMARY

This thesis is based on the papers published in the last five years on different subjects about a central theme: liquid helium four.

In Chapter one a breef resumé of liquid helium four is given. The purpose of this chapter is to serve as a link relating the next three chapters. This resumé contains in its last paragraph an up-to-date list of books.

Chapter two is devoted to superleaks, a device which can play an important rôle in the future technological applications of superfluidity which are indicated in this chapter. Some new materials for superleaks are described, namely: a porous silicon carbide and two natural materials: an aluminium silicate from South Africa called wonderstone and a magnesium silicate from Rabenwald in Austria called talc-stone. A new method of measuring the onset point of a superleak is described as well.

Chapter three deals with the electrical breakdown strength of liquid helium. This quantity became important not only from a scientific point of view but also for the new technology of superconductivity in which liquid helium is the means of cooling down the materials as well as the insulating dielectric. The apparatus and the total instrumentation for measuring the electrical breakdown are described. Five published
papers on this subject containing all the measurements performed during the last eight years are included. Existing theories on the electrical breakdown of liquid dielectrics are discussed as well.

Chapter four contains the description of a new method of measuring ionic mobilities in liquid dielectrics in high electric fields. Also it contains the description of the classical method of measuring ionic mobilities. Two published papers with our own results are included.

Appendix one contains the description of a modified helium Dewar. The modification relates to the way the Dewar is filled with the liquid helium. This gives some advantages with respect to the conventional way. A tube which is in the vacuum chamber joins the bottom of the inner container of the Dewar to the external siphon.

Appendix two is a general discussion on each one of the four chapters mentioned containing criticisms and plans for future work on each of the subjects dealt with.

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## SAMENVATTING

Dit proefschrift is gebaseerd op de artikelen die de laatste vijf jaar zijn gepubliceerd over verschillende onderwerpen rond een centraal thema: vloeibaar helium vier.

In het eerste hoofdstuk wordt een kort overzicht van de eigenschappen van vloeibaar helium vier gegeven. Dit hoofdstuk laat de samenhang zien tussen de drie hoofdstukken die erop volgen. Dit overzicht bevat in zijn laatste paragraaf een uitgebreide bibliografie.

Het tweede hoofdstuk is gewijd aan superlekken, een hulpmiddel dat een belangrijke rol kan spelen in de toekomstige technologische toepassingen van superfluïditeit die in dit hoofdstuk zijn aangegeven. Er worden enige nieuwe materialen voor superlekken beschreven, namelijk: een poreus silicium carbide en twee natuurlijke materialen: een aluminium silicaat uit Zuid Afrika dat "wonderstone" wordt genoemd, en "talc-stone", een magnesium silicaat uit Rabenwald in Oostenrijk. Er wordt tevens een nieuwe methode beschreven om het "onset point" van een superlek te bepalen.

Het derde hoofdstuk gaat over de electrische doorslag van vloeibaar helium. De veldsterkte waarbij de doorslag optreedt is niet slechts belangrijk uit wetenschappelijk oogpunt, maar ook voor de nieuwe technologie van de supergeleiding, waarbij vloeibaar helium het middel is om de materialen te koelen en tegelijkertijd een deel vormt van de electrische isolatie. Het apparaat en de volledige experimentele opzet om de electrische doorslag te meten, wordt beschreven. In dit hoofdstuk zijn vijf gepubliceerde artikelen verwerkt, die alle metingen over dit onderwerp bevatten die gedurende de laatste acht jaar zijn uitgevoerd. Bestaande theorieën over de electrische doorslag van vloeibare diëlectrica worden eveneens besproken.

Hoofdstuk vier bevat de beschrijving van een nieuwe methode om de beweeglijkheid van ionen te meten in vloeibare diëlectrica in sterke electrische velden. Tevens wordt de klassieke methode om de beweeglijkheid van ionen te bepalen, erin beschreven. Twee gepubliceerde artikelen met eigen resultaten maken er ook deel van uit.

Appendix één bevat de beschrijving van een gemodificeerde helium dewar. De modificatie heeft betrekking op de wijze waarop de dewar met vloeibaar helium wordt gevuld. Deze biedt enige voordelen ten opzichte van de gebruikelijke manier. Een buis in de vacuümruimte verbindt de bodem van het binnenglas met de vulhevel.

Appendix twee vormt een discussie van ieder der vier genoemde hoofdstukken; het bevat enige critische beschouwingen en plannen voor toekomstig werk aan elk der behandelde onderwerpen.

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## STELLINGEN

1 The following experiments could provide a relevant information on the pre-breakdown phenomena in liquid He: a) the change of the temperature in points as near as possible to the gap; b) the optical observation and recording of the interelectrode space as soon as the voltage is applied and during the raising of the field till the breakdown takes place, and c) the application of a photomultiplier for the detection of the pre-breakdowns.

This thesis, Chapter III and Appendix II.

2 The electrical breakdown of solid He has never been measured. Modifying the high pressure cell of the existing equipment for solid He in this laboratory, this quantity could be measured as well as the electrical breakdown of liquid He as a function of the density.

Solid He equipment in Vos, J.E., Thesis, Delft, 1968.

- 3 In the study of the electric charging of a capacitor by friction with rotating liquid helium, a discontinuity in the charge of the capacitor could be expected at the  $\lambda$ -point.
- 4 In order to keep constant the temperature of a cryostate below the  $\lambda$ -point, a manostat can be made, the Adritron, which has the advantage that no gas reference pressure is necessary and which can be built in a mercury manometer. This is a mechano-electric instrument with an electronic control unit.
- 5 Since the liquifaction of He was achieved, there is an important problem in Low Temperature physics: the creeping up of a He II film on the walls of the dewar. It seems possible to eliminate this He II film by means of a trap made on the wall.
- 6 Träuble and Essmann using electron optics and the so-called decoration technique, visualized the magnetic structure of superconductors. Quantized vortex lines in rotating He II could be visualized as well using the fact that negative ions are trapped in these vortex lines. Applying an electron lens to the accelerated emergent electrons and with the aid of photographic techniques, the structure and distribution of vortices could be studied.

Träuble, H.T. and Essmann, V., Phys. Stat. Sol. 18 (1966) 813; Douglass, R.L., Phys. Letters 28A (1969) 560; Schoepe, W. and Dransfeld, K., Phys. Letters 29A (1969) 165. 7 Fermi wrote once: "One can go back to the books on method (I doubt whether many physicists actually do this) where it will be learned that one must take experimental data, collect experimental data, organize experimental data, begin to make a working hypothesis, try to correlate and so on, until eventually a pattern springs to life and one has only to pick out the results. Perhaps the traditional scientific method of the textbooks may be the best guide, in the lack of anything better".

About that Segre said: "I believe that he always had a working hypothesis ..." and later on "... he did not want to miss great discoveries by not being bold enough".

Any physicist always had to ask himself: which is the way to be followed?

> Fermi, E., Physics Today <u>5</u> (1952) 6; Emilio Segre, "Enrico Fermi, Physicist", The University of Chicago Press, Chicago and London, 1970.

8 Scientists in general and, due to the consequences of the development of nuclear weapons, physicists, in particular, must share responsibility about what is happening in the whole world. Furthermore, any citizen and any human being, cannot be indifferent to millions of people which are suffering either hunger by economical aggression, or oppression by racial prepotency, or violence from the very powerful. For violence can only generate violence.

> Max Born, Physics in my generation, Springer Verlag New York Inc., Heidelberg Science Library, 1969 and The Autobiography of Bertrand Russell, Volumen III, George Allen and Urwin Ltd., London, 1969.

9 Very often one hears from dilettanti that there are intellectual differences between artists and scientists. Doubtless, it seems that J.B. Bach or I. Strawinsky, if they had been trained, could perfectly well solve differential equations or plan some complicated physical experiment. On the other hand, it is not possible to doubt that Ch. Huygens or A. Einstein, if they had been trained, could compose music or write a novel as transcendent as their scientific works. Fortunately, none of them changed their habits.