IONIZATION BY ALPHA-PARTICLES IN LIQUIDS AT LOW TEMPERATURES



A. N. GERRITSEN

DIBLIOTHEEK GORLAEUS LABORATORIA Postbus 9502 2300 RA LEIDEN Tel.: 071 - 527 43 66 / 67





IONIZATION BY ALPHA-PARTICLES IN LIQUIDS AT LOW TEMPERATURES

kast dissertaties

N.V. VAN DE GARDE & CO'S DRUKKERIJ, ZALTBOMMEL

IONIZATION BY ALPHA-PARTICLES IN LIQUIDS AT LOW TEMPERATURES

PROEFSCHRIFT

TER VERKRIJGING VAN DE GRAAD VAN DOCTOR IN DE WIS-EN NATUURKUNDE AAN DE RIJKSUNIVER-SITEIT TE LEIDEN, OP GEZAG VAN DE RECTOR MAGNIFICUS MR J. C. VAN OVEN, HOOGLERAAR IN DE FACULTEIT DER RECHTSGELEERDHEID, VOOR DE FACULTEIT DER WIS- EN NATUURKUNDE TE VERDEDIGEN OP WOENSDAG 30 JUNI 1948 DES NAMIDDAGS TE 2 UUR

DOOR

ALEXANDER NICOLAAS GERRITSEN GEBOREN TE 'S-GRAVENHAGE



's-gravenhage MARTINUS NIJHOFF 1948 Promotor: PROF. DR. H. A. KRAMERS.



Opgedragen aan mijn moeder aan mijn gezin en aan de nagedachtenis van mijn vader



CONTENTS

IN	TRODUCTION	VIII
1.	MEASUREMENTS IN LIQUID NITROGEN AND LIQUID HYDROGEN	
	Summary. 1.1. Introduction 1.2. Some remarks on the theory of column ionization in gases and liquids 1.3. The ionization chamber. 1.4. The method of measuring the ionization currents. 1.5. Some remarks concerning the experiments. 1.6. Results of the measurements in liquid nitrogen. 1.7. Results of the measurements in liquid hydrogen	1 2 8 11 14 15
	1.8. Conclusions	21
2.	MEASUREMENTS IN LIQUID HELIUM AND LICUID ARGON Summary. 2.1. Introduction 2.2. The results in liquid helium. 2.3. The results in liquid argon 2.4. The experiments in liquid helium at different temperatures 2.5. A comparison between the results and their interpretation for the four liquids	26 27 27 28 32 34 38 44
3.	THE STOPPING POWER OF A METAL FOR POLONIUM ALPHA- PARTICLES.	
San	Summary. 3.1. Introduction 3.2. The experimental method. 3.3. The results for tin and aluminium References IENVATTING	45 45 47 49 54 55

the second

INTRODUCTION

From some experiments on X-ray ionization in liquid helium followed an ionization current-voltage characteristic, which showed more analogy with a characteristic in a gas than with that in a liquid. This and the fact that with X-rays it was hard to determine the saturation current, were the reasons to start a new series of experiments on ionization in liquids at low temperatures. They were carried out with the α -particles of polonium as an ionizer in liquid nitrogen, argon, hydrogen and helium. Some series were carried out with a metal foil over the polonium, in order to reduce its intensity of radiation.

In the *first chapter* we describe the ionization chamber and the simple measuring method. Next the general outlines of J a f f é's solution of the problem of column ionization and a modified solution of K r a m e r s, more adapted to experiments in liquids at low temperatures, precede a report on the experimental results in nitrogen and hydrogen. A comparison between the measured currents and those calculated by both theories are given in the last section of this chapter, and it seems that the K r a m e r s solution and the results agree.

In the *second chapter* we describe the experiments in liquid helium I (and helium II) and in liquid argon. In these cases both theoretical solutions fail.

The comparatively large values of the ratios of the measured currents and their saturation values in the inert liquids argon and helium can be interpreted by a coëfficient of recombination, which is small compared with its value in nitrogen and hydrogen.

A model of feather ionization is introduced, from which a possible explanation for the failure of the Kramers solution follows and which may give an estimate of the correction that must be applied to this solution.

In the *third chapter* we report a series of measurements on the stopping power of aluminium and tin foils (the latter in their normal and super conductive state) which is, in contradiction with V o n W e i s z ä c k e r's prediction, independent of temperature.

CHAPTER I

MEASUREMENTS IN LIQUID NITROGEN AND LIQUID HYDROGEN

Summary

After a review of the theories of J affé and K r a m e r s concerning column ionization in liquids at low temperatures a simple method for measuring the ionization currents is described. The results of several series of measurements in liquid nitrogen and liquid hydrogen are given. It seems probable that in nitrogen the linear dependence of the current on the field is, in contradiction to J affé's theory, due to the corrective influence of the diffusion on the distribution of the ions in the liquid. K r a m e r s's theory proves to be in qualitative agreement with the results.

1.1. Introduction. The phenomena of ionization and the relation between an electric field and an ionization current in a gas have been studied by many authors. The curve that describes the connection between ionization current and field strength has been called the saturation curve. It is known that the form of this curve depends on the way in which the ionization is established and that besides recombination also the diffusion of the ions play a part. In the case of ionization by α -particles, the so-called column ionization, the theory of the saturation curve offered many difficulties and many details must be taken into account. In a following section we shall mention the formulae of G. J a f f é for column ionization.

Not so many studies and experiments have been devoted to the ionization of non-electrolyte liquids and solids. Besides the measurements of J a f f é¹) and other authors ²) in liquids at room temperature (hexane, tetrachloric carbon and some other liquids) and of Miss F o l m e r ³) in paraffin, the only known experiments

_ 1 _

in liquids at temperatures lower than room temperature are those of $M c L e n n a n^4$) in liquid air. The ion-densities in the columns and the magnitude of the coefficients of ion-diffusion and recombination are different in liquids and solids from those in gases, so that the shape of the saturation curve will be quite another.

The facts that liquefied gases are in general very pure and that a liquid has a high density, led Prof. dr W. J. d e H a a s to suggest an investigation in liquefied gases in connection with a possible search to the use of these liquids as a detecting medium for cosmic rays. Moreover such investigations in liquid helium would perhaps be worth while in connection with the properties of the helium below the λ -point.

Our first experiments with X-rays ⁵) showed not only that ionization of the liquefied gases nitrogen and helium was possible, but also that the saturation curves in these liquids showed differences that required a closer investigation. For this investigation, however, it is desirable to know the values of absolute saturation (or in other words the specific ionization of the X-rays in the liquids) and this could not be determined because of the unknown absorption of the X-rays before entering the ionization chamber and the unknown influence of the secundary electrons set free from the solids in the apparatus.

An ionizer for which such difficulties do not exist is polonium (RaF). We have investigated the saturation curve in four liquids at low temperatures (nitrogen, hydrogen, argon and helium), with the α -rays of polonium as ionizer.

1.2. Some remarks on the theory of column ionization in gases and liquids. We shall give some theoretical considerations and report the experiments and their results later on in this and the next chapter.

1.21. J a i j i e's theory of column ionization. Jaffé⁶) started his considerations with L a n g e v i n's assumption of column ionization, that is that all the ions formed by the passing α -particle are collected in a cylinder around the path of this particle. The initial distribution is modified by the recombination between the ions in the cylinder (thus the ion density is lowered), by the diffusion of the ions (thus the cylinder is broadened) and by the electric field (that separates the ions of different sign from each other). One is interested

in the fraction of the initial number of ions, which per unit length of the column in the course of time is set free by the field and can reach the electrodes.

Introducing an initial linear density N_0 of ions along the axis of the cylinder (z-axis) and supposing a gaussian distribution of the number n_0 of ions per cm³:

$$n_0 = \frac{N_0}{\pi b^2} e^{-\frac{x^2 + y^2}{b^2}},\tag{1}$$

where b is a measure for the radius of the cylinder and assuming the mobility of the ions of both signs to be the same, the solution of the equation of motion:

$$\frac{\partial n_{\pm}}{\partial t} = \mp v F \sin \varphi \frac{\partial n_{\pm}}{\partial x} + D \Delta n_{\pm} - \alpha n_{+} n_{-}$$
(2)

is required with $n_{+} = n_{-} = n_{0}$ at t = 0. In this equation are introduced the symbols $v_{+} = v_{-} = v$ for the velocity per unit of fieldstrength of an ion of charge + e and - e, respectively; D for the coefficient of ion diffusion and α for the coefficient of recombination. The field F was chosen in the x, z-plane, the angle between its direction and the z-axis being φ .

It seems impossible to establish the exact mathematical solution of (2) and one has to apply some approximative procedure. J a f f é derives first the exact solution of (2) in the case $\alpha = 0$; in this solution, which is easily obtained n_+ and n_- contain both N_0 as a constant factor. He then takes recombination into account by replacing N_0 by a function N(t) of time, which is assumed to satisfy the equation which follows from (2), by integrating both sides over x and y. By this procedure the effect of recombination is overestimated in regions where n_+ and n_- are comperatively small and underestimated where they are comperatively large. Denoting by ϕ the ratio of the number of ions set free per unit time to that produced per unit time, J a f f é finds:

with

VE

where $H_0^{(1)}$ is the Hankel function of the first kind of order zero. Using the expressions v = eu, D = kTu, and L ang evin's

well known approximate expression 7) $\alpha = 8\pi u e^2$ (where u is the mobility of an ion) *), we get

$$\dot{p} = \frac{1}{1 + \frac{e^2 N_0}{kT} f(\xi)}, \quad \sqrt{\xi} = \frac{b \, e F \sin \varphi}{2kT}.$$
 (3b)

The result is independent of the mobility; this is not astonishing and should also hold for the exact solution of (2) since a change in the mobility only means a change in the rate at which ions in the column are liberated and not in their final number.

Since for large values of ξ (or large values of F) we have

$$f(\xi) = \sqrt{\frac{\pi}{2\xi}} \left(1 - \frac{1}{8\xi} + \frac{9}{128\xi^2} \dots \right),$$

we see that for sufficient large values of ξ^{*} the solution (3b) is transformed into:

$$p_{1} = \frac{1}{1 + \frac{\sqrt{2\pi} e N_{0}}{b} \frac{1}{F \sin \varphi}},$$
(4)

which changes into a linear function of current and field:

$$p_1 = \frac{b}{\sqrt{2\pi} e N_0} F \overline{\sin \varphi} \tag{5a}$$

in an F-domain in which the second term in the denominator is large compared to unity. For still larger values of F eq. (4) becomes:

$$\phi_2 = 1 - \sqrt{2\pi} \frac{eN_0}{b} \frac{1}{F \sin \varphi}.$$
(5b)

In these expressions $\sin \varphi$ is an average appropriate to the distribution of the directions of the α -particles in the experiment under consideration.

The solutions given by J a f f é are in general in agreement with experimental results in various gases at normal conditions of density and temperature. For gases at high densities modifications seem necessary which pertain to details of the recombination of the ions in the column. For a complete review of these theories and their

^{*)} In this formula we omitted the dielectric constant, which differs only a few percents from unity in the liquids under consideration.

applications we may refer tot the thesis of P. H. Clay⁹) and the original communications.

The accuracy of our measurements is too low to decide whether modifications of this kind are essential, but quite apart from this our experimental results led to the conclusion that a modification of the theory of J a f f é is required for quite a different reason.

1.22. Kramers's modification of the theory of Jaffé. Starting from the results of the measurements that will be referred to later on, Kramers suggested another way of solving the equation of motion (2) ¹⁰). He pointed out that Jaffé's procedure will no longer be justified in cases where the influence of the recombination largely outweighs the effects of diffusion and field strength. Such cases will precisely present themselves in liquids, where the initial ion densities are much larger than in gases, in particular in liquified gases at low temperatures where the diffusion is small due to the low temperatures. The following consideration will clarify this point.

With the given initial distribution (1) it is possible to deduce from equation (2) the ratio of the initial contributions of the several terms relative to each other. By measuring the time in units of 1/u and by replacing D and α by their expressions in terms of the fundamental quantities u, e and T eq. (2) becomes:

$$\frac{\partial n_{\pm}}{\partial t} = \mp eF \sin \varphi \, \frac{\partial n_{\pm}}{\partial x} + kT \varDelta n_{\pm} - 8\pi e^2 n_{+} n_{-}, \tag{6}$$

or in symbolic form:

$$\frac{\partial n_{\pm}}{\partial t} = [f] + [d] + [r]. \tag{7}$$

At t = 0 the order of magnitudes of these terms are:

$$[f]_0 \approx \frac{eFN_0}{\pi b^3}, \ [d]_0 \approx \frac{kTN_0}{\pi b^4}, \ [r]_0 \approx \frac{8\pi e^2 N_0^2}{\pi^2 b^4},$$
(8)

from which it follows that their ratios are:

$$\left[\frac{d}{f}\right]_{0} \approx \frac{kT}{beF}, \quad \left[\frac{f}{r}\right]_{0} \approx \frac{Fb}{8eN_{0}} \text{ and } \left[\frac{d}{r}\right]_{0} \approx \frac{kT}{8e^{2}N_{0}}.$$
 (9)

The ratios (9) will be different for gases and liquids, and will also depend on temperature. Taking for the value of b a number of the

order of magnitude 10^{-3} cm for gases under normal conditions ¹¹) and assuming b to be inversely proportional to the density of the medium, its magnitude in liquids will be of the order of 10^{-6} cm. As the number of ions produced by an α -particle in a gas is of the order 10^4 ions per cm ¹²) this number increases in liquids to 10^7 ions per cm.

Hence the numbers (9) will be of the order of magnitude:

$$\left[\frac{d}{f}\right]_{0} \approx 3 \times 10^{-4} \frac{T}{F}, \quad \left[\frac{f}{r}\right]_{0} \approx 3 \times 10F, \quad \left[\frac{d}{r}\right]_{0} \approx 8 \times 10^{-3}T \quad (10a)$$

for gases and

$$\left[\frac{d}{f}\right]_{0} \approx 3 \times 10^{-1} \frac{T}{F}, \quad \left[\frac{f}{r}\right]_{0} \approx 3 \times 10^{-5} F \text{ and } \left[\frac{d}{r}\right]_{0} \approx 8 \times 10^{-6} T \quad (10b)$$

for liquids.

These numbers show that:

a. As soon as $3 \times 10^{-1} T \ll F$ (F given in e.s.u.) the influence of the diffusion is in liquids small compared with that of the field. This will be so for fields larger than 27 kV/cm, 7.2 kV/cm, 1.8 kV/cm and 0.4 kV/cm at temperatures of 300 °K, 80 °K, 20 °K and 4 °K respectively. In gases, however, the diffusion at 300 °K has a negligible influence compared with the field when this becomes larger than the small value of 27 V/cm.

b. The ratio [f/r] may be of the order of magnitude 1 for gases at 10 V/cm, for liquids fields of 10^7 V/cm are needed to obtain this result.

c. The expression for [d/r] shows that at room temperature in gases the influence of the diffusion is 2.4 times as large as that of the recombination. In liquids on the other hand the recombination has an influence 400 times greater than the diffusion at room temperature and 30,000 times at 4°K.

In view of this consideration K r a m e r s looked for a solution of (6) without the diffusion term, and corrected afterwards for its influence.

Without diffusion his solution runs as follows:

 $p = f\psi(f) \tag{11}$

with:

 $f = \frac{F \sin \varphi}{F_0}, \quad F_0 = \frac{4\sqrt{\pi e}N_0}{b}.$

7

 $\psi(f)$ is given by:

$$\psi(f) = \frac{2}{\sqrt{\pi}} \mathcal{F}(-\ln f)$$

where $\mathcal{F}(\eta)$ is the function defined as:

$$\mathscr{T}(\eta) = \int_{0}^{\infty} \frac{\sqrt{\zeta} \, d\zeta}{e^{\zeta - \eta} + 1} ;$$

it has been calculated by McDougall and Stoner¹³) in tables for $-4.0 \le \eta \le 20.0$.

Using the property of $\mathcal{F}(\eta)$ that for $\eta \to -\infty$ (or: $f \to +\infty$):

$$\mathcal{F}(\eta)_{\eta \to -\infty} \to \frac{\sqrt{\pi}}{2} \left(e^{\eta} - \frac{e^{2\eta}}{2\sqrt{2}} \right),$$

it follows that $p(f)_{f\to\infty} \to 1 - \frac{1}{2f\sqrt{2}}$, or inserting the value of f, we obtain:

$$p_2 = 1 - \sqrt{2\pi} \frac{eN_0}{b} \frac{1}{F \sin \varphi}$$

which is the same as J a f f é's solution for large values of F (compare (5b)).

Accounting for the corrective influence of the diffusion, K r am e r s deduced the approximative solution:

$$p^* = j\psi(j^*) + p_D \tag{12}$$

with

$$f^* = f \left(\frac{2}{\sqrt{\pi}} \int_0^{cf} e^{-\xi^*} d\xi \right)^{-1}$$
 and $c = 4 \theta \sqrt{\pi} \frac{e^2 N_0}{kT}$

 θ is a number between 1 and 5.

For large values of f the integral $\int_{0}^{cf} e^{-\xi^{2}} d\xi$ tends to $\sqrt{\pi}/2$ and f^{*} to f and consequently p^{*} to p given by (11*a*).

This solution has been obtained by following the solution of (6) without diffusion only up to the moment where the diffusion term becomes comparable with the recombination term. The number of ions liberated after that moment is represented by p_D . In weak fields this quantity will soon increase to a constant saturation value $\ln c'/c'$, with $c' = 2c/\sqrt{\pi}$, and is of the same type as the first term in the right hand number of eq. (17) in section 1.64. Due to the uncertainity in θ this value of p_D is not very accurately predicted.

For small values of j, $f\psi(f^*)$ will be smaller than $f\psi(f)$, and shows

a linear behavour for f sufficiently small. Indeed as f tends to zero, then η tends to $+\infty$, and $F(\eta)$ to $\frac{2}{3}\eta^{3/2}$ so (11) becomes

$$p_3 = \frac{4}{3\sqrt{\pi}} i \left(-\ln t \right)^{3/2}. \tag{13}$$

On the other hand, since for $i \to 0$, $j^* \to \sqrt{\pi}/2c$, eq. (12) becomes

$$p_{3}^{*} = \frac{4}{3\sqrt{\pi}} f \left(\ln \frac{8 \, \theta \, e^{2} N_{0}}{kT} \right)^{3/2} + p_{D}, \tag{14}$$

OF

$$p_3^* = C_T f + p_D. \tag{15}$$

While the linearity in J a f f e's solution (5*a*) is a result of omitting the influence of the diffusion in (3), it results here precisely from taking diffusion into account.

In order to compare with experiment the formulae should still be averaged over the different values of φ and the variation of N_0 along the path of the α -particle should be taken into account.

1.23. Conclusions. Summarizing we see that:

1) In gases the theory of J a f f é will in general be in agreement with the experimental results because of the greater influence of the diffusion relative to that of the recombination.

2) In liquids the influence of the recombination is always stronger than that of the diffusion, and J a f f é 's calculations will in general not be in agreement with the experiments. Here K r a m e r s's calculations give quite different results and might afford a better base.

3) Both theories however will describe the approach to saturation in the same way.

In the next sections the apparatus will be described and the results of the measurements with liquid nitrogen and hydrogen will be reported and compared with the theoretical formulae discussed in this section.

1.3. The ionization chamber.

The dimensions of an ionization chamber have to satisfy the requirements for obtaining homogeneous electric fields between the electrodes and for almost absolute electric insulation. This last condition can be fulfilled at low temperatures without much difficulty

because of the freezing of the moisture on the surfaces of the insulators which causes the leakages at normal temperature. The smallness of the space in the cryostat makes it difficult to fulfil the first condition. Early experiments showed that to avoid large errors in the determination of the ionization current a symmetrical chamber was indicated. The dimensions of the chamber were limited by the inner diameter of the cryostat vessel (which was about forty millimeters). A compromise had to be found for a largest possible measuring electrode with a broadest possible guard ring around it.

In the case of α -particle ionization the apparent saturation will be reached at relatively low fields if the direction of the field is perpendicular to the path of the α -particle ¹⁴) (a so called transversal ionization chamber). Now the path of an α -particle of normal radioactive substances is in liquids only some hundredths of a millimeter in length, so very great difficulties are encountered if one tries to construct a transversal ionization chamber. In the chamber used for our measurements there was a continuous distribution of all directions of the paths of the α -particles relative to that of the field.

A sketch of the simple apparatus in the cryostat is given in the figure 1. In the german silver top of the cryostat was soldered an exhaust tube a of the same material. This tube carried a brass disc bto which three metal strips c were soldered, which at their lower end were screwed to a basal disc supporting an electrode e_1 . The second electrode e_2 opposite e_1 was surrounded by a copper guard ring *i*. The mechanical connection between e_2 and the tube *i* by which it could be lifted was made by a disc g of insulating material, screwed in a cap h. It was possible to vary the distance between the electrodes from 0 to 4 mm with an accuracy of 0.01 mm by a screw outside the cap of the cryostat. This distance was adjusted by placing the electrodes in contact with each other, and lifting e_2 over a distance read on a scale along the tube i outside the cryostat. This was of course only permissible when the polonium, that was placed in the electrode e_1 , had a protective shield (metal foil), because of the danger of damage to the polonnium layer and contamination on the electrode e_2 .

The electrode e_1 was fixed by three insulated metal springs to three quartz rods l placed on a disc k, and could be adjusted parallel to e_2 by three screws j, that could turn in the basal disc. The adjusting was done optically by observing the small strip of air on

the background of a lighted screen: After the adjustment of the apparatus a brass cylinder m was shoved around it and soldered to the top b. Thus the ionization chamber was protected from the liquid in the cryostat. In our first experiments the electrodes were placed directly in the cryostat, but measurements were impossible, because of bubbles which entered between the electrodes from the outside. In a closed chamber they can be avoided by putting the liquid in it under a higher pressure than that of the liquid in the cryostat.



Figure 1. The ionization chamber.

The electrode e_1 was connected with the outside of the ionization vessel by a rigid wire of constantan which ended in a drop of mercury in a glass sphere n in the bottom of the cylinder. A platinum wire through this glass made the electric contact between the mercury and a lead in the cryostat.

We mention the following dimensions: diameter electrode e_2 : 18 mm; outer diameter of the guard ring: 33 mm; distance between electrode and ring: 0.5 mm; diameter electrode e_1 : 28 mm; inner diameter cylinder m: 39 mm.

In the lower electrode a copper strip was inserted with polonium precipitated on it (Po figure 1).

During the measurements the upper electrode was connected with the electrometer, the lower one with the voltage supply. In the construction of the apparatus we had to guard against the possibility of a discharge between the wire at high voltage (voltages to 6 kV were used) and the metal parts of the cryostat. The chance of such discharges depended on the pressure of the vapour above the liquid in the cryostat. We met some difficulties at low temperatures: in the top of the cryostat the vapour assumes room temperature and discharges could easily taken place through the gas of low density. They were avoided by surrounding the high voltage wire with a rubber tube along its whole length.

1.4. The method of measuring the ionization currents.

1.41. General remarks. For measuring ionization currents several methods present themselves. The difficulties of obtaining materials (the investigations started at the end of 1940) led us to keep the method as simple as possible. Another argument for simplicity was that some preliminary experiments showed that a simple electrostatic method with an electrometer reasonably fulfilled, the requirements of accuracy. The expectation that the difficulties would be most in the experiment itself and not so much in the method of measuring, was another argument.

The simplest way of measuring an ionization current is by determining the amount of electric charge that the ions transport in a certain time to one plate of a condenser. This is done by compensating this charge by changing the voltage on the second plate of the condenser, using the electrometer as a zero instrument. This method has two important advantages:

a. The total system of the electrometer and the measuring electrode in the ionization chamber that is connected with it remain at zero potential, thus no capacities are introduced in the calculations.

b. During the measurements there is no change in the voltage difference between the electrodes.

One of the disadvantages of this method is, however, that measuring one current takes some time and instantaneous changes are not detected. As our measurements deal only with currents that are not, or only slightly varying with time this was not considered important.

The electric circuit consisted of two parts:

1. the (high) tension circuit.

2. the electrostatic measuring circuit. These circuits being in general the same as we have used for our measurements with X-rays. we only give here the diagram (figure 2) *) and a short summary.

1.42. The voltage circuit. Since rather high fields were required. we had to use high voltages though the distance between the electrodes in the chamber was always small (of the order of magnitude of some tenths of a millimeter). We used three supplies:

1. a rectifier 0 V-300 V (Philips, G.M. 4560) regulable continuously by a potentiometer circuit outside of the apparatus:



Figure 2. The measuring circuit.

2. a rectifier 0 V-2000 V, constructed in the physical laboratory of Philips, Eindhoven, output stabilized with neon tubes, regulable in discontinuous steps of 100 V each;

3. a generator 1000 V-6000 V, constructed by Smit, Slikkerveer, regulable by adjusting the excitation of the generator †).

The value of the output voltage was measured by an electrostatic

^{*)} loc. cit., figure 1.

^{†)} This generator was only used for the measurements with the covered polonium sample (see following sections) as it was lost by german requisition.

voltmeter and a voltmeter consisting of a milliammeter and a resistance of $6.7 \text{ M}\Omega$.

1.43. The measuring circuit and the measurements. The electrometer electrode (Cr, figure 2) in the ionization chamber was connected directly with a Lutz-Edelmann electrometer with a highest sensitivity of 5 mV per scale division. The capacities C_1 and C_2 (that could be given values varying from 25 to 4000 pF) could be connected by means of magnetically controlled electrostatic switches parallel to the electrometer and chamber system. The compensation voltage was given by the compensation current (which was regulated with R_2) and a resistance R_1 fixed at one measurement. Except for some condensers the electrostatic system was placed in a metal box that was kept dry by continuous heating with a heating coil. All the condensers, were compared electrostatically with a cylinder condenser of 24.46 pF, made and calibrated in the Physical Laboratory of the University of Amsterdam ¹⁵) *).

1.44. Sensitivity and accuracy of the method. Our measurements did not require very strict conditions as to sensitivity and accuracy since the irreproducibility of the results was in general of the order of magnitude of a few percent. One hundredth of a picoampère could be measured with an accuracy of 2%, for a picoampère the error was smaller than $\frac{1}{2}\%$.

When measuring a given current the following points are to be considered:

a. the constancy of the voltage,

b. the speed of the electrometer wire,

c. the time of measurement,

d. electrostatic leaks.

The first condition was adequately fulfilled. Only at high voltages was the ripple of the 7000 Volt generator perceptible. This was not serious (except for measurements in nitrogen), since at these voltages the currents could be measured with a lower sensitivity of the electrometer.

The speed of the wire depends on the capacity of the system of conductors and the sensitivity of the electrometer, the time of measurement depends upon the value of R_1 and the number of volts

^{*)} We wish to thank Prof. dr J. Clay for kindly placing this condenser at our disposal.

available for the compensation. In general a measurement lasted 60 seconds and was repeated three times or more.

The observer can regulate the speed of the wire so as to be most suited for his purpose. The condition was that the passing of the zero point should be easily observed and measured with a stopwatch.

1.5. Some remarks concerning the experiments.

1.51. The ionizer. From the first experiments it followed that a polonium sample with an intensity of ionization in gaseous air under normal circumstances of 3000 pA was very suitable for our measurements. A higher ionization intensity caused currents which only after some time became stationary, a phenomenon to which we shall return in a following article. A lower intensity on the other hand reduced the value of the currents in liquid nitrogen below the observable limit. The series of measurements that are reported were obtained with a polonium sample that was covered by a tin foil to reduce its intensity and with a sample without a cover.

As a theory of V o n W e i s z ä c k e r¹) predicts an increase of the stopping power of metals at low temperatures, we had to check this theory by experiment. The results have shown ¹⁷), that no change exists in the cases of aluminium and tin, so the use of these metals as a shielding foil meets no difficulties with regard to the range of the α -particle in the liquid after passing the foil.

1.52. Corrections for unevennesses in the polonium sample. In our first experiments we thought we found a considerable influence of the distance of the electrodes on the current at a given field. Closer analysis showed that this was due to an unevenness in the electrode with the polonium coating. Direct determination of this unevenness and, independently, the analysis of the ionization currents as a function of the field allowed to establish a correction on the assumed value of the applied field which was the same for different liquids and which caused the apparent distance effect to vanish. Measurements carried out with a second, uncovered sample *) which had a smaller unevenness showed also, that no real distance effect exists. In order to avoid possible errors in the determination of this dis-

^{*)} We wish to express our sincere thanks to dr E. Broda of the Cavendish Laboratory, Cambridge, for placing some polonium at our disposal, and to dr F. Barendregt (Vrije Universiteit Amsterdam), who precipitated the polonium on the given copper electrode.

tance between the electrodes during a measurement, it was fixed by quartz rods placed in holes in the largest electrode. The distances between the tops and the surface of the electrode were measured with a tachometer.

1.6. Results of the measurements in liquid nitrogen.

1.61. The currents as a function of the field. In this section we shall report the results in liquid nitrogen, which were obtained with the covered and the uncovered sample, respectively.



Figure 3. The ionization current in liquid nitrogen (77°K) as a function of the field, for low and high fields separately. The curves are calculated with Kramers's formula for p^* (dotted lines: $p = f\psi(f) + p_D$). Measured points: uncovered sample, positive (\bigcirc) and negative (\bigcirc) ions; covered sample, positive (\bigcirc) and negative (\bigcirc) ions, respectively.

The liquid in the chamber was condensed from a vessel with pure and dry gas and was put afterwards under a pressure that was about 10 cm higher than that over the liquid in the cryostat glass. With this extra pressure we avoided bubbles in the chamber.

From the measurements of i on a given day we calculated the values p = i/I, where I is the saturation current in nitrogen gas measured on another day and corrected for the decay of the polonium. The determination of I in the case of the covered sample could be performed in the ionization chamber in the cryostat because the density of the gas at 90 °K was large enough to keep the

F	$\frac{i}{I}$	× 10 ⁴	F kV/cm	$\frac{4}{I} \times 10^4$		
kV/cm	uncovered (April 10, 1946)	covered (March 3, 7, 1944)		uncovered (April 10, 1946)	covered (March 3, 7, 1944	
0.118	2,28		10.4	18.9		
0.277	3.07		13.8	23.4	3.97	
0.417	3.48		17.3	27.5		
0.705		0.762	20.9	31.8		
0.710	4.39		26.4	39.0		
1.04	5.14		26.9		6.72	
1.38	5.70		39.7		9.31	
1.41	0.00	1.16	51.1	65.5		
2.08	6.89	220.20	52.6		11.7	
2.00	0.07	1.41	67.6		15.1	
2.11		1.81	73.1	91.4		
5.47		2.21	83.9		18.1	
6.05	14.1		101.5	and a state of the	21.4	
7.24	1.4.4	2.73	111	128		
7.24		2.83	119		25.3	

TABLE I

 α -rays from reaching the other electrode. The ionization intensity of the uncovered sample was measured in a large chamber filled with gas under normal conditions.

The results of the measurements in liquid nitrogen, corrected for the non-uniformity of the surfaces of the electrodes, are given in figure 3 for weak and strong fields separately. For each case we have chosen a series with currents of positive ions which is representative for the results. The data of figure 3 are recorded in table I.

Our results are in qualitative agreement with those obtained by M c L e n n a n in liquid air, which are reproduced in figure 4. A comparison between measurements in fields of opposite sign shows that a difference between the current of positive ions (single marks in figure 3) and that of negative ions (double marks) did not surpass the accuracy of the measurements (about one percent).

1.62. The influence of the distribution of φ . There is a marked difference in the magnitude of *i* and of *i*/*I* between the measurements with the covered and those with the uncovered polonium sample. One feels tempted to ascribe this mainly to a difference in the distribution of the angles between the field and the columns. The angle φ

corresponds to a random distribution in space between 0 and $\pi/2$ for the uncovered sample and between 0 and about $\pi/4$ for the covered sample (the last number being estimated from the range equivalent of the foil that was found to be 0.706-times the total range of the α -particles in tin).





1.63. The influence of temperature. A measurement showed that at 65°K higher fields are needed for the same currents than at 77°K. From table II we see that for two series with the uncovered sample there is a constant ratio $F_{77}/F_{65} = 0.90$.

1.0.1	10.00		The second second second	and the second second second second second second by				
$p \times 10^{*}$	18.7	32.0	43.5	54.0	65.5	74.0	83.5	
F77	10.0	20.0	30.0	40.0	50.0	60.0	70.0	
F 65	11.3	22.5	35.5	43.8	55.0	66.0	78.3	
F_{77}/F_{65} .	0.89	0.89	0.90	0.91	0.91	0.91	0.0	

TABLE II

1.64. Comparison of the results with the theory. 1.64a. Jaffé¹⁸) described the results of his measurements in liquids at room temperature by

$$i = i_D + aF. \tag{16}$$

The second term on the right hand corresponds to (5a) which represented an approximation in which the influence of the diffusion on the solution of equation (2) was neglected. The current

2

 i_D may be regarded as a small diffusion effect that exists at this temperature. In fact diffusion will prevent the recombination of a small fraction of the ions, which are easily brought to the electrodes even by a very small field (compare page 7). Dividing (16) by the value of the saturation I it becomes:

$$p = p_D + a'F. \tag{17}$$

Our measurements in nitrogen can also be described by this formula, so we will use it to test the J a f f é theory.

For nitrogen at 77°K we deduce form our measurements

$$\phi_{D} = 1 \times 10^{-4}$$
 and $\phi_{D} = 4 \times 10^{-4}$

for the measurements with the covered and the uncovered sample, respectively. These values are of the same order of magnitude as would follow from M c L e n n a n's measurements with liquid air, which gave $p_D = 10^{-4}$ (there is an uncertainty in the value of I which we had to estimate from his data on the measurements in a gas at high density) ¹⁹).

Considering the results with the uncovered sample, we see (compare figure 3) that they can be represented by a straight line at weak fields up to about 15 kV/cm. For stronger fields, however, the p(F)-curve is concave against the *F*-axis. This feature is clearly in contradiction with J a f f é's formula, since this formula would require that once the linear dependency of p on *F* has started, it would stretch to much larger values of p. Indeed, deviation from linearity would first show up when the first term (1) in the denominator of (4) makes itself felt beside the second term, but this would require p-values, much larger than those ($p \approx 0.015$) appearing even at the highest *F*-values ($F \approx 200 \text{ kV/cm}$) in our experiments.

Our conclusion is that the relation between field and current in the case of liquid nitrogen does not agree at all with the Jaffé theory.

1.64b. Since a similar result holds for the measurements in liquid hydrogen, K r a m e r s was led to modify J a f f é's calculations, which modifications are described in 1.22. In order to compare with the formulae given there, we proceeded as follows.

As remarked on page 8, the formula (12) should be averaged over the different values of φ present in the experiment. This can

be done by replacing $\sin \varphi$ in the formula for f by a suitable average value $\sin \varphi_0$ which will take different values in the case of the covered and that of the uncovered sample, but which in each of these cases may be expected to be practically independent of the field. This was corraborated by some numerical integrations based on (11), which showed that for the uncovered sample (φ distributed as $\sin \varphi \, d\varphi$ between 0 and $\pi/2$) $\sin \varphi_0$ may be taken approximately equal to 0.80 whereas for the covered sample (φ distributed between 0 and about $\pi/4$; the ranges are shorter for larger angles) $\sin \varphi_0$ may be taken approximately equal to 0.46. Still there may be reasons, as we will suggest later on, to consider calculations with a $\sin \varphi_0$ different from 0.8 illusory.

Furthermore we have replaced N_0 in the formulae for F_0 and c by an appropriate average value, taking the variation of the ionization along the path of the α -particle into account. From a consideration of the Bragg ionization curve²⁰), the total ionization in gases²¹, the range equivalent of the foil and the density of the liquid²²) we decided on the value $N_0 = 2.57 \times 10^7$ ion pairs/cm for the uncovered sample and on the value $N_0 = 3.38 \times 10^7$ ion pairs/cm for the covered sample, respectively, in liquid nitrogen.

In the case of the covered sample the influence of the diffusion is given by the ratio (10b)

$$\left[\frac{d}{j}\right]_0 \approx \frac{0.3}{0.46} \frac{77}{F} = \frac{50}{F}$$

so that if $F\!>\!{\rm about}$ 15 kV/cm the diffusion may be neglected relative to the field.

We shall compare the results with the formula (12), in which the values $p_D = 1.0 \times 10^{-4}$ (which is of the right theoretical order of magnitude), sin $\varphi_0 = 0.46$ and $c = 10.5 \times 10^3$ (calculated with the somewhat arbitraty choice $\theta = 2$ and the average $N_0 = 3.38 \times 10^7$) are inserted. With these values and using the tables of McD o u-g a 11 and S t o n e r *) we calculated the curve for $p^*(f)$. At the values $p_{\text{measured}} - p_D$ at a field F (taken from a smooth curve which represented the experimental results within fair approximation) the corresponding numbers f were found in the curve $p^*(f)$. Divided into 0.46F they gave values F_0 which were plotted against F (figure 5).

*) loc. cit.

We see that except for weak fields F_0 is a constant and equal to 4.6×10^5 kV/cm. Inserting this F_0 into (11) and (12), the curves for p and p^* that are given in figure 3 could be calculated. In order to compare the curve for p with that for p^* , p was drawn as $p = f\psi(f) + p_D$. These curves show, that K r a m e r s's formula (12) for p^* is in agreement with the experimental results.

In the formulae b represents an effective value of the diameter of the columns, which is based on the model of a gaussian distribution of the density of the ions round the path of the a-particle. Now at the moment the ionization is produced, the distribution is certainly of a type quite different from the gaussian distribution (1). Still, in gases where the relative effect of the diffusion is large, a gaussian distribution may be established pretty soon \dagger). In liquids, however, things are quite different, and here b_{hig} can only be interpreted as an effective value, which need not be simply related to b_{gas} .

This is brought out by a comparison between the values of $b_{\rm gas}$ and $b_{\rm liq}$ that can be calculated from experimental data. From the value $b_{\rm gas} = 1.8 \times 10^3$ cm (which is given by J a f f é and others as a result of ionization experiments in air and oxygen) and the assumption that b is inversely proportional to the density of the medium, there follows a value $b_{\rm l,g} = 30 \times 10^{-7}$ cm. Inserting the value F_0 given above, and $N_0 = 3.38 \times 10^7$ in the equation (11), there follows for nitrogen in the case of the experiments with the covered sample $b_{\rm lig} = 0.75 \times 10^{-7}$ cm, *i.e.* 40-times smaller than $b_{\rm l.g.}$

In the case of the uncovered sample the same procedure for calculating F_0 was followed. Now we used the values: $\sin \varphi_0 = 0.80$, $N_0 = 2.57 \times 10^7$, $c = 7.95 \times 10^3$ and $\phi_D = 4.0 \times 10^{-4}$. Again an approximate constant value F_0 could be found: $F_0 = 1.15 \times 10^5$ kV/cm (compare figure 5). Using this value, we see that the curve for ϕ^* represents the results with fair accuracy (figure 3).

With the just mentioned values of N_0 and F_0 there follows now that $b_{\rm liq} = 2.5 \times 10^{-7}$ cm, *i.e.* 3-times the value of $b_{\rm liq}$ in the preceding case and 1/12 of the value of $b_{\rm l,g}$.

As well the big difference between the b_{liq} -values in the cases of the covered and the uncovered sample and their difference from the expected $b_{\text{l.g}}$ -value, may at first sight seem strange and

t) reference 6, page 316.

difficult to reconcile with the theory. In a following article, dealing with measurements in liquid argon and helium, we shall come back to this diffeculty and suggest a natural explanation.

The influence of the temperature on the ionization current can be understood on considering, that since $F_0 = F/f$, $F_0 \sim N/b \sim \varrho^2$ (where ϱ means the density of the liquid which depends on the temperature), F for a same value of ρ at different temperatures will change as $F \sim \varrho^2(T)$. Now $\varrho_{77^o}/\varrho_{65^\circ} = 0.93^{23}$), so $F_{77^o}/F_{65^\circ} =$ 0.87. Experimentally the value 0.90 (table II) has been found.

1.7. Results of the measurements in liquid hydrogen.

1.71. The results and their comparison with the theory. The measured relative currents i/I were in general larger than those measured in nitrogen at corresponding fields.





The reproducibility of the measurements was reasonable: on different days with recondensed liquid the irreproducibility did not exceed 3 percent of the mean values. The difference was larger (5%) for measurements with different foils, because of the irreproducible way in which the foil passed over the non-uniformities in the polonium surface.

In figure 6 some values of currents of positive ious, that are representative for the results and which are tabulated in table III, are given in separate graphs for fields to 20 kV/cm and to 200 kV/cm

F	$\frac{i}{I}$	< 10 ⁴	F	$\frac{i}{I} \times 10^4$			
kV/cm	uncovered (April 11, 1946)	covered (March 9, 1944)	kV/cm	uncovered (April 11, 1946)	covered (March 9, 1941)		
0.125	4.16	8	30.1		35.7′		
0.277	6.06		39.0		45.5"		
0.425	7.66		39.5	ALC: NOT THE REAL OF	42.4*		
0.53	1	2.05'	50.2		56.5*		
0.68	9.73		50.3	about the second	56.0'		
1.04	12,2		58,6		63.6"		
1.05		2.25'	62.2		62.8*		
1.39	14.3		69,1	Commission of Commission	73.6'		
2.08	17.8	3.42'	73.0	188			
3.16		4.62'	97.1		94.4'		
5.18	The Lot Press of	6.68'	100.8	State Street	101 *		
6.95	35.4		112	261			
9.75	10000	12.9 "	134		123 *		
10.4	45.4		139	310			
10.8		12.8 '	152		133 *		
13.7		16.9 *	168	1	145 *		
13.9	54.0		171	367			
19.5		23.6 "	185		154 *		
20.8	71.0	25.9 '	202		162 *		
26.4	84.4		Distar	ices between the	electrodes:		
26.7	7.21.2	29.8 *	uncov	ered: 0.0195 cr	n		
29.2		35.0 //	covere	d: (1) 0.010 cn	1		

	-		
10.00	1.0	11. 10. 10.	
1.1.1.1		10.00	

kV/cm, respectively. Only in the case of the covered sample the current is linear in the fieldstrength at low fields, and corresponds to $p_D \approx 1 \times 10^{-4}$. With the uncovered sample there is no region with clearly defined linearity. The drawn curves in this figure are calculated with (12) and with the values of F_0 which are deduced from the lower graph in figure 5.

The curve 1 in figure 6 represents the results with the covered sample rather well with $F_0 = 7.5 \times 10^4 \,\mathrm{kV/cm}$ and $p_D = 1.0 \times 10^{-4}$. From this value of F_0 follows (compare (11)) $b_{\mathrm{liq}} = 1.6 \times 10^{-7} \,\mathrm{cm}$ when we use the value 1.20×10^7 for the number N_0^{21}). With a value $b_{\mathrm{gas}} = 1.9 \times 10^{-3} \,\mathrm{cm}^{-9}$ ²²), one should expect $b_{\mathrm{l,g}} = 24 \times 10^{-7} \,\mathrm{cm}$, *i.e.* about 15-times our experimental value.

In figure 6 the dotted line gives the relation between current and

field if the correction for the diffusion is omitted. The first condition of (10b) shows that the influence of the diffusion should be negligible for fields larger than 4 kV/cm.

Also for the case of the measurements with the uncovered sample, *i.e.* with the measurements with higher effective fields (sin $\varphi_0 = 0.8$), an approximatively constant value of F_0 could be found.



Figure 6. The ionization current in liquid hydrogen (20.4°K) as a function of the field, for low and high fields separately. The curves are calculated with Kramers's formula for p (dotted lines: $p = f\psi(f) + p_D$). Measured points: uncovered sample, positive (\bigcirc) and negative (\bigcirc) ions; covered sample, positive (\frown) and negative (\boxdot) ions.

With a chosen value $p_D = 9.0 \times 10^{-4}$ there follows from the measurements that F_0 equals 4.25×10^4 kV/cm (figure 5). Using this value we have computed the curve 2 in figure 6, and we see, that (12) agrees with the results within the accuracy of the measurements. Since for these experiments we have $N_0 = 7.70 \times 10^6$, we found the value $b_{\rm liq} = 1.8 \times 10^{-7}$ cm, which does not differ much from the corresponding value in the case of the covered sample and which is about 1/13-th of the theoretical expected value.

1.72. The influence of temperature. Most measurements in hydrogen were performed at 20°K, but we measured one series at 14°K. As follows from table IV there is a nearly constant ratio between the fields required for obtaining the same current at 20°K and at 14°K. Assuming again $F \sim \varrho^2$, and using $\varrho_{20^\circ}/\varrho_{14.5} = 0.935^{24}$),

Aug. 1			
10.00	1.120	10.125	1.17
10.02	N 12.	1.2.2.2.2.2.2.2.2.2.2.2.2.2.2.2.2.2.2.2	CL 1 W / -

$p \times 10^{4}$	60	120	180	240	300	360	
F_{10} ,	16.0	40.5	69.0	100	133	167	
F 14.5	20.0	50.5	85.0	122	159	198	
F / F	0.80	0.80	0.81	0.82	0.83	0.84	

one should expect $F_{20}/F_{14.5} = 0.87$, which value differs slightly from the ratio deduced from table IV.

1.73. Currents of positive and negative ions. In hydrogen a difference has been found between the measurements with positive and negative ions. The saturation current in gas was found the same within less than one per cent for both directions of the field. These measurements were done both with the covered and the uncovered polonium sample, so that for a comparison in table V the effective fields ($F_{\rm eff}=F\sin\varphi_0$) are given.

The two series join each other and it appears that for high fields the ratio p_{-}/p_{+} is a constant and equals 1.03 *).

 0.000		-		
 V 1.2	10.00	100	A	
 5.05	1.21	12.00	AC	
 Sec. 61.07.				

Foff	covered						uncovered		
kV/cm	9,2	18.4	28	37	46	48	64	80	
p_ × 104	24	46	65]	82	96	160	202	240	
$p_{-} \times 10^{4}$	26	49	68	86	100	165	207	247	
p_1p,	1.08	1.07	1.05	1.05	1.04	1.03	1.03	1.0	

1.8. Conclusions.

In this section we mention some conclusions that can be drawn so far from our experiments. In the following chapter more detailed considerations will be given after a discussion on the results of the experiments in liquid argon and liquid helium.

Our measurements are in accordance with the fundamental ideas of the theory of column ionization applied to liquids.

It has been ascertained, however, that the formulae of the J a f f é theory cannot be applied to liquids at low temperatures because of the preponderance of recombination over diffusion. It appears that the modifications given by K r a m e r s are qualitatively in

^{*)} I have the impression that this somewhat surprising dissymmetry is real, although I can offer no explanation for this effect, which has been found only in hydrogen.

agreement with the results as regards the influence of the diffusion at low temperatures, and his formulae give a fair description of the ionization current as a function of the field.

Still the fact that the calculated values of the column diameters deviate considerably from those that are calculated from the known values in gas under normal conditions suggests that the modified theory requires further corrections. Two points where such corrections may be important present themselves immediately. One is the disturbing influence of the high concentration of the ions, that causes a strong initial recombination, on the applicability of the formula for volume recombination within the column, the other refers to the assumptions of an initial gaussian distribution of the ions in the column. Whereas, when applied to gases, these assumptions are not unreasonable, the relative smallness of the diffusion effect makes their validity appear questionable in liquids at low temperatures.



REFERENCES

- 1) G. Jaffé, Le Radium 10, 136, 1913.
- 2) Among others: H. Greinacher, Phys. Z. 10, 986, 1909.
- 3) H. Folmer, Proc. kon. Akad. Amsterdam 23, 759, 1929.
- 4) J.C. McLennan and D.A. Keys, Phil. Mag. 26, 876, 1913.
- A. N. Gerritsen and Jacqueline Koolhaas, Physica, 's-Grav. 10, 49, 1943; Commun. Kamerlingh Onnes Lab., Leiden No. 266b.
- 6) G. Jaffé, Ann, Physik, (4) 42, 303, 1913.
- 7) P. Langevin, Thesis, Paris 1902.
- 8) H. Zanstra, Physica, 's-Grav. 2, 815, 1935.
- 9) P. H. Clay, Thesis, Delft 1942.
- 10) H. A. Kramers, to appear shortly in Physica, 's-Grav.
- 11) See reference 9, page 45, and the original communications.
- 12) Int. Crit. Tables I, 367, 1926.
- J. McDougall and E. C. Stoner, Phil. Trans. roy. Soc., London (A) 237, 67, 1938.
- 14) H. Stücklen, Hdb. d. Phys. Bd. XIV, Kap. 1, 1927.
- 15) J. Clay, Physica, 's-Grav. 3, 757, 1936.
- 16) C. F. von Weiszäcker, Ann. Physik (5) 17, 869, 1933.
- A. N. Gerritsen, Physica, 's-Grav. 12, 311, 1946; Commun. Kamerlingh Onnes Lab., Leiden, No. 270b.
- 18) See reference 6, page 331.
- 19) J.C. McLennan and D.A. Keys, Phil. Mag. 30, 484, 1915.
- 20) G. A. W. Rutgers, Thesis, Utrecht 1940.
- 21) Landolt Börnstein, Phys. Chem. Tabelle, Eg. III 6, 1315, 1935.
- 22) Values of ϱ in the liquid are taken from Int. Crit. Tables 1, 102, 1926.
- 23) E. Mathias, H. Kamerlingh Onnes and C. A. Crommelin, Commun. Kamerlingh Onnes Lab., Leiden No. 145c.
- 24) H. Kamerlingh Onnes and C.A. Crommelin, Commun. Kamerlingh Onnes Lab., Leiden No. 137a.
CHAPTER II

MEASUREMENTS IN LIQUID HELIUM AND LIQUID ARGON

Summary

In continuation of a previous article on liquid nitrogen and hydrogen the results are reported of ionization measurements with *a*-particles of polonium in liquid helium and liquid argon. In both cases the results can not quantitatively be described by the formulae of Kramers's theory in contrast to the cases of nitrogen and hydrogen. It is argued that this may be due to the unadequacy of J a f f é's assumption of an initial gaussian distribution of the densities of the ions along the path of the *a*-particle; a correction calculated on the assumption of a spreading of the ratio between the local linear ion density and the local diameter of the column leads to a better applicability of the theory.

The currents in the liquified inert gases are 10 to 20 times larger than in liquid nitrogen and liquid hydrogen. This seems to require a value of the recombination of the ions in liquid helium and liquid argon, which is much smaller than that given by L ang e v i n's classical expression.

Apart from an irregularity in the immediate neighbourhood of the λ -point, the currents in liquid helium are nearly the same above and below the transition temperature. The results with α -particles are similar to those with X-rays as an ionizer.

2.1. Introduction. The experiments in nitrogen and helium with X-rays ¹) showed that the order of magnitude of the ionization currents in liquid helium was many times larger than that of the currents in liquid nitrogen. As this difference might be caused by a different specific ionization by the X-rays in the two liquids it was of interest to repeat the measurements with an ionizer which has a known intensity of ionization in each liquid. So the measurements that are described in our previous article on this subject ²) (which we will cite as **1**) were also carried out in liquid helium.

- 27 -

Here again we found that with a strong sample it took a long time, before the ionization current reached a stationary value, and again we could reduce this effect by putting a metal foil over the sample. In a separate article this and related effects, which can be interpreted as a consequence of a polarization of the electrodes, will be discussed in some detail.

The experiments were, just as in nitrogen and hydrogen, carried out with two polonium samples of which the stronger one was covered by a tin foil with a range equivalent of 0.706 times the total range of the α -particles in air, such, that both samples had about the same effective intensity of radiation.

For the details of the experiments we refer to 1, sections 3, 4 and 5.

After the measurements in liquid helium were done, it became of interest to compare the results with those in another liquified inert gas. Hence a few measurements were done with the uncovered polonium sample in liquid argon.

2.2. The results in liquid helium.

2.21. General remarks. In this section the results are reported of measurements in liquid helium at about 4°K. The results in the neighbourhood of the transition temperature and at lower temperatures are reported in section 2.4.

After the cryostat was filled with liquid helium, helium gas was condensed into the vessel with the ionization chamber (compare 1, figure 1). With the covered sample the measurements were carried out with several distances between the electrodes. Below we have recorded some results for positive ions which we have chosen as representative for all measurements. The currents could be reproduced within some 5% on different days, on one day they were reproducible within 1%.

In the experiments with the uncovered sample the fixed distance between the electrodes was 0.019_5 cm, which was somewhat smaller than the range of the α -particles that moved in a direction perpendicular to the electrodes. Thus a correction to the value of the saturation current in the liquid should have been applied. This correction would depend on the density of the liquid and would not exceed 6% at 4°K and 3% at 1.3°K. Its exact value is rather uncertain, however, and its neglect will not materially influence our discussion of the results.

		/I		<i>i/I</i>		
F kV/cm	uncovered April 12, 1946	covered F 46 March 24, 1944 kV/cm May 5, 1944 kV/cm	F kV/cm	uncovered April 12, 1946	covered March 24, 1944 May 5, 1944	
3.47	0.0242		51.9	Un van de la la	0.114"	
3.58		0.0130+	62.2	and the second second	0.126×	
4.43		0.0172+	73.1	0.250		
6.94	0.0564		77.9		0.142"	
12.2		0.0463*	81.9		0.145'	
20.5		0.0651'	83.7		0.146×	
20.9	0.133		104.1	17 1 1 VI 1 1	0.162"	
24.3		0.0766*	111.3	0.292		
26.0		0.0768"	123		0.176'	
36.3		0.0976*	130		0.182"	
38.4	0.184	and the second second	149	0.316		
38.8		0.0995+	156		0.192"	
40.9		0.101'	171	0.340		

TABLE I

Distance between the electrodes: uncovered: 0.0195 cm; covered: 0.010 cm('), 0.020 cm (''), 0.070 cm (×) and 0.120 cm (*) (March 24), 0.040 cm (+) (May 5).



Fig. 1. The ionization current in liquid helium at 3.98 °K, as a function of the field. Measured points: uncovered sample, positive (\bigcirc) and negative (\bigcirc) ions; covered sample, positive (\bigcirc) ions, respectively. The curves 1, 2' and 2" are calculated with K r a m e r s's formula for $p = j\psi(f)$, with $F_0 = 1600 \text{ kV/cm}$, 1200 kV/cm and 900 kV/cm, respectively. The points ($-\frac{1}{2}$) have been measured at 1.3 °K.

2.22. The current as a function of the field. Some representative series of the current *i* of positive ions at 4°K are recorded in table I and figure 1, which give the ratios $\phi = i/I$, where *I* is the saturation value, as a function of the electric field. The field has been corrected for the unevennesses in the surfaces of the electrodes (compare 1. 5).

The saturation current in gaseous helium was measured by introducing helium gas into the ionization chamber at 20.4°K. The measured ratios of the saturation currents in hydrogen and helium in their gaseous state relative to air, compared with the values of these ratios given by G u r n e y and others ³), show agreement within one percent. Since the ionization chamber is not particularly adapted to measurements in gases, this agreement is satisfactory.

The measurements at 4°K were carried out with distances between the electrodes varying from 0.010 cm to 0.120 cm with the covered sample, and without large deviations (only one point of the given series at 40.9 kV/cm differs 5% from the smooth curve) the results could all be represented by the curve a within fair approximation, which supports the correctness of the assumption made in correcting for the unevennesses in the surfaces of the electrodes. Measurements with the uncovered sample with a distance of 0.0195 cm between the electrodes, can be represented by the curve b. The data for low fields are not inserted, since these dit not reproduce probably because of small thermo-electric and other electro-motoric forces between the electrodes, which resulted into small ionization currents when no external voltage was applied.

2.23. Comparison of the results with the theory.

2.23*a*. There is no region of linear dependency of current on fieldstrength as J a f f e's formula (1. (5a)) (this refers to equation (5a)in article 1) should require. Hence the condition for applying the formula of K r a m e r s (1.(12)) seems to be realised at first sight. From the estimate for the several influences acting on the ions (1. (10)), it follows that the influence of the diffusion may be neglected relative to that of the field when:

$$\left[\frac{d}{j}\right] \approx 0.3 \, \frac{T}{F \sin \varphi_0} \ll 1,$$

which at $T = 4^{\circ}$ K will be realised for the covered sample (sin $\varphi_0 = 0.46$)

for fields stronger than $0.8 \,\mathrm{kV/cm}$. The diffusion may also be neglected relative to the recombination since according to (1.(10)):

$$\left[\frac{a}{r}\right]_{0} \approx 8 \times 10^{-6} T = 3 \times 10^{-5} \ll 1.$$

So there are no objections against applying the expression (1.(11)) $p = f\psi(f)$ (with $f = F \sin \varphi_0/F_0$ and F_0 a parameter that has to be found from the experiments) on the results.

Although we remarked before (1.64b) that calculations with a factor $\sin \varphi_0$ different from 0.8 may be illusory, we will analogous to our considerations on the experiments with nitrogen and hydrogen use $\sin \varphi_0 = 0.46$ for the results with the covered sample.



Fig. 2. $F_0(F)$ for helium (upper graph) and argon (lower graph). Full points: covered sample; circles: uncovered sample.

From the experimental value of p the corresponding values of f are calculated by the same procedure as described in (1. 64b), supposing $p_D = 0$, however. In the upper part of figure 2 the values of F_0 are given as a function of the field F (dots), and we see that a constant number F_0 can not be deduced, which was required for the validity of K r a m e r s's formula. In figure 1 we have drawn a curve (1) for $p = j\psi(f)$ with $F_0 = 1.6 \times 10^3 \, \text{kV/cm}$. Since ³) $N_0 = 8.8 \times 10^6$ this would mean for the diameter of the column: $b_{\text{hig}} = 55 \times 10^{-7} \, \text{cm}$. This curve and the experimental curve do not coincide.

2.23b. The results with the uncovered sample are similar. The *p*-values are larger now, similar to the experiments in nitrogen and hydrogen.

Again the formulae of J a f f é cannot be applied to the results for the same reason as before, and similarly no constant F_0 can be deduced from the results when the K r a m e r s formula is used (circles in figure 2).

We calculated for these experiments two curves for p with (1.(11)), inserting $F_0 = 1200 \text{ kV/cm}$ (curve 2') and $F_0 = 900 \text{ kV/cm}$ (curve 2'), respectively.

With the number $N_0 = 6.6 \times 10^6$ and the value $F_0 = 900 \text{ kV/cm}$ we found for the average diameter of the columns $b_{\text{lig}} = 75 \times 10^{-7} \text{cm}$.

The mentioned values of $b_{\rm liq}$ are of the same order of magnitude as the value $b_{\rm l,g} \approx 27 \times 10^{-7}$ cm, which can be calculated from the value of the diameter of the column in gaseous helium under normal conditions, for which we took the hydrogen value $b_{\rm gas} \approx 2 \times 10^{-3}$ cm⁵).

2.24. Currents of negative ions. In table II the results of experiments with fields of opposite directions with the covered and uncovered sample are plotted against the effective field $(F \sin \varphi_0)$. The measured saturation currents for positive and negative ions in gas were found to be the same within 1%.

The currents	of positive and	l negative ion the uncove	s in helium in red sample	the case of the	covered and
$F \sin \varphi_{\delta}$ kV/cm	5.2	10,4	21	16,7	84
$p_+ \times 10^2$	6.70	9.76	13.2	13.3	29.0
$p_{-} \times 10^{2}$	6.79	9.76	13.2	13.5	28.7
p_/p_	1 015	1.00	1.00	1.015	0.99
		covered $T = 2.29^{\circ} \mathrm{K}$	unco T = 3	vered 98°K	

TABLE II

The differences between p_+ and p_- are unimportant and similar to our results in nitrogen.

2.3. The results in liquid argon.

2.31. *General remarks*. Since the results in helium differed so much from those in nitrogen and hydrogen, we thought it of interest to measure another inert gas. We chose argon, which has its boiling point near that of oxygen.

The gas was condensed into the vessel in the cryostat which was filled with liquid oxygen boiling under reduced pressure. The temperature of the argon was 88°K, and one series of measurements was carried out with the uncovered sample.

2.32. The results in liquid argon. The values of p which were . calculated from the measured currents (the number I was found by measuring the saturation current in air and multiplying by the factor 1.38; comp.³)) were smaller than in helium at corresponding fields but a good deal larger than in the diatomic liquids. The values of p are tabulated in table III and plotted against the field in figure 3.

TABLE III

The measured currents i in liquid argon (May 28, 1946) at 88°K divided by the saturation current I in gaseous argon, as a function of the fieldstrength							
F kV/cm	$\frac{i}{I}$	F kV/cm	$\frac{i}{I}$				
0.139	0.00187	6.95	0.0277				
0.348	.00367	13.9	.0406				
0.695	.00616	26.4	.0566				
0.681	.00616	38.4	.0686				
1.39	.00996	63.6	.0904				
2.08	.0132	87.8	.1050				
3.47	.0184	111	.1210				



Fig. 3. The ionization currents in liquid argon at 88 °K as a function of the field for positive (\odot) and negative (\odot) ions. The curves 1 and 2 are calculated with Kramers's formula for $p^* = /\psi(/^*)$, with $F_0 = 5000 \text{ kV/cm}$ and 1000 kV/cm, respectively.

33

2.33. Comparison of the results with the theory. The conditions for applying K r a m e r s's formula seem to be realised in this case also. Just as in the case of helium, however, no constant value of F_0 could be found (compare lower graph of figure 2). For strong fields F_0 may be chosen $F_0 = 5000$ kV/cm. With this value we calculated curve 1 in figure 3 with the formula (1.(11)) for p. When we use the value $F_0 = 1000$ kV/cm we get the curve 2 that coincides with the measured curve for weak fields. With the former value of F_0 and $N_0 = 4.01 \times 10^7$ we calculated $b_{\text{liq}} = 81 \times 20^{-7}$ cm. This corresponds with $b_{\text{l,g}} = 88 \times 10^{-7}$ cm, or $b_{\text{gas}} = 6.4 \times 10^{-3}$ cm, which is of the same order of magnitude as given by C1 a y and V a n K 1 e e f ⁶) for ionization by γ -rays.

2.34. *Currents of negative ions*. The currents of positive and negative ions proved to be practically the same within the limits of our accuracy. In table IV the data are collected.

The currents of positive and negative ions in argon							
F kV/cm	3.72	6.95	19.2	55.6	105.3		
$p_+ \times 10^4$ $p \times 10^4$	184 191	277 284	479 479	835 828	1170 1160		
p_/p_	1.03	1.02	1.00	0.99	0.99		

TABLE IV

2.4. The experiments in liquid helium at different temperatures.

2.41. The results between $4^{\circ}K$ and $1.3^{\circ}K$. In this section we shall give in more detail the results in liquid helium at different temperatures.

The results of a series with the uncovered sample (April 12, 1946) are given in the figure 4. Here the values of p are plotted against the field at different temperatures. From this graph we took the data for figure 5, in which for some effective fields *i.e.* $F_{\rm eff} = F \sin \varphi_0 = 0.8 F$, the currents are given as a function of temperature. In this diagram we have also inserted a series (February 24, 1944) with the covered sample at 85.0 kV/cm (*i.e.* $F_{\rm eff} = 39$ kV/cm, since we took $\sin \varphi_0 = 0.46$). The currents (of about 650 pA at the larger field) in the immediate neighbourhood of the λ -point are given in figure 6 for some series with the uncovered (at $F_{\rm eff} = 89.6$ kV/cm and 16.7 kV/cm) and with the covered sample (at $F_{\rm eff} = 12.5$ kV/cm).

Here we found large irregularities, the character of which seemed to vary strongly with varying field, but which in our experiments certainly were not reproducible. In the case of the covered sample and of ionization by X-rays the situation was quite similar. Since the physical properties of helium vary so abruptly at the λ -point, it is quite understandable that with our technique it proved actually difficult to establish in the immediate neighbourhood of this point a sufficient isothermal homogeneous state of the liquid at a sharply defined temperature between the electrodes.



Fig. 4. The ionization current of positive ions in liquid helium at different temperatures (uncovered sample).

 $\bigcirc T = 3.97^{\circ}\mathrm{K}, \ \boxdot T = 2.78^{\circ}\mathrm{K}, \ \bigtriangleup T = 2.34^{\circ}\mathrm{K}, \ \diamondsuit T = 1.90^{\circ}\mathrm{K}, \\ \bigtriangledown T = 1.30^{\circ}\mathrm{K}.$

Looking apart from the questionable irregularities of the current at the transition temperature, we conclude that no essential differences between the ionization currents in helium I and helium II have been found. Still we notice, that the values of the currents in helium II are at weak fields somewhat larger than at corresponding fields in helium I; at strong fields they are somewhat smaller.

2.42. A comparison with the results with X-rays. It may be of interest to compare our results with those, obtained with X-rays as



Fig. 5. The current as a function of temperature for different fields (uncovered sample). The numbers denote the values of the effective fields in kV/cm. Dotted line is a graph for the covered sample.



Fig. 6. The currents in the neighbourhood of the λ -point. Uncovered sample: $\bigcirc F_{\rm eff} = 89.6 \, {\rm kV/cm}$; $\triangle F_{\rm eff} = 16.7 \, {\rm kV/cm}$. Covered sample: $\bigcirc F_{\rm eff} = 12.5 \, {\rm kV/cm}$.

an ionizer. In table V we have collected the values of some currents of a series with the uncovered sample (i_u at 20.9 kV/cm), a series with the covered sample (i_c at 4.00 kV/cm) and a series with X-rays (i_x at about 20 kV/cm; comp. ⁱ)).

currents measured in a series with the covered sample (i_c , 4.00 kV/cm, May 5, 194 and a series with the uncovered sample (i_u , 20.9 kV/cm, $April$ 12, 1946), compare with a series measured with X-rays (i_x , 20 kV/cm), at different temperature							
T °K	i _c pA	i _u pA	i _x pA	i _c /i _x	$i_{\rm u}/i_{\rm x}$		
4.0	36	540	34	1.06	16		
2.8	32.5	490	30.5	1.06	16		
2.3		463	31	and a second	15		
2.2	31.5	466	31.5	1.00	15		
1.9		520	35		15		
1.4	(39)		35.5	(1.10)			
1.3	11 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1	540	36	AL LINE A	15		

TABLE V

The ratios of these currents in the fifth and the last column show, that apart from the temperature region in the neighbourhood of the λ -point, the currents are in fair approximation similar for *a*-particle and X-ray ionization.

2.43. The application of Kramers's theory. The application of Kramers's theory to the experiments at 1.3°K led to an analogous result as in helium I. Again no constant value of F_0 could be found, at strong fields the value $F_0 = 1200 \text{ kV/cm}$ (which is of the same order of magnitude as the F_0 -value in helium I) could be used for a an approximate description of the experimental curve. With this value of F_0 should follow $b_{\text{tig}} = 47 \times 10^{-7} \text{ cm}$, which is again of the same order of magnitude as the theoretical value for which we calculated: $b_{1,e} \approx 25 \times 10^{-7} \text{ cm}$ taking for b_{eas} the hydrogen value.

In a following section (2.53) we will show how a formal refinement of K r a m e r s's theory might give a natural explanation of the apparent failure in helium and argon.

We may notice here, that in view of the small coefficient of internal friction in helium II a large mobility of the ions might perhaps be expected. Whether this is the case cannot be deduced from our experiments, since the magnitude of the ionization current depends on the ratio of the ion mobility to the coefficient of recombination

(1.21). The experiments indicate that this ratio differs not very much in helium I and helium II. A direct measurement of the ion mobility would certainly be of interest.

The theories on the properties of helium II for which we refer to W. H. K e e s o m's "Helium"⁷) and which assume excited atoms in the liquid with mean free paths which are extraordinary large compared with those in the case of a normal liquid, would perhaps suggest larger currents than in normal liquids, since now collision ionization might occur. There is certainly no indication of such an effect.

2.5. A comparison between the results and their interpretation for the four liquids.

2.51. A comparison between the results. In this section we will first compare the results of the measured ionization currents and next we will try to suggest a natural explanation for the difficulties we met in understanding them theoretically.

We are interested in the following facts. When we apply K r amers's theory on the results in nitrogen and hydrogen where the currents were rather small (1), we found that this theory described the results within fair approximation, but that the empirical values of the diameter of the ion columns $(b_{\rm liq})$ that could be calculated with the formulae of the theory, were many times smaller than the *b*-values which theoretically might be expected when we extrapolate from the gases. Also an unexpected large difference was found between the *b*-values in the experiments with the covered and the uncovered sample, respectively.

In argon and helium, on the other hand, we measured much larger currents and Kramers's formula did not describe the results. When we tried to apply it formally, not a constant value of b_{hiq} was found, but one which decreased with increasing field. The order of magnitude of these *b*-values was however the same as the theoretically extrapolated values in gas.

The values p = i/I taken from the smooth curves relating to the uncovered sample, are plotted as a function of the effective field $(F_{\text{eff}} = 0.8 F)$ in table VI and as a function of the actual field in figure 7.

In table VII we recapitulate for the various cases the values of N_0 that were used for the calculation of $b_{\rm hiq}$ from the empirical values F_0 , the theoretical values $b_{\rm hg}$ and their ratios to $b_{\rm hiq}$.

Feff	nitrogen	hydrogen	argon	helium		
kV/cm	77°K	20° K	88°K	3.98°K	1.30° K	
5	0.00135	0.00342	0.0262	0.056	0.057	
10	.0022	.0051	.038	.096	.099	
15	.0030	.0064	.048	.126	.126	
20	.0038	.0082	.055	.149	.148	
- 30	.0051	.0111	.068	.184	.178	
40	.0064	.0140	.079	.212	.200	
50	.0078	.0167	.089	.234	.219	
60	.0089	.0192	.098	.259	.236	
70	.0104	.0216	.107	.268	.247	
80	.0116	.0241	.114	.282	.260	
90	.0128	.0264	.122	.294	.271	
100	.0140	.0285	.130	.305	.283	

TABLE VI



Fig. 7. The current as a function of the field in liquid nitrogen, hydrogen, argon and helium.

For argon and helium F_0 and $b_{\rm liq}$ have been computed from the measurements at highest fieldstrength, at fields about 10 times smaller the values of $b_{\rm liq}$ are larger by some 50% and 30%, respectively.

			See Asso			
Liquid	T °K	$N_0 \times 10^{-7}$	$F_0 imes 10^{-7}$ V/cm	$b_{ m liq} imes 10^7$ cm	$b_{\rm l,g} \times 10^{\circ}$ cm	$b_{\rm liq}/b_{\rm l,g}$
nitrogen (a)	77	2.57	11.5	2.5	30	0.08
nitrogen (b)	77 -	3.38	46	0.75	30	0.025
hydrogen (a)	20.4	0.77	4.25	1.8	24	0.075
hydrogen (b)	20.4	1.20	7.5	1.6	24	0.067
argon (a)	88	4.01	0.50	81	88	0.90
helium I (a)	3.98	0.66	0.09	75	(27)	(2.3)
helium I (b)	4.00	0,88	0.16	55	(27)	(2.0)
helium II (a)	1.30	0.75	0.12	47	(25)	(1.8)

1020	14.1	1003	0.010	
1.1.1.1	D 1	E	V 1.	

2.52. The applicability of the theory in nitrogen and hydrogen. Following J a f f é, K r a m e r s assumed an initial gaussian ion distribution along the path of the *a*-particle. The real situation in our liquid is much different however. From the path of the *a*-particle are ejected sideways small tracks of ions, that are made by a primary electron. The number of pairs of ions in each track depends on the initial energy of the electron and it may vary from one to ten. These branches may show small side-branches, if a secundary electron has enough energy to make one or more ion pairs itself.

We may describe this situation by the expression: feather ionization. In gases of usual temperature where a strong influence of diffusion compared with that of the recombination disturbs this feather picture, a gaussian ion distribution along the path of the α -particle will be established a short time after the formation of the column.

In our liquids, however, the featherlike distribution will persist during most of the lifetime of the column, as it will not markedly be disturbed by the influence of the diffusion which now is very small relative to that of recombination and field.

As a consequency of this, the justification for applying K r am e r s's theory seems lost. Still it would seem allowed to apply it in a *formal* way if we replace its original assumption of a single column with a definite N_0 and a definite b_{liq} by the assumption that we have to deal with a great number of such columns, each of which is characterized by its own N_0 and its own b_{liq} and which more or less may correspond to the branches in the featherlike model.

The current which is extracted per unit length from each column by the field is, when the diffusion is neglected, given by the expres-

41

sion of the form (1.(11)). Now this expression depends besides on F, only on the ratio $v_{\rm liq} = N_0/b_{\rm liq}$ which appears in the definition (1.(11)) for F_0 . Therefore, when the experimental results are to be interpreted on K r a m e rs's theory, we need in first instance only to introduce an average value $\tilde{v}_{\rm liq} = \overline{N_0/b_{\rm liq}}$, taken over all individual columns, which takes the place of the original $N_0/b_{\rm liq}$. It is clear that $\tilde{v}_{\rm liq}$ may easily be larger than the ratio $v_{\rm l,g} = N_0/b_{\rm l,g}$ which, assuming the J a f f é model in the gas is computed by the separate values N_0 and $b_{\rm l,g}$.

If our treatment is permissible, we see that in nitrogen for the covered sample $(\tilde{v}_{\rm liq})_{\rm cov} \approx 40 v_{\rm l,g}$ and in the case of the uncovered sample $(\tilde{v}_{\rm liq})_{\rm unc} \approx 12 v_{\rm l,g}$.

In hydrogen the corresponding ratios are $(\bar{v}_{\rm liq})_{\rm cov} \approx 15 v_{\rm l,g}$ and $(\bar{v}_{\rm hq})_{\rm unc} \approx 13 v_{\rm l,g}$, respectively. For the ratio $(\bar{F}_0)_{\rm cov}/(\bar{F}_0)_{\rm unc}$ we find 4.0 for nitrogen and 1.75 for hydrogen. The difference between the latter ratios can be brought in connection with the fact, that the B r a g g-ionization curve shows in gaseous nitrogen a relatively much broader lump near the end of the range than in the case of hydrogen. In fact this suggests that the distribution of *r*-values in hydrogen is not so much influenced by the covering foil as in nitrogen. It seems that such a situation agrees with what could be expected from the theoretical mechanism of ionization. Near the end of the range bigger effective N_0 -values and smaller effective *b*-values will appear than in the beginning.

The directions of the individual columns will not coincide with the directions in which the *a*-particles travel, but more or less perpendicular to them. This implies that in the case of the covered sample the assumption $\sin \varphi_0 = 0.46$ should only have a meaning near the end of the range. Both for the covered and uncovered sample we will therefore, as a first approximation, reckon with a random distribution, $\sin \varphi_0 = 0.80$.

Thus in the case of the covered sample our old values of F_0 and $b_{\rm liq}$ have to be multiplied (divided respectively) by a factor 0.80/0.46 = 1.7, so that we will now have the larger ratios $(\overline{F}_0)_{\rm cov}/(\overline{F}_0)_{\rm unc} = 7.0$ for nitrogen and 3.0 for hydrogen.

2.53. The applicability of the theory in argon and helium. Our consideration in (2.52) points towards a natural explanation of the fact that the K r a m e r s formula does not describe the experiments so

42

very well in argon and helium. In fact the ionization was considered as an average over many columns, each with their individual value of the quantity $v = N_0/b_{\text{liq}}$. In nitrogen and hydrogen this averaging process was replaced by inserting \bar{v} in the formula for ϕ (*i.e.* we put $\bar{p}(\bar{v}) = \phi(\bar{v})$). It can be demonstrated that in the present case, where the \dot{p} -values are so much larger, such a simple treatment is no longer allowed. This demonstration could be based directly on K r a m e r s formula. Now this formula is rather complicated, and we may also proceed in the following way:

As Prof. K r a m e r s pointed out to us, formula (1.(11)), in which the diffusion was neglected, can within fair approximation and looking apart from the saturation region, be described by

$$\phi = \frac{F}{F_0} \ln \frac{F_0}{F} \tag{1}$$

where F_0 does not differ very much from F_0 defined by (1.(11)) and is still proportional to ν .

Now averaging this formula over some *r*-distribution we get clearly with $(\overline{F_0})^{-1} = (\widetilde{F_0})^{-1}$:

$$p = \frac{F}{\tilde{F}_0} \ln \frac{\tilde{F}_0}{cF}, \qquad (2)$$

where c depends on the details of the distribution and is always bigger than 1.

If the individual ν -values may show ratios of the orders of magnitude 1 to 10, c may easily amount to several units.

Now in argon the measurements are pretty well described by (2) with c is about 5 and $\tilde{F}_0 = 1035 \text{ kV/cm} (F/\tilde{F}_0 \approx 10^{-1} \text{ at the highest}$ field), and those in helium with c is about 2 and $\tilde{F}_0 = 276 \text{ kV/cm} (F/\tilde{F}_0 \approx 0.5 \text{ at the highest field})$. Values of this order of magnitude will hardly have an influence in nitrogen where F/\tilde{F}_0 never exceeds 1×10^{-3} at the highest field, and an unimportant influence in hydrogen where the ratio is smaller than 6×10^{-3} .

2.54. A recombination effect in liquified inert gases. We have no reason to assume a picture of feather ionization in the inert gases different from that in nitrogen and hydrogen. Thus the large values of p are rather surprising. We are practically compelled to explain

them by the assumption of a coefficient of recombination in the liquified inert gases which is a good deal smaller than the theoretical value originally assumed.

In the computation of the formulae for p we had introduced the L angevin expression for a, the coefficient of recombination:

$$a=4\pi e^2\left(u_++u_-\right),$$

where u_+ and u_- are the mobilities of a positive and a negative ion, respectively.

Now a smaller recombination is formally expressed by a factor q < 1:

$$a = 4\pi q e^2 \left(u_+ + u_- \right).$$

In gases — except at small densities, where the mean free path of the ions and their kinetic energy (diffusion) diminish the probability for ions to recombine — this number equals 1. In our experiments where effects of this kind do not exist, we should also expect q = 1, quite analogous to the experiments with gases at large densities.

Under the assumption that in the cases of nitrogen and hydrogen q = 1 (which seems to be quite reasonable), the experiments lead to the assumption that in liquified inert gases we have to deal with a value of q which is much smaller than unity.

The formal consequence of such a smaller value of q is, that in the expression for $F_0 = 4\sqrt{\pi e} N_0/b$ the factor $v = N_0/b$ is replaced by qv. Since q is the same for each separate value of v, this means that in the graph for p versus F the scale of the F-values is to be stretched in the ratio 1/q, so that at a given field F we expect a larger value for p than with q = 1.

If we assume that the distributions of v are similar in nitrogen and argon, and that the same holds for hydrogen and helium, we deduce from the average $N_0/b_{\rm liq}$ (compare table VII) that $q \approx 0.05$ in argon and $q \approx 0.02$ in helium. We will not enter here on an explanation for this recombination effect. Suffice it to suggest the possibility that in the liquified inert gases the electrons which are liberated at the moment of ionization will not be attached by neutral atoms, so that in these liquids no heavy negative ions are present. Such a situation makes the considerations on which the L a n g e v i n formula with q = 1 is based, disputable.

Now the four gases under consideration are in general supposed to behave similar with respect to their electronegative property, i.e. in

all gases the possibility that an electron is attached to a neutral particle is very small. Clay⁸) has drawn attention to this fact and he proposed a modification of the Jaffé calculation on this account. A closer investigation of this electronegative property would be of interest, since in view of our suggestion large differences in this property should be expected in the liquified normal and inert gases, respectively.



REFERENCES

- A. N. Gerritsen and Jacqueline Koolhaas, Commun. Kamerlingh Onnes Lab., Leiden No. 266b; Physica, 's-Grav. 10, 49, 1943.
- 2) A. N. Gerritsen, Commun. Kamerlingh Onnes Lab., No. 275a; Physica, 's-Grav.
- Landolt-Börnstein, Phys. Chem. Tabellen, Eg HHb, Tabelle 161qq, 1315, 1935.
 - R. W. Gurney, Proc. roy. Soc. London A 107, 332, 1924.
- 4) Internat. Critical Tables 1, 102, 1926.
- 5) G. Jaffé, Ann. Physik, (4) 42, 303, 1913.
- 6) J. Clay and G. van Kleef, Physica, 's-Grav. 4, 651, 1937.
- W. H. Keesom, *Helium*, ed. by Elsevier, Amsterdam 1942, from which we also took the data for the densities of liquid helium.
- 8) P. H. Clay, thesis, Delft 1942.

CHAPTER III

THE STOPPING POWER OF A METAL FOR POLONIUM ALPHA-PARTICLES

(reprinted from: Physica, 's-Grav. 12, 311, 1946)

Summary

V on Weiszäcker's theory concerning the contribution of the conductivity electrons to the stopping of α -particles in a metal predicts a great influence of the temperature. In an attempt to complete V on Weiszäcker's theory Kronig and Korringa arrive at results from which the temperature dependence cannot be predicted with certainty. From the results of a few measurements with aluminium and tin as a stopping medium no influence of the temperatures.

3.1. Introduction. An α -particle penetrating a metal will loose its energy to the electrons of the metal. Studying the stopping processes one has to take into account that the electrons are bound. Completely free electrons can take over arbitrary quantities of energy from the α -particle, hence such electrons in the metal would cause an infinite stopping power.

In consequence of the interchange of energy between the latticeand the conductivity electrons the latter are, however, more or less bound and so the loss of energy by the α -particle remains finite.

The total stopping power can be divided into two parts:

a) the stopping caused by the electrons bound to the nucleus,

b) the stopping caused by the interaction with the conductivity electrons.

B o h r¹) and B e t h e²) have given formulae for the stopping due to *a*, but they have not taken into account separately the contribution of the conductivity electrons. This was done by V o n W e i s z ä c k e r³) who computed this contribution and was led to a correction to B e t h e's formulae for the total stopping power.

- 45 -

46 STOPPING POWER OF A METAL FOR POLONIUM &-PARTICLES

The total energy loss thus corrected is:

$$-\frac{\Delta\varepsilon}{\Delta x} = \frac{4\pi\varepsilon^2 E^2}{mV^2} NB \ln 10; \quad B = \Sigma s f_s \log \frac{2mV^2}{A_s} + z \log \frac{2mV^2}{h\gamma}.$$
 (1)

In these formulae are:

 $\Delta z/\Delta x$ the decrease of energy of the α -particle per cm path, E = 2e and V the charge and the velocity of the α -particle, m the electron mass, N the number of atoms per cm³, z the number of conductivity electrons per atom, j_s the oscillator sum of all possible electron transitions from the state of binding s to a level of higher energy, and A_s an appropriate average value of the change of energy that belongs to these transitions.

The coefficient γ is given by

$$\gamma = zN \frac{e^2}{m} \rho, \qquad (1a)$$

where ρ means the specific electric resistance of the metal. The inverse of γ has the dimension of a time and represents the time in which a current in a conductor without inductance decreases to 1/e of its original value: $i = i_0 e^{-\gamma t}$. In V o n W e i s z ä c k e r's theory γ may be compared with the time between two successive collisions of the electron with the lattice ions.

Since

$$\log \frac{2mV^2}{h\gamma} = \log \frac{1}{\rho} + const. \text{ and } \rho \sim T^p \quad (p > 1)$$
 (2)

the energy loss (1) will increase with decreasing temperature. Really free electrons (e.g. electrons in a superconductor) will take an infinite energy per cm path from the α -particle. The value of the change in the stopping power due to a change of temperature can be deduced from (1). We shall do this for the case of aluminium by computing the influence of a temperature decrease from 273°K to 20°K on the expression (1*a*) and for *B* and by comparing this with its experimental value at 273°K. According to Bethe this value is 18.7. The change in question is equal to $\log \gamma_{273°K}/\gamma_{20°K} = \log \rho_{273°K}/\rho_{20°K}$. Now the specific resistance at low temperatures depends very much on the purity of the sample. For the aluminium used in the experiments described hereafter, the ratio $\rho_{273°K}/\rho_{20°K}$ was experimentally found to be 16. Hence, as $\log 16 = 1, 2$ an increase of stopping power amounting to 6% per conductivity electron is to be expected.

STOPPING POWER OF A METAL FOR POLONIUM &-PARTICLES 47

After Von Weiszäcker a new method to calculate the stopping process has been proposed, which leads to quite different a result. Using throughout a hydrodynamic description Kronig and Korringa⁴) deduced a formula for the energy loss per conductivity electron

$$Q \simeq \frac{4\pi e^2 E^2}{m V^2} \ln \frac{\varepsilon^{3/4} V^2}{4\sqrt{\pi} \, \varrho \, \eta} \,. \tag{3}$$

Their suggestion is that the conductivity electrons can be treated as a fluid, with a density of charge (ρ) and a density of mass (ϵ). This negatively charged fluid moves with respect to the fixed substrate (the metal ions), and between them a frictional force acts which shows itself in an electric resistance of the metal. Besides this they suppose an internal frictional force in the fluid itself which can be described by means of a coefficient η . It is not known how η changes when the metal becomes superconductive. It might eventually be independent of, or only slightly dependent on temperature. In that case the change in the stopping power may be imperceptible.

As far as we know, a systematic search for the influence of temperature on the stopping power of α -particles is not mentioned in literature. Some years ago C a s i m i r started some experiments, which, however, gave no conclusive results. In the following we describe some experiments on aluminium and tin, performed in 1943 and 1944. They show that the effect of the temperature on the stopping power must be very small if it exists at all.

3.2. The exprimental method. The best way to measure a range is to count the number of α -particles passing the stopping layer. With the instruments for the electrostatic measurements at our disposal we could only perform ionization experiments. The electrostatic circuit has been described in 1.43⁵). We had two ionization chambers which could be used for a measurement with a tin- and an aluminium foil respectively.

3.21. The ionization chambers.

1) The first chamber had been used for ionization measurements in liquids with z-rays as an ionizer (a "longitudinal" chamber). This chamber has been described in chapter 1. We remind that this chamber consisted of two copper plates of different diameter which were placed parallel to each other at a variable distance. A little slab coated with polonium could be slid into the largest electrode. The

48 STOPPING POWER OF A METAL FOR POLONIUM α-PARTICLES.

other electrode could be connected with the compensation circuit and was surrounded by a guard ring at zero potential. The tin foil was pressed against the polonium. The two electrodes were placed within a copper cylinder in which hydrogen or helium could be condensed.

2) The second chamber used for measuring the stopping power in the aluminium was constructed especially for this purpose. It had to be built from parts of chambers which we had at our disposal. Besides the two electrodes e_1 and e_2 (see fig. 1) there were two copper



Fig. 1. Apparatus for measuring the range equivalent of an aluminium foil in gas atmosphere.

K the ionization chamber with electrodes e_1 and e_2 ; F the aluminium foil;

Po the polonium coating on a metal disk e_1 ;

c, g, k insulation material.

grids. The lower grid b was kept at earth potential, whereas the upper one a was connected with the tension circuit. Thus the measuring chamber K was protected by a chamber V, in which the field is practically the same but opposite to that in K. This prevents ions formed outside of K from contributing to the current in K. An aluminium foil F was placed at a distance of 14.24 mm from these chambers (fixed by means of two rings j). It was kept in position by a flat ring j on a thick ring of copper l at 2.50 mm distance from the polonium Po, so that the total distance which the α -particle had to travel through gas from Po to a was 16.64 \pm 0.08 mm. Our measurements with the two ionization chambers described should be interpreted in the following way.

3.22. Interpretation of the measurements.

a. Suppose that the loss of range by traversing a metal foil is given by the part OA of the Br a g g ionization curve in fig. 2. With

STOPPING POWER OF A METAL FOR POLONIUM &-PARTICLES 49

an ionization chamber as described, sub 1) the area of the curve between A and C will be measured provided the chamber is sufficiently deep. If thes topping power of the foil increases (which means that the line AA must be displaced to BB), only the area BCwill be measured and the ionization current decreases. For our purpose this method is not very suitable. It was incidentally applied to tin at the occasion of an ionization measurement in liquid helium where a tin foil was used for lowering the intensity of the activity of the polonium sample.



Fig. 2. The positions of the extrapolated ionization range (Λ_i) , of the range equivalent (λ_m) of the foil with a supposed change from AA to BB, and of the ionization chamber K relative to the Bragg curve for helium gas.

b. With the second chamber described sub 2) the density of the gas can be chosen so high that no α -particle reaches the shallow chamber K (K_2 , fig. 2). When the pressure is sufficiently lowered the shape of the curve between B and C will be flattened and stretched past K (or, leaving the curve unchanged, the effective position of the chamber is moved from K_2 to K_1 , to the left of C), so that an ionization current can be measured. The chamber has to be shallow if we wish to measure the specific ionization with some accuracy so that precise data about the extrapolated ionization range (Λ_i) can be obtained.

3.3. The results for tin and aluminium.

3.31. The measurements and the results for tin. The temperature at which tin becomes superconductive is 3.72° K (*i.e.* in the temperature

50 STOPPING POWER OF A METAL FOR POLONIUM α-PARTICLES

region obtained with liquid helium), so that this metal is well suited for the investigation of an eventual influence of superconductivity on the stopping process. In this temperature region it is difficult, however, to measure the range by means of chamber 2 with helium gas as a stopping medium. Our actual measurements constitute only a byresult of an investigation on ionization currents in liquefied gases. We have used for this purpose chamber 1 filled with liquid helium.

The polonium layer which emitted the α -particles was covered by a tin foil of about 13 μ thick, which is equivalent to $\lambda_m = 0.709 \Lambda_i$. Ionization currents were measured at various temperatures below and above the transition point of the tin foil (from 3.77°K to 1.42°K).

Some currents were measured three times, first some time after adjusting the temperature of the liquid, then when an external magnetic field was applied so as to disturb the superconductivity, finally in the superconductive state again. The strength of the disturbing field was always higher than the critical field strength at the lowest temperature ⁶). The superconductivity was checked with some samples cut from foils from 6 to 20 μ thickness.

The results of the measurements are given in table I. The distance between the electrodes was 0.020 cm except for the point at 2.38°K where the distance was 0.060 cm. The field strength in these two cases was 84.9 kV/cm and 97.7 kV/cm respectively.

The current in liquid helium at different temperatures below and above the transition point of tin								
Т	3.77	3.58	2.92	2.25	2.14	1.86	1.42	2,38°K
H = 0	524	516	497	489	573	593	596	617 pA
$H > H_k$	526	516	497	487	573	597	598	610 pA
H = 0	526	516				593 pA		

TABLE I

We see from this table that the result is negative: within the limits of accuracy, or rather, of reproducibility (1%) we find no difference in the current whether the foil is superconductive or not.

To interprete this result in terms of an eventual change in the value of the range, we have to estimate the rest range as a function of the current. The range equivalent of the tin foil λ_{A1} was as great as 0.709 Λ_i , so the rest range of the α -particle was 0.291 Λ_i . Now we see from the Bragg curve in helium that at the point $\lambda = 0.291 \Lambda_i$ a change of 1% in the total current corresponds to the same relative

change of the rest range, which means a change of 0.4% in the range equivalent of the tin foil.

So our final conclusion is that the change in the stopping power of tin at the transition into the superconductive state is less than 0.4%. This holds not only at the transition temperature itself, but also at lower temperatures.

3.32. The measurements and the result for aluminium. In our second apparatus 1 we used helium gas as a stopping medium, but since a density of 2.5 was required to prevent the α -particles from reaching the measuring chamber K and since the apparatus was not constructed for high pressures, we had to use nitrogen in stead of helium at temperatures higher than 180°K. The aluminium foil had a



Fig. 3. Current-density curves at 90.5°K (a), 20.2°K (b) and 13.3°K (c).

thickness of 18.4μ (corresponding to a range equivalent $\lambda_{AI} = 0.818 \Lambda_{II}$ for polonium) so that our measurements covered the whole region of the top of the Bragg curve which lies in the last 13%. At each measurement, after adjusting the temperature of the bath, so much gas was introduced that no ionization current could be observed. Then the pressure was decreased by small steps, and each time the ionization current was measured. From the temperature of the bath and the pressure of the gas the density of the latter could be determined. The curves that were obtained from these measurements could be fitted into the known Bragg curve. In fig. 3 three of the measured current-density curves are given, for 90.5°K, 20.2°K and

52 STOPPING POWER OF A METAL FOR POLONIUM &-PARTICLES

13.3°K. We see from these curves that a linear extrapolation along the tangent to the curve at half the top value is possible. This extrapolation may be effected for a curve that is obtained from saturation currents in the ionization chamber (V > 200 V, that is F > 1000 V/cm) as well as for currents registered in the case of non-saturation (50 V, that is F = 250 V/cm). An accidental discharge somewhere in the apparatus was the cause of a bad determination of the top in the curve at 90°K.

For comparison we have expressed the results obtained with nitrogen also in terms of helium. The ratio of a current in helium to that in nitrogen is 1.2^{7}) and the ratio of the ranges in nitrogen and in helium we find from their values 3.89 cm^7) and 21.5 cm^8) respectively (normal pressure, 15° C). To calculate the range equivalent λ_{AI} of the foil we use the formula

$$\Lambda_{\rm iHe} = A \ \rho + \lambda_{\rm A1} \left(T \right) \tag{4}$$

in which $\Lambda_{i\text{He}}$ means the extrapolated ionization range in helium gas at 273°K and 760 mm pressure ($\Lambda_{i\text{He}} = 20.38 \text{ cm}^8$)), ρ the corresponding relative density of the gas and A the distance between the polonium surface and the grid a of the ionization chamber K: A = 1.66 cm at 273°K.

From (4) we see that a change in the stopping power of 0.1% will not escape observation. At 20°K the helium equivalent of the foil proved to be about 16.7 cm. As $\Delta \lambda_{AI} = -A \Delta \rho$, $\Delta \lambda_{AI} = 0.01$ cm corresponds to $|\Delta \rho| = 0.006$. Since $\rho = 1$ at 20°K corresponds to a pressure of 5.57 cm, this $\Delta \lambda_{Al}$ would correspond to $\Delta p = 0.033$ cm Hg which is easily measured indeed. The results have been collected in table II. The numbers in the first column indicate the chronological order in which the measurements were performed. The fourth column gives the values of the ionization currents I in the top of the Bragg curve in arbitrary units. They are the same in the measurements 1 to 5, but in 6, 7 and 8 they are smaller. This is presumably due to a contamination of the polonium coating or of the foil. In fact the measurements at 90°K with helium gas were preceded by a measurement at room temperature on the same day and it is possible that the ionization chamber had not been sufficiently exhausted or washed with dry gas. The next day the two measurements 7 and 8 were performed and the apparatus had meanwhile not been warmed to room temperature. The contamination may have been water or oil.

STOPPING POWER OF A METAL FOR POLONIUM &-PARTICLES 53

The number and the position of the measured points on the current density curve do not permit a very precise extrapolation to the density that belongs to the extrapolated ionization range and therefore three persons have independently drawn the curves through the points and performed the extrapolation. The values of the extrapolated ionization range λ_{A1} obtained thus are collected in the columns a, b and c of table II. The last column but one gives the mean values $\overline{\lambda}_{A1}$ of these numbers. The dispersion of λ_{A1} about the mean values $\overline{\lambda}_{A1}$ proves to be very small and does not surpass 0.3%. In the last column the ratio of $\overline{\lambda}_{A1}$ to its value at 294°K is given. Only in the case of measurements 6, 7 and 8, which anyhow were questionable, we find deviations surpassing 1/2%.

No.	T	gas	I	λ_{A1}			-	$\lambda_{A1}(T)$
	°К	Bank	pA	a	b	G	XAI	λA1(294° K
5	294	N ₂	33.0	16.57	16.58	16.57	16.57	1
7a	169.4	N ₂	24.7	16.90	16.92	16.88	16.90	1.020
7b	169.4	He	26.7	17.01	17.02	16.97	17.00	1.026
8	142.6	He	26.3	16.81	16.88	16.83	16.84	1.016
6	90.5	He	28.8	16.60	16.58	16.55	16.58	1.000
i	77.4	He	32.6	16.59	16.65	16.58	16.61	1.002
2	63.4	He	32.6	16.65	16.71	16.63	16.66	1.005
3	20,4	He	32.8	16.59	16.60	16.51	16.56	1.000
4	13.3	He	32.9	16.58	16.58	16:63	16.60	1.002

A 44 1	ALC: NAME	100		
1000	1.10	1000	200	1.7
1000	4.155			
100.00	Sec. 3. 1	1000		

In the calculation of λ_{A1} the change of the distance A caused by the cooling of the apparatus is neglected. Using the value for the dilatation coefficient of brass 1.85×10^{-5} and supposing that this coefficient is constant between 273° K and 100° K, A will in this interval decrease only by 0.07 cm and this means that the largest correction to $\overline{\lambda}_{A1}$ due to the dilatation will be smaller than 0.1%. Inspection of the current-density curves (fig. 3) proves that the shape of the curves is unchanged within the limits of our accuracy over the whole temperature region from 294°K to 20°K.

3.33. Conclusions. It is clear that the measurements described are open to criticism and that it would have been possible to increase the accuracy in several respects. On the other hand it seems safe to conclude that neither the transition in the superconductive state nor

54 STOPPING POWER OF A METAL FOR POLONIUM *x*-PARTICLES

a lowering of the temperature has any influence on the range of α -particles higher than $1/2^{0}/_{0}$. This is in accordance with the negative results of M c L e n n a n's measurements with swift electrons' passing through a foil of superconductive lead 9).

Our results are in contradiction with the theory of Von' Weiszäcker, which for aluminium leads to expect a change of some 6% per conductivity electron when the temperature decreases from 300° K to 20° K. No conclusion can be drawn in regard to the theory of Kronig and Korringa since the temperature dependence of the coefficient of internal friction is not known.

Prof. K r a m e r s informed us, that the theory concerning the stopping process in metals can be brought into accordance with the absence of a temperature influence on the stopping power if the effect of polarization of the metal is introduced 10).



REFERENCES

- 1) N. Bohr, Phil. Mag. (6) 25, 10, 1913.
- 2) H. A. Bethe, Ann. Physik (5) 5, 325, 1930.
- 3) C. F. von Weiszäcker, Ann. Physik (5) 17, 869, 1933.
- 4) R. Kronig and J. Korringa, Physica, 's-Grav. 10, 406, 1943.
- A. N. Gerritsen and Jacqueline Koolhaas, Commun. Kamerlingh Onnes Lab., Leiden No. 266b; Physica, 's-Grav. 16, 49, 1943.
- W. J. de Haas and A. D. Engelkes, Commun. No. 247d; Physica, 's-Grav. 4, 325, 1937.
- Cf. Landolt-Börnstein, Physik. chem. Tab., dritter Ergänzungsband II, 1315 (161qq) and 1306 (161gg), 1935.
- 8) G. A. W. Rutgers, Thesis for the doctorate, Utrecht 1940.
- J. C. McLennan, Trans. roy. Soc. Canada 23 (III) 269,1929 and Journ. Inst. mech. Eng. 75, 693, 1934.
- 10) H. A. Kramers, Commun. Suppl. No. 96a; Physica, 's-Grav. 13, 401, 1947.

Uit enige op verzoek van prof. dr W. J. de Haas verrichte ionisatieproeven met röntgenstralen in vloeibaar helium, volgde de noodzaak een onderzoek in te stellen naar het gedrag van ionisatiestromen in vloeistoffen bij lage temperaturen, met een bekende intensiteit van de ionisator in de te ioniseren ruimte.

De resultaten van dit onderzoek worden in dit proefschrift beschreven.

Over ionisatiemetingen in gassen zijn vele uitvoerige publicaties verschenen, die niet zelden door grote nauwkeurigheid der beschreven resultaten de aandacht hebben getrokken: wij denken hierbij vooral aan de proeven van prof. dr J. Clay en medewerkers.

Het is dan ook niet zonder enige schroom dat wij minder nauwkeurige resultaten van metingen van ionisatiestromen, veroorzaakt door *a*-deeltjes in vloeibare stikstof, waterstof, argon en helium mededelen. Bij het meten van ionisatiestromen in deze vloeistoffen treden vele storende nevenverschijnselen op, zodat het zeer moeilijk zal zijn bij deze metingen eenzelfde nauwkeurigheid als in gassen te bereiken. Voor een overzicht en een critische beschouwing van ionisatiemetingen in gassen zij naar het proefschrift van P. H. Clay verwezen.

a. De resultaten van de proeven. In het eerste hoofdstuk wordt het eenvoudige ionisatievat beschreven waarmede de metingen werden verricht. De vloeistof tussen de electroden in het ionisatievat werd door de *a*-deeltjes van een in een der electroden geschoven koperplaatje met polonium geïoniseerd.

Het bleek tengevolge van polarisatie der electroden niet mogelijk enigszins reproduceerbare metingen te verrichten wanneer de sterkte van het polonium, uitgedrukt in de waarde van de verzadigde ionisatiestroom in gasvormige lucht 10⁻⁸ A bedroeg. Nadat de sterkte van het polonium tot ongeveer een derde van de oorspronkelijke waarde was verminderd door dat er een dun blaadje aluminium of tin op

- 55 -

werd gelegd, bleek het mogelijk de stromen met een nauwkeurigheid van enkele procenten te meten.

Door afzonderlijke metingen, welke in het derde hoofdstuk worden besproken, hebben wij aangetoond, dat het remmend vermogen van de gebruikte blaadjes metaal niet van de temperatuur van het metaal afhangt. Dit resultaat is in tegenstelling met een voorspelling van V o n W e i s z ä c k e r, welke op theoretische overwegingen was gebaseerd.

In het eerste hoofdstuk worden de metingen der ionisatiestromen in afhankelijkheid van de electrische veldsterkte tussen de electroden voor vloeibare stikstof en waterstof beschreven, in het tweede hoofdstuk voor vloeibaar helium en argon.

De stroom-veldkrommen zijn voor iedere vloeistof verschillend van vorm. Het quotient p van de gemeten stroomsterkte en de in gas gemeten verzadigingsstroom bedraagt bij een veld van 100 kV/cm tussen de electroden: 0,014, 0,028, 0,130 en 0,305 voor stikstof, respectievelijk waterstof, argon en helium. Deze getallen zijn ontleend aan een reeks metingen met een niet door bladmetaal bedekt poloniumneerslag, waarvan de ionisatiesterkte ongeveer 2,5.10⁻⁹ A in lucht bedroeg (vergelijk fig. 7 in het tweede hoofdstuk).

Het verband tussen de stroomsterkte en het veld is voor stikstof en misschien voor waterstof in een groot veldgebied lineair, voor argon en helium is hiervan geen sprake.

Metingen bij verschillende temperaturen in eenzelfde vloeistof hebben aangetoond, dat de invloed van de temperatuur voor het grootste deel aan de veranderingen in de dichtheid van de vloeistof kunnen worden toegeschreven.

Behalve in waterstof, waarvoor het effect slechts 3% bedraagt, is in geen der vloeistoffen een verschil groter dan de meetnauwkeurigheid tussen de stromen van positieve en negatieve ionen gevonden.

De stromen en de overeenkomstige p-waarden zijn voor de metingen met het bedekte polonium steeds kleiner dan in het geval van de metingen met het onbedekte preparaat, met ongeveer dezelfde ionisatiesterkte. Deze verschillen zijn groter dan uit beschouwingen over de gemiddelde hoek tussen het veld en de richting der *a*-deeltjes kan worden verwacht.

De resultaten in helium II verschillen weinig van die in helium I. De onregelmatigheden in de omgeving van het λ -punt moeten waar-

schijnlijk daaraan worden geweten, dat de vloeistof in de omgeving van de overgangstemperatuur niet in evenwicht verkeert.

b. *Theoretische beschouwingen*. De beweging der ionen wordt beschreven door een partiële differentiaalvergelijking van de tweede orde, waarin de diffusie- en de recombinatie coëfficiënten en de grootte van het electrische veld als coëfficiënten voorkomen.

In het eerste hoofdstuk worden de oplossingen van deze bewegingsvergelijking, zoals zij door J affé en door Kramers zijn afgeleid, vergeleken. In het algemeen kan worden verwacht dat de door J affé gegeven oplossing niet de toestand in vloeistoffen zal beschrijven, daar hij de invloed der recombinatie steeds klein ten opzichte van die der diffusie onderstelt. Daar Kramers bij het oplossen juist de invloed van de diffusie als klein beschouwt, biedt zijn oplossing voor metingen in vloeistoffen bij lage temperatuur meer perspectief. Desondanks blijkt zij slechts voor stikstof met de resultaten der metingen binnen de meetnauwkeurigheid overeen te stemmen. In waterstof is de overeenstemming minder goed, in helium en argon schijnt de oplossing te falen.

De aanpassing tussen de oplossing van J affé en de meetresultaten wordt voor gassen verkregen door op geschikte wijze een parameter te kiezen, welke wordt geïnterpreteerd als de gemiddelde diameter van de ionenkolom. In de oplossing van K r am er s komt een parameter voor met de dimensie van veldsterkte, welke geschreven wordt als het product van enkele constanten en de verhouding tussen de lineaire ionendichtheid en de diameter van de kolom. Bij een bepaalde keuze van deze parameter kan de bijbehorende waarde van de kolomdiameter worden berekend. Wanneer dit voor onze metingen wordt uitgevoerd, dan blijken de waarden van de met K r a m e r s berekende kolomdiameters zowel in stikstof als in waterstof aanmerkelijk kleiner te zijn dan de uit de gasmetingen te verwachten waarden. Bovendien zijn de kolomdiameters kleiner voor de metingen met het bedekte dan voor die met het onbedekte polonium.

In het laatste deel van het tweede hoofdstuk trachten wij deze verschillen te verklaren. Het moge zijn, dat in gassen het beeld van de kolom-ionisatie met de werkelijkheid overeenkomt, in vloeistoffen en zeker in vloeistoffen bij lage temperatuur zal dit beeld misleidend zijn.

De aard van het ionisatieproces brengt met zich mede, dat de verspreiding der ionen langs de baan van het ioniserend deeltje steeds een veervormige structuur vertoont. In deze structuur, die in gassen door de aanwezigheid van de diffusie spoedig in een min of meer regelmatige kolom overgaat, zullen van plaats tot plaats sterk verschillende waarden van de lineaire ionendichtheid en van de breedte van de afzonderlijke kolommetjes langs de banen der primair- en secundair ioniserende deeltjes optreden.

Dit veervormige model leidt tot het formeel invoeren van een verdeling langs de baan van het ioniserend deeltje van de grootheid v, die het quotient van de *plaatselijke* lineaire ionendichtheid en de *plaatselijke* waarde van de kolomdiameter voorstelt.

Wij hebben er van af gezien een theoretisch quantitatief berekend model van deze verdeling te ontwerpen. Uit de metingen in stikstof en waterstof kan alleen worden geschat, dat plaatselijke verschillen in v zeer groot kunnen zijn en dat een gemiddelde waarde van v in deze vloeistoffen 40-, respectievelijk 15-maal de waarde die door extrapolatie uit de gastoestand zou volgen, kan bedragen. Daar voor argon en helium door de grote waarden van ϕ niet met een gemiddelde waarde van v mag worden gerekend, tonen wij hiervoor aan dat met het geschetste model een schatting van de correctie op de oplossing van K r a m e r s mogelijk is, waardoor deze met de resultaten in overeenstemming is te brengen.

Uit de litteratuur is bekend, dat in de uitdrukking van L ang evin voor a, de coëfficiënt van recombinatie:

$$a = 4\pi q e^2 \left(u_+ + u_- \right) / \varepsilon$$

(e is de lading van een ion, u_+ en u_- zijn de bewegelijkheden der positieve en negatieve ionen, ε is de dielectrische constante) de coëfficiënt q gelijk is aan zijn theoretisch maximale waarde 1 voor gassen onder enkele atmosferen druk, dan zal namelijk de diffusie de kans op recombinatie niet meer verminderen.

Uit de metingen in argon en helium volgt in tegenstelling tot de verwachting, dat in deze vloeistoffen een recombinatie optreedt, die met $q \ll 1$ moet worden beschreven. Het vergelijken met de resultaten in stikstof en waterstof leert, dat q van de orde van grootte 0,02, respectievelijk 0,05 in helium en argon zal kunnen zijn.

Ik wil op deze plaats mijn hartelijke dank betuigen aan mijn vrouw, wier toegewijde hulp tijdens de metingen van grote waarde is geweest; en aan het technisch personeel van het Kamerlingh Onneslaboratorium voor de medewerking bij deze en andere onderzoekingen voortdurend ondervonden.

Zonder de samenwerking met prof. dr H. A. Kramers en de leiding die hij mij bij de theoretische problemen heeft gegeven, zou het mij nimmer mogelijk zijn geweest uit de met elkaar schijnbaar strijdende meetresultaten een enigszins gefundeerde theoretische verklaring te verkrijgen. Ik dank u voor uw belangstelling en voor de prettige uren in wetenschappelijk gesprek doorgebracht.

Mijn leermeester, prof. dr W. J. de Haas ben ik dank verschuldigd voor de verschillende onderzoekingen welke ik onder zijn leiding in het Kamerlingh Onnes-laboratorium heb mogen verrichten.

Tenslotte wil ik bij de wetenschappelijke bekroning van mijn universitaire opleiding mijn moeder grote dankbaarheid betuigen voor haar zorg, waardoor zij mij het volgen van deze opleiding mogelijk heeft gemaakt.







STELLINGEN

Ι

Uit bepalingen van de stroom-veld krommen met α -deeltjes van verschillende dracht, kan proefondervindelijk een beeld van de v-verdeling langs de baan van het α -deeltje worden verkregen.

Dit proefschrift, bladzijden 43 en 44

Het meten van de bewegelijkheden van ionen in vloeibaar helium en andere vloeibare gassen is wenselijk en is waarschijnlijk reeds met een kleine intensiteit van de ionenbron mogelijk.

II

III

De uitkomst van de berekeningen van Kramers over de invloed van de temperatuur op het remmend vermogen van een metaal, zou ondanks het optreden van de drachtspreiding, aan lithium proefondervindelijk kunnen worden getoetst.

H. A. Kramers, Physica, 's-Grav. 13, 401, 1947.

IV

Tengevolge van de polarisatie der electroden zullen diamanttellers, of tellers gevuld met vloeibare gassen, slechts voor het meten van die verschijnselen kunnen worden gebruikt, welke zich met zeer korte perioden herhalen.

D. E. Woolridge, A. J. Ahearn and J. A. Burton, Phys. Rev. 71, 913, 1947. Dit proefschrift, bladzijde 15.

V

Het is voor het verkrijgen van inzicht in het mechanisme en de oorzaken van de polarisatieverschijnselen, welke optreden bij proeven met ionisatiestromen door niet geleidende vloeistoffen noodzakelijk, deze proeven uit te voeren met volkomen zuivere electroden van verschillend materiaal.

VI

De resultaten van metingen van de warmtegeleiding van vast normaal en zwaar methaan kunnen een inzicht verschaffen in het mechanisme van de orde - wanorde overgangen in deze stoffen. A. N. Gerritsen and P. van der Star, Physica, 's-Grav. 9, 503, 1942.


VII

De door Frederik beschreven adaptometer voor het constateren van nachtblindheid onder uitsluiting van de mogelijkheid tot simulatie van de proefpersoon, zal, in tegenstelling tot de adaptometer van Kentgens, waarschijnlijk niet aan deze voorwaarde voldoen. W. S. Frederik, Philips Techn. Tijdschr. 9, 211, 1947.

S. K. Kentgens, Proefschrift, Utrecht 1938.

VIII

Het invoeren van de quantumvoorwaarde voor het impulsmoment van het electron door middel van de debrogliegolflengte is didactisch misleidend.

J. A. Prins. Grondbeginselen van de hedendaagse natuurkunde, 4e druk, blz. 245.

IX

Zolang aan instituten voor medisch speurwerk geen natuurkundigen zijn verbonden, zal een korte natuurkundige opleiding der medische onderzoekers, bijvoorbeeld na het artsexamen, zeer wenselijk zijn.

Х

De waarde van een natuurkundig practicum zal stijgen, naarmate de verhouding tussen de aantallen "standaard"- en "constructie"proeven ten gunste van de laatsten toeneemt.

XI

Lezers van periodieken blijken advertenties welke op de rechter pagina zijn geplaatst beter te reproduceren dan advertenties op de linker pagina. Deze voorkeur voor de rechter pagina is niet gebleken met met tekst bedrukte kaarten. De grotere opmerkzaamheid zal een gevolg zijn van een onwillekeurige leespauze voor het omslaan der rechter bladzijde; deze pauze zou uit filmopnamen van te voren niet ingelichte lezers kunnen worden bepaald.

F. Roels, Psychologie der reclame Amsterdam, 1938, blz. 171.

XII

Bij het beschikbaar stellen van gelden voor de uitbreiding van het zuiver wetenschappelijk onderzoek zullen tevens de mogelijkheden voor financiele steun bij de opleiding van technische hulpkrachten moeten worden uitgebreid. trag van anderhinderid under vielenting van de anorskelent int an makin van de proeferansen aal, basejenaste beginnen de anorskelenter ener Kastigena, waardinaaten aal, basejenaste beginnen in seitenen ener Kastigena, waardinaaten inte ener hene van trage van seitenen energena kastigena waardinaaten interes van trage van seitenen in trage van seitenen van seitenen van seitenen van seitenen in seitenen in trage van seitenen van seitenen van seitenen van seitenen in seitenen in trage van seitenen van seitenen van seitenen van seitenen in seitenen van seitenen in seitenen van seitenen v

JIV

van het siestene van de generententer en en de hetendende van het interdienen. Van het siestene date mitdak van de delengtingelikenne is datas-

20

evenue united that was particle becautered averaging the interaction of the second state of the second sta

70

De mande son een mittelenden practicus of sigger, onegante of

12

terson une persolution bision advertenties veille no de maler qui que ais person bares la reproductor de activit perior a sin qui bisina rev sauter. Dass conducts ven de rechter perior a sin qui bisina con sei relati balandes beres parte per la filles promes ven la restant der rechte basinable, deve parte per et il filles present ven in vons der rechte basinable, deve parte per et il filles present ven in vons der rechte basinable, deve parte per et il filles present ven in vons der rechte basinable, deve parte per et il filles present ven in vons sin ingelieble brenn businable personable la present ven in vons der rechte basinable, deve parte per basinable la present ven in vons der rechte basinable, deve parte per basinable la present ven in vons sin ingelieble brenn businable personable la present ven in vons sin ingelieble brenn businable personable la present ven in vons sin ingelieble brenn businable personable la present ven in vons sin ingelieble brenn businable personable la present ven in vons sin ingelieble brenn businable personable pe

IN M

In her hearing have maken was usiden voor de uitbruding van het aanter aerenscharpelijk conternati milien terven de moeren lien voor termeriste steve hit de opleting was remetable belgenericen antern voorbe uitretuent



