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F. Scaramuzzi: FURTHER MEASUREMENTS OF IONIC MOBILITIES IN SUPERFLUID HELIUM BY THE METHOD OF THE COMPLETE SPACE CHARGE LIMITATION. -

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INTRODUCTION.

In a previous communication[1], henceforth referred to as BS, a new method for the measurement of ionic mobilities was presented. It was pointed out that this method is limited by technical reasons to mobilities not greater than ∿ 2 cm^2/ /volt.sec. The results of measurements made for positive ions in superfluid helium were reported and analyzed in the light of present knowledge about this subject.

In this paper further measurements made by the same method are reported. Still considering superfluid helium, they have been extended to negative ions. Furthermore, the temperature region close to the lambda-point (2.18°K) has been accurately scanned for both positive and negative currents in order to precisely determine the anomalous temperature dependence of the mobility in this region.

The validity and accuracy of the method are discussed, and the possibility of improving it to obtain a better absolute method is examined.

THE METHOD.

It will be helpful to the reader to briefly review the method, which has been described in detail in BS.

The apparatus consists of a simple diode whose dielectric is liquid He^4. It has two main electrodes (a guard ring is also used to avoid boundary effects). One of the electrodes has a Po^{210} source deposited on it. The alpha-particles of polonium have a range of ~ 0.2 mm in He^4 liquid, so they produce a highly ionized layer close to this electrode. An electric field of the proper sign can draw positive or negative ions out of this layer. The ions move through the liquid helium and are collected on the second electrode. The current passing through the helium can be measured as a function of the applied potential.

"Complete space charge limitation" is defined by the vanishing of the electric field at the emitting electrode. In this case Poisson's equation can be easily solved for simple geometries[2]. It gives

$$i = a\mu V^2, \tag{I}$$

i.e., the current measured is proportional to the mobility μ and to the square of the applied potential V. a is a coefficient which takes into account the geometrical parameters of the system. For a cylindrical geometry (coaxial infinite cylinders), like that used in the present research, equation (I) becomes:

A) Internal emitter ($r_o < r$),

$$i = \frac{\mu}{2} \frac{V^2}{r^2 R}, \tag{I_A}$$

where

$$R = \left\{ \left[1 - \left(\frac{r_o}{r}\right)^2 \right]^{1/2} - \left(\frac{r_o}{r}\right) \cos^{-1} \left(\frac{r_o}{r}\right) \right\}^2$$

B) External emitter ($r_o > r$),

$$i = \frac{\mu}{2} \frac{V^2}{r_o^2 R} \tag{I_B}$$

where

$$R = \left\{ \left[1 - \left(\frac{r}{r_o}\right)^2 \right]^{1/2} + \ln \frac{r/r_o}{1 + \left[1 - (r/r_o)^2 \right]^{1/2}} \right\}^2$$

These formulas are in electrostatic c.g.s. units. i is the current per unit axial length of the electrodes. r_o is the radius of the emitter, and r the radius of the collector.

To verify that the condition of "complete space charge limitation" applies, one checks that the plot of i versus V^2 is a straight line. The mobility is then evaluated from the slope $s = i/V^2$ of such a straight line.

Two different ways can be followed to calculate the mobility:

1. By introducing in equation (I_A) or (I_B) the values of the geometrical parameters of the diode used (r and r_0), it is possible to calculate the value of the constant a of equation (I). Knowing both a and $s = i/V^2$, the mobility value is immediately obtained. In this way the method is an absolute one.

2. Since for liquid He^4 II other mobility measurements exist, obtained by other techniques (3 ÷ 7), it is possible to calibrate the whole experimental set-up (including the geometrical constant a and the current detecting devices), by introducing in equation (I) a known value of the mobility together with the value of $s = i/V^2$, measured with the present technique under the same physical conditions (i.e., at the same temperature, pressure, etc.). This enables one to derive a value of a, which is now an instrumental (not only geometrical) constant and takes into account all the causes of systematic errors. This value of a can then be used to deduce the values of the mobility for other physical conditions, for example at different temperatures. In this way the method is not absolute.

In this paper we decided to follow the second of these two methods, because no particular care was taken in designing the experiment to avoid systematic errors. However, in the last chapter an "a posteriori" comparison of the two methods is made and turns out to be rather satisfactory.

THE EXPERIMENT.

In BS reliable results were obtained for a cylindrical geometry (diode A): the inner cylinder (4 mm diameter) was the emitter, the outer (12 mm internal diameter) was the collector. Further measurements have been made with another cylindrical diode (diode B), in which the emitter is the external cylinder (internal diameter 20 mm) and the collector is the internal cylinder (10 mm diameter). For both diodes, guard electrodes were provided in order to be able to regard the geometry as infinite in the direction of the cylindrical axis. In diode A the lenght of the collecting electrode was 20 mm, in diode B it was 2 mm. Fig. 1 shows schematic views of the two diodes.

The polonium source for diode A was prepared in these

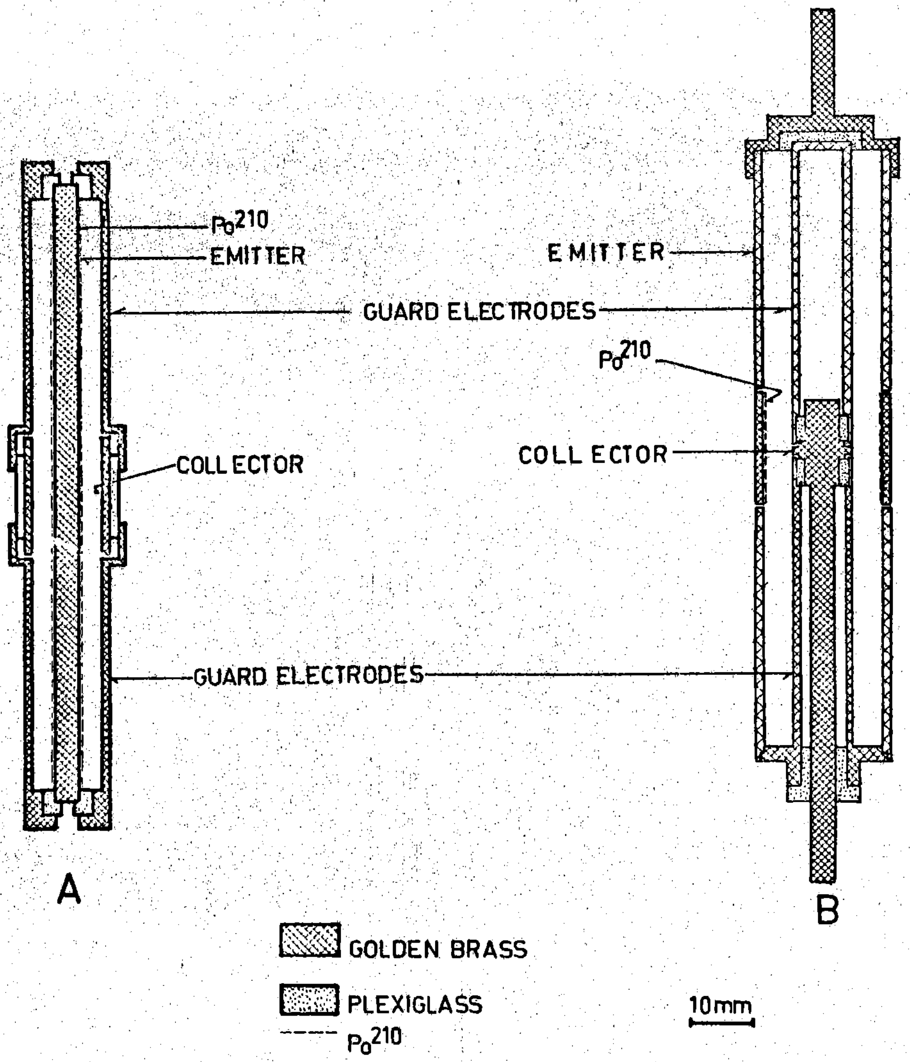

FIG. 1 - Schematic views of the two diodes used for the mobility measurements. Diode A has been used for the measurements reported in BS. The emitting electrode is the inner cylinder. For diode B, the emitting electrode is the outer cylinder.

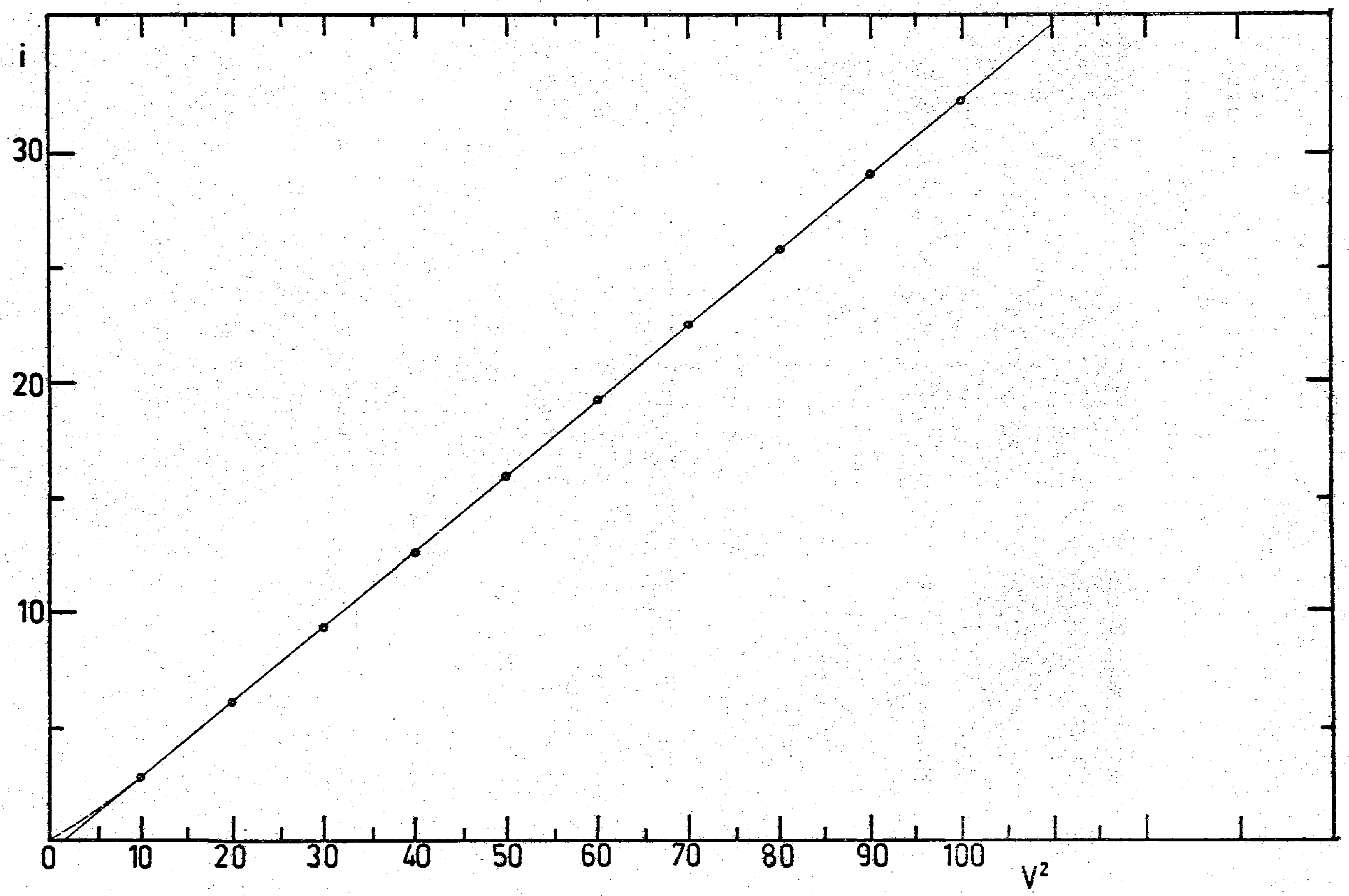

FIG. 2 - Typical plot of the electric current i versus the square of the applied voltage V. Currents are in units of 10^{-13} amp, applied potentials in volt (FERO 49, T = 2.167°K).

Laboratories(*), by autodeposition from a solution of 50 mC of polonium nitrate in nitric acid, and by then coating the alpha-source with a thin layer of plexiglass first and gold afterwards. For diode B the alpha-emitting ring was prepared by the "Radiochemical Centre"+: It had 50 mC of polonium deposited on it and was then covered with a thin gold layer.

A Keithley mod. 241 Regulated High Voltage Supply unit, which can give up to 1000 volt in steps of 0.01 volt, was used (accuracy 0.05%).

The current was measured by an EKCO mod. 616A Vibrating Reed Electrometer (accuracy 5%), whose output was displayed on a Speedomax Recorder.

Measurements were taken in seven runs. The temperature region investigated is from $\sim 1°K$ to the lambda-point.

A set of ten values of the applied voltage was systematically used, corresponding to V^2 varying from 10 to 100 $volt^2$ in steps of 10 $volt^2$.

ANALYSIS OF DATA.

The plots of the current versus the square of the applied voltage were to a very good approximation straight lines over the entire range of V used, except at the lowest temperatures, where the highest two or three current values were low with respect to the straight line, thus showing that the condition of complete space charge limitation begins to be invalid.

Fig. 2 shows an example of an i vs. V^2 plot, in a case where the condtion of complete space charge limitation is fulfilled(x).

(*) - With the help of the "Sezione Dosimetrica".

(+) - UKAEA, The Radiochemical Centre, Amersham, Buckinghamshire, England.

(x) - It will be noted that the straight line passing through all the experimental points does not pass exactly through the origin. This means that in the low voltage region ($V \lesssim 1$ volt) the current is not a linear function of the square of the applied potential (this is qualitatively indicated by the dotted line in Fig. 2). This could be ascribed to the mechanism of ion extraction from the strongly ionized layer created by the polonium alpha-particles. Duprè[8] found a similar effect with a slightly different geometry, and found it to be affected by a heat flow. Following Duprè's suggestion, we tried heating the helium at the bottom of our diode B,

./.

Table I gives, as a function of temperature, the slopes s of the i vs. V^2 plots and the mobilities, for every run. The values of the coefficient a for the two diodes have been evaluated according to paragraph 2 of page 4, by using the values of s (one for each diode) indicated by a star in the table. This gives:

$$\text{for diode A,} \qquad a = 128.5$$

$$\text{for diode B,} \qquad a = 6.061$$

(i is in 10^{-13} amp, μ in cm^2/volt.sec., V in volt).

Fig. 3 is a plot of the logarithm of the mobility versus the inverse of the absolute temperature. Also the experimental points obtained with diode A (already published in BS) are shown. The straight lines are best fits to the points in the low temperature region.

To demonstrate the validity of our method we compare the results shown in Fig. 3, i.e., the temperature dependence of the mobility, with the corresponding results obtained by other techniques.

Meyer and Reif in their most recent paper on this subject(6), report a similar plot of log μ versus $1/T$ for a larger temperature interval ($0.6 \lesssim T \lesssim T_\lambda$). For temperatures above 0.8°K the experimental points lie on straight lines (one for positive and one for negative currents), whose slopes(+) are

$$S_+ = 8.8\,°K\,, \quad \text{for positive ions,}$$

$$S_- = 8.1\,°K\,, \quad \text{for negative ions.}$$

In the range from approximately 1.5°K up to 2.0°K, however, the experimental points tend to lie slightly above the straight lines, as though they fitted a straight line with a smaller slope. In fact, Cunsolo(7) shows his results for positive ions by fitting them with two straight lines, the change in slope occurring at ∿ 1.3°K. The two slopes, corresponding to the

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(x) - thus producing a heat flow in the direction of the cylindrical axis. This did in fact change the shape of the i vs. V^2 plot, eliminating in almost all cases the curved part of it at the lowest voltages, and always leaving unaffected to within ∿ 1% its slope. The value of the slope s used for the mobility calculations has been then evaluated in the straight line region.

(+) - All the slopes S regarding the log μ vs. $1/T$ plots are evaluated by using the natural logarithm of the mobility.

TABLE I

Values of the slopes $s = i/V^2$ of the i vs. V^2 plots as a function of the temperature T. The mobilities μ are then evaluated following the method described in paragraph 2 on page 3. In the column "Run" the conventional name of the run is indicapted. i is in units of 10^{-13} amp, V is in volt, μ is in cm^2/volt.sec.

Positive Currents

Run	T	$s = i/V^2$	μ
Diode B			
FERO 49	1.248	6.225	1.027
" 52	1.256	5.888	0.972
" 46	1.323	4.625	0.763
" 47	1.323	4.725	0.780
" 49	1.337	4.150	0.685
" 49	1.431	2.775	0.458
" 51	1.431	2.675	0.441
" 49	1.536	1.875*	0.310
" 49	1.672	1.246	0.206
" 49	1.745	1.029	0.170
" 47	1.799	0.918	0.151
" 49	1.828	0.845	0.139
" 49	1.905	0.715	0.118
" 48	1.960	0.636	0.105
" 47	1.996	0.586	0.0967
" 51	2.004	0.550	0.0908
" 52	2.004	0.540	0.0891
" 49	2.008	0.553	0.0912
" 49	2.049	0.495	0.0817
" 49	2.066	0.474	0.0782
" 48	2.079	0.464	0.0759
" 49	2.083	0.446	0.0736
" 49	2.105	0.420	0.0693
" 49	2.128	0.385	0.0635
" 48	2.141	0.371	0.0612
" 49	2.155	0.349	0.0576
" 49	2.167	0.329	0.0543
" 49	2.171	0.321	0.0530
Diode A			
PCS 14	1.131	251.0	1.953
"	1.273	116.0	0.902
"	1.446	57.70	0.449
"	1.658	27.75*	0.216
"	1.894	15.60	0.121
"	2.090	10.00	0.0778
"	2.156	8.025	0.0624

Negative Currents

Run	T	$s = i/V^2$	μ
Diode B			
FERO 50	1.030	12.05	1.988
" 50	1.256	3.45	0.569
" 46	1.323	2.650	0.437
" 47	1.323	2.613	0.431
" 50	1.342	2.375	0.392
" 50	1.435	1.659	0.274
" 50	1.541	1.148	0.189
" 50	1.678	0.796	0.131
" 46	1.799	0.615	0.101
" 50	1.832	0.565	0.0932
" 50	2.004	0.381	0.0629
" 50	2.062	0.326	0.0538
" 50	2.101	0.290	0.0479
" 48	2.141	0.244	0.0403
" 50	2.146	0.234	0.0386
" 50	2.165	0.215	0.0355
" 50	2.169	0.205	0.0338

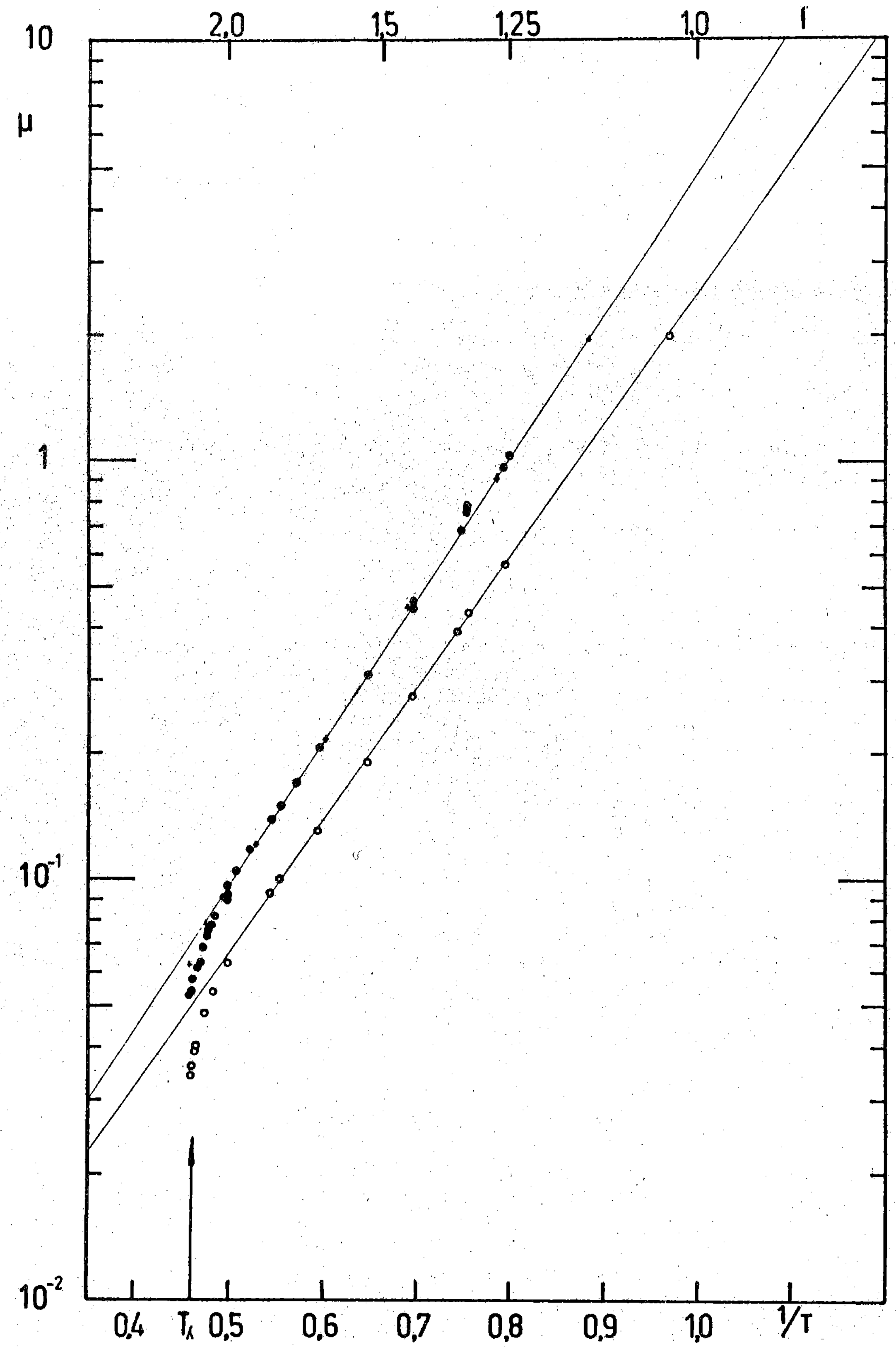

FIG. 3 - Plot of the logarithm of the mobility μ versus the inverse of the absolute temperature T. The upper curve is for positive mobilities, the lower one for negative mobilities. The experimental points indicated by a cross are those already published in BS, Fig. 7. The straight lines are best fits to the points at temperatures below 2°K. Mobilities are in cm^2/volt sec, temperatures in degrees Kelvin.

two temperature regions, are (*):

$$\left.\begin{array}{l} S_{+}\,(T \lesssim 1.3°K) = 9.1°K \\ \\ S_{+}\,(1.3°K \lesssim T \lesssim T_{\lambda}) = 7.5°K \end{array}\right\} \text{for positive ions}$$

The value (9.1°K) found by Cunsolo in the low temperature region is in good agreement with the value (8.8°K) found by Meyer and Reif for positive ions.

Coming back now to our measurements, the slopes of the two straight lines of Fig. 3 are:

$S_{+} = 7.8°K$, for positive ions,

$S_{-} = 7.1°K$, for negative ions.

Since these measurements have been taken in the temperature range from $\sim 1.2°K$ to the lambda-point, the value (7.8°K) found for positive ions has to be compared with the value (7.5°K) found by Cunsolo for the higher temperature region, and is in a rather good agreement with it.

It can be helpful at this point to briefly summarize what is known at present about the ionic mobilities in superfluid helium in the temperature and electric field regions in which only elastic collisions between ions and excitations are postulated to occur. The only approach to the problem made so far is a gas-kinetic one, considering both excitations (in the temperature region in which we are interested these are only rotons) and ions as hard-spheres and describing the mobility in terms of the ions mean free path[4,6]. This approach gives a mobility temperature dependence of the form

(II) $$\mu \propto T^{-1} \exp(\Delta/KT).$$

Δ/k is the roton excitation energy and equals 8.68°K. In the proportionality constant are included the effective mass of the roton and that of the ion and the ion-roton cross-section. The experimental results, as seen before, show, in a first approximation and in a limited temperature region ($0.8°K \lesssim T \lesssim 2.0°K$), a linear dependence of the logarithm of the mobility on the inverse of the absolute temperature. This means that the T^{-1} factor of equation (II) does not appear to exist. Meyer and Reif[6] interpret this by saying that the hard-sphe-

(*) - The same plot, with the same values of the slopes, is shown in BS, Fig. 1.

re model used to deduce the above formula (II) is a poor approximation and that the introduction of a temperature-dependent ion-roton cross-section would be more realistic and would cancel the T^{-1} factor. But, also if this correction is accepted, one should also expect the slope of the log μ vs. 1/T plot to be the roton excitation energy Δ/K, both for positive and negative mobilities. In contradiction to this, these slopes are different for negative and positive ions and are different in different temperature regions. So it appears that the hard-sphere model is in any case a very rough approximation to the problem .

Another peculiar aspect of the temperature dependence of the mobility is its behaviour in a very narrow temperature region close to the lambda-point ($2.0°K \lesssim T \lesssim T_\lambda$). In this region, as can be seen in Fig. 3, the dependence of log μ on 1/T deviates from the straight line, exhibiting much lower mobility values. This was already shown in BS for positive currents. In the same paper an attempt was made to interpret this in terms of the temperature dependence of the roton excitation energy. In fact, such a dependence has been observed by Yarnell et al.[9] and by Henshaw and Woods[10,11] by inelastic neutron spectroscopy. The former made measurements in the temperature region from 1.1°K to 1.8°K and found a change of Δ/K from 8.68°K to 8.15°K . The latter worked over a larger temperatures interval, up to the lambda-point and into the He^4 I. At T_λ , Δ/k is 5.4°K, thus showing a very strong decrease in the temperature range from 2.0°K to 2.18°K.

Describing the anomalous temperature dependence of the mobility in the range $2.0°K \lesssim T \lesssim 2.18°K$ in terms of the temperature dependence of the roton excitation energy Δ/k, however, is neither easy nor convincing for at least two reasons: Firstly, because altered values of Δ/k appear as the slopes of the straight lines of Fig. 3 (7.8°K and 7.1°K instead of 8.68°K). Secondly, because using the hard--sphere model and treating the rotons as quasi-particles in a temperature region where the roton density is extremely high, is physically unsound. All that can be reasonably said is that the decrease of the roton excitation energy would have the effect of decreasing the ionic mobilities. We think, however, that this peculiar aspect also must be faced in a more general perspective, and that we are still at only a first approximation in the theoretical interpretation of the ionic mobilities in superfluid helium.

DISCUSSION OF THE METHOD.

Using the method as an absolute one would require two main features:

1. An accurate calibration of the instruments measuring the current and the applied voltage. For the latter,

the problem is easy to solve, since the values of the potential used are of the order of a few volts. For the current, the problem of accurately knowing its absolute value is not so simple, since the currents vary from 10^{-13} to 10^{-10} ampere (for example, the accuracy of our EKCO Vibrating Reed Electrometer is 5%).

2. An accurate knowledge of the distance between the electrodes (of the radii of the electrodes, for the case of cylindrical geometry). This does not mean simply the geometrical distance (which is not easy to measure, since it is of the order of a few millimeters and thermal contraction must be taken into account). The presence of the intensely ionized layer, which presumably acts as a conductor, shifts the $E = 0$ surface towards the collecting electrode so that the geometrical distance between electrodes is effectively decreased by~0.2 mm.

As a matter of fact, we compared the calibration values of the coefficients a in equation (I), found in the way described above (see paragraph 2, page 3), with those calculated with the nominal dimensions of the diodes (see paragraph 1, page 3) and assuming the emitting surface to be shifted 0.2 mm towards the collecting one. This gives:

	experimental	theoretical	deviation (%)
diode A	128.5	127.6	+ 0.7
diode B	6.061	6.135	- 1.2

The calculated deviation is then of the order of 1% with respect to measurements made by other techniques. It must be remembered, however, that these are given with an accurancy not better than 5%, as far as the absolute values of the mobility are concerned.

Regarding the accidental errors, the good alignment of the experimental points on a straight line in the i vs. V^2 plots (see Fig. 2) and the small differences found between measurements performed in different runs (see Table I) enable us to estimate these errors to be about 2%. Taking into account also the small uncertainty in the slope (see footnote at page 6), which has been estimated to be not more than 1%, we can say that, at the present state of this technique, the accidental errors in the mobility measurements are not more than 3%.

To conclude, we want to point out two lines for the development of this technique in the future. One is the possibility of making it a better absolute method, and work is in progress in this direction. The second is the possibility of applying it to the measurement of the mobility of liquids, such as He^3, which has mobilities much less than those of He^4. The method is much more rapid than a time-of-flight method and could then also be used in difficult experimental conditions, such as, for example, an adiabatic demagnetization cryostat.

BIBLIOGRAPHY.

(1) - A.L. Bartoli and F. Scaramuzzi, CNEN, Laboratori Nazionali di Frascati, report LNF-62/106 (1962).

(2) - H.F. Ivey, Adv. in Electr. and Electr. Phys., 6, 137 (1954).

(3) - L. Meyer and F. Reif, Phys. Rev., 110, 279, (1958).

(4) - G. Careri, F. Scaramuzzi and J.O. Thomson, Nuovo Cimento 13, 186, (1959).

(5) - A. Dahm, J. Levine, J. Penley and T.M. Sanders, Proc. 7th Int. Conf. Low Temp. Phys., Toronto (1960), p. 495.

(6) - L. Meyer and F. Reif, Phys. Rev., 119, 1164, (1960).

(7) - S. Cunsolo, Nuovo Cimento, 21, 76, (1961).

(8) - F. Duprè, private communication.

(9) - J.L. Yarnell, G.P. Arnold, P.J. Bendt and E.C. Kerr, Phys. Rev., 113, 1379, (1959).

(10) - D.G. Henshaw and A.D.B. Woods, Proc. 7th Int. Conf. Low Temp. Phys., Toronto (1960), p. 539.

(11) - D.G. Henshaw and A.D.B. Woods, Phys. Rev., 121, 1266, (1961).