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Nota interna: n° 173
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A.L. Bartoli,[(+)] F. Scaramuzzi: Completely Space Charge Limited Currents in Superfluid Helium: a Method for the Measurement of the Ionic Mobilities in the Vicinity of the Lambda--Point.

Introduction.

The mobility of helium ions in liquid He^4 below the lambda-point (T = 2.18°K) has been measured by different authors with different techniques[(1,2,3,4,5,6)] at temperatures down to a few tenths of a degree. Both positive and negative mobilities change very strongly in this short temperature interval (from ~ 0.03 cm^2/ volt.sec at the lambda-point up to several hundreds at temperatures lower than one degree) and are in not bad agreement with the Landau-Khalatnikov picture of superfluid helium[(7)]. The methods which give the most reliable results until now are based on

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time-of-flight measurements. Here a new method, based on the measurement of the (D.C.) voltage-current characteristics of a diode under conditions of complete space charge limitation is reported and the mobilities of the positive ions, as a function of temperature, are shown and compared with the known values. (Measurements on negative mobilities are in progress.)

The peculiar behaviour of the mobility in the vicinity of the lambda-point is emphasized and an attempt is made to correlate it with the temperature dependence of the activation energy for rotons measured by Yarnell, Arnold, Bendt and Kerr(8) and by Henshaw and Woods.(9)

The precision and the limits of such a method are then discussed and the conclusion is reached that this method is likely to be used for mobilities not too high (roughly, less than 2 cm^2 / volt. sec); this suggests that it could be employed with success on other liquids, like He^3 for example, which have mobilities much lower than superfluid helium.

The Method.(+)

We define "complete space charge limitation" in a diode, following Ivey,(10) to be the situation where the current is so high that the value of the electric field

$$E = - \text{grad } V$$

(+)- Electrostatic c.g.s. system is used all through this report.

at the emitter[x] is zero.

In this case Poisson's equation

$$\nabla^2 V = -\frac{4\pi\rho}{K}$$

(V = electric potential; ρ = space charge density; K = dielectric constant of the medium) can be solved for a dense gas, if we introduce the definitions

$$J = \rho v \quad ; \quad v = \mu E$$

(j = current density; v = drift velocity; μ = mobility; E = electric field) and provided that the following conditions are fulfilled:

a) mobility independent of the electric field (this condition is satisfied if the energy gained by an ion from the field between two successive collisions with the gas molecules is small compared with its energy);
b) charges of only one sign moving in the field;
c) absence of initial velocities of the ions.

Ivey in his review article[10] gives the solution of this problem for two simple geometries[+]:
-plane electrodes (i.e., two indefinite parallel planes):

(x) - In all this report we will call the "emitter" the electrode where the charges are created and the "collector" the electrode where the current is collected, in place of the more common "cathode" and "anode".
(+) - The dielectric constant of the medium, K, is taken =1.

$$J = \frac{9}{32} \frac{\mu V^2}{\pi x^3}$$

(x= distance between the electrodes);

- cylindrical electrodes(i.e.,two concentric indefinite cylinders)

(1) $$i = \frac{\mu}{2} \frac{V^2}{r^2 R(r/r_o)}$$

(i= electric current per unit axial length of the electrodes; r_o= emitter radius; r= collector radius). The function $R(r/r_o)$ is different according to which of the electrodes (internal or external) is the emitter:

- internal emitter ($r_o < r$)

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

external emitter ($r_o > r$)

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

We point out that both solutions (for plane and cylindrical geometries) can be written as follows:

(3) $$I = \alpha \mu V^2$$

where I is the total current collected at the collector and α is a geometrical factor which does not depend on the applied electric field, nor on the physical conditions of the medium.

This simple relation suggests a method for measuring the mobilities of ions in a dense medium, provied that the above conditions are satisfied. The procedure is very simple. A plot is made of the current versus the square of the voltage; the linearity of this plot is a proof that the currents are completely space charge limited; the slope of the straight line, once the geometrical factor α is known, gives the mobility.

Ionic Mobilities in Superfluid Helium.

The measurement of the mobility of thermal helium ions in liquid He^4, below the lambda-point, has been performed by many authors with different techniques[1,2,3,4,5,6]; these measurements are interesting because the helium ions can be considered microscopic probes, and from their interactions with the excitations[11] of liquid helium, information can be drawn on its superfluidity.

We shall not be concerned here with the inelastic collisions between ions and excitations because these can be revealed more favourably with high electric fields and at temperatures lower than 1°K. The method reported here, as will be shown, is not practically applicable at temperatures lower than 1.1°K (improvements in the technique probably will allow reaching slightly lower temperatures, but 1.0°K will most likely not be feasible).

Fig. 1 is a plot of the logarithm of the mobility of positive ions, measured by other techniques, as a function of the inverse of the absolute temperature. The solid line is a best fit to the experimental results, greater weight having been given to the measurements of Careri, Cunsolo, Duprè[5,6] and Meyer, Reif[2,4], based on time--of flight methods, which were more reliable technically. The best fit curve is composed of two straight lines, the change in slope occurring at $\sim 1.25°K$.

A theoretical calculation of the mobility can be performed following the theory of Zharkov and Khalatnikov[7] for the case of He^3 atoms dissolved in liquid He^4; the result is reported by Careri, Scaramuzzi, Thomson[3] and by Meyer, Reif[4] and gives the following temperature dependence of the mobility

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

(k=Boltzmann's constant) Δ/k is the roton activation energy (expressed in degrees Kelvin), postulated by Landau[7] and measured by Yarnell, Arnold, Bendt, Kerr[8] and by Henshaw, Woods[9], and is found to be slightly temperature dependent; Yarnell and coworkers give the following empirical relation:

(5) $$\Delta/K = 8.68 - 0.0084\, T^7 \ °K.$$

The experimental results exibit the following behaviour:

(6) $$\mu \propto \exp(\Delta/KT).$$

The two slopes shown in fig. 1 correspond to two values of Δ/K:

$$1/T > 0.8\ °K^{-1}\ ,\quad \Delta/K = 9.2\ °K$$

$$1/T < 0.8\ °K^{-1}\ ,\quad \Delta/K = 7.5\ °K$$

The combined values of Δ/K agree in a first approximation with the value measured by the neutron scattering experiments.[8,9] No trace of the T^{-1} factor of equation (4) seems to exist in the experimental results; Meyer and Reif[4] impute this discrepancy to the fact that, in deriving equation (4), a hard sphere interaction between ion and roton has been used, which is a poor approximation: if a velocity-dependent cross-section is introduced, one obtains a correction in the proper direction. In the same paper the log μ versus 1/T plot is represented, for positive ions, by one straight line only: the change in slope of fig. 1 is accounted for saying that in the range from 1.5°K up to 2.0°K the experimental points tend to lie slightly above the straight line. Surprisingly, this is attributed to the temperature dependence of the activation energy, given in equation (5), which has, on the contrary, the effect of lowering the mobility values.

Finally it has to be noted that, in all these measurements while the temperature dependence of the mobility can be measured whith good precision, the absolute values can be incorrect by a common scale factor, depending on the fact that, to compute the mobilities, the distance between two electrodes must be introduced, whatever the technique.

The Experiment.

The apparatus consist of a diode; one of the electrodes is plated with Po^{210}, which emits alpha-particles of 5.3 MeV, with a range in superfluid helium of 0.22 mm; the same electrode (emitter) is connected to a regulated voltage supply, while the collector is connected to a vibrating reed electrometer, which can measure currents down to 10^{-15} ampere. The output of the electrometer is sent to a recorder. Fig.2 shows a block diagram of the system.

The apparatus is immersed in the helium bath with a standard low thermal conductivity support and the temperature is kept constant, with a heater-thermoregulator, within 0.001°K.

Two kinds of geometries have been used: a plane one, shown in fig.3, and a cylindrical one, shown in fig.4; both drawings are with cylindrical symmetry.

The condition of complete space charge limitation is checked, as already said above, by plotting the current versus the square of the applied voltage and checking that it lies on a straight line: fig.5 shows a typical plot of i versus V^2 . From such plots the slope i/V^2 is computed; it corresponds to the mobility except for the scale factor α of equation (3), which does not depend on temperature. α has been found, in a first approximation, from the mobility values of other authors for one temperature: with this value of α the mobilities μ for positive ions have been calculated from the slopes i/V^2 . Fig.6 shows the results for the plane geometry, and fig.7 those for the cylindrical geometry; the solid line in both is the best fit line for other authors' measurements, already shown in fig.1.

Discussion.

We first want to make a few observations about the extent to which the conditions, described in the section on "The Method", have been fulfilled:

a) the mobility can be considered independent of the electric field in the temperature region in which we have measured it; in fact, the thermal energy of an ion at a temperature around 1°K can be taken to be of order of 10^{-4} ev; this must be much greater than the energy gained by an ion from the electric field between two successive collisions, which is

$$L = e E l$$

(e = electron charge; l = mean free path). We used a maximum field of 20 volts/cm, a minimum temperature of 1.13°K, to which corresponds a mean free path for rotons (and then for ions) $l = 10^{-5}$ cm; as a result L_{max} is of the order of 10^{-8} ev.

b) The ions moving in the diode are of one sign except in the region, 0.22 mm thick, in which ionization by alpha-particles takes place. In this layer, due to the very high ionization, the situation can be summarized by saying that it has a very large electrical conductivity with respect to the region in which ions of only one sign move. This effect can be taken into account by supposing that the emitter surface is not confined to the electrode boundary, but is displaced towards the end of the layer: this means that the distance between the two electrodes is in fact decreased by the thickness of the layer.

Following this suggestion, the empirical value of , deduced by comparing the experimental results with the mobilities measured by other authors, has been introduced in equations (1) and (2) and the value of r_0 calculated: it was found to be greater than the emitter radius by a quantity $\delta r_0 = 0.19$ mm, close to the range of the alpha-particles.

c) The mean free path of the ions, in the temperature region in which the measurements have been taken, is so small that the effects due to initial velocities of the ions are confined to the highly ionized layer quoted above and are then negligible.

Besides these three conditions, the geometrical condition of indefinite electrodes must be considered. Looking at fig.3, it can be seen that for the plane-electrodes apparatus the ion source is smaller than the collector, which means that the boundary effects on the beam of ions are felt by the apparatus, and a priori these are not temperature-independent. For the cylindrical-geometry apparatus this condition has been changed (see fig.4). The presence of guard electrodes guarantees that the current which reaches the collector is negligibly affected by the boundaries of the beam. Therefore, in this case, the electrodes can be considered indefinite. This difference between the two kinds of diodes could explain the different results: the plane-electrodes data (see fig.6) show a slope slightly different from the best fit curve, while the cylindrical-electrodes results (see fig.7) are in very good agreement with it. Then in the following considerations we refer only to the latter geometry.

Comparing fig.1 and fig.7 (in which the experimental errors are not larger than the linear dimensions of the representative points) it can be seen that the temperature dependence of the mobility can be measured with good precision by the method here reported.

A limitation of the method consists in the following: looking at fig.5, if we increase the applied voltage, at a certain value V_0 the curve begins to deviate from the straight line[(+)], bending towards the V^2 axis. This means that the condition of complete limitation of the current by space charge is no longer true; decreasing the temperature, the mobility increases as do the currents, V_0 becoming smaller and smaller. When V_0 is a few volts, the method is no longer reliable. The minimum temperature, of course, depends on the intensity of the Po^{210} source: with the most intense source available, we were able to measure the mobility of positive ions down to a temperature of 1.13°K.

The measurement of the mobility with the technique reported here does not give a new result, except in the region between 2.0°K and the lambda-point. Fig.7 (and also fig.6) shows in this region a bending of the curve towards the 1/T axis. In fig.1 (results of other authors) the effect is not shown, because measurements were not taken at temperatures higher than 2.0°K; such an effect was shown by Meyer and Reif in a first short communication[(2)], but in the final, more complete paper[(4)], they do not quote it. An at-

(+) - A practical criterion must be established to define V_0 .

tempt has been made to interpret this effect in terms of the temperature dependence of the rotons activation energy: introducing equation (5) in equation (6) a correction in the proper direction is obtained, but not in quantitative agreement with the data of fig.7. Using the same T^7 dependence, the coefficients are different:

$$\Delta/K = 7.63 - 0.0018\ T^7\ {}^{\circ}K\ .$$

It must be emphasized that:
- Yarnell and coworkers[8] obtained equation (5) in the range 1.1°K - 1.8°K ; in the same range the mobility measurements do not show any similar phenomenon;
- Henshaw and Woods[9] do not derive any relation, but they observed a strong decrease of Δ/K going towards the labda-point; they took measurements also in the range 2.0°K - -2.18°K and in this range Δ/K varies from 7.5°K down to 5.4°K;
- it is hard to speak of excitations as quasi-particles in the temperature region close to the lambda-point, due to the very strong interactions between them.

Concluding, the theoretical approach described in the chapter "Ionic Mobilities in Superfluid Helium" must be considered only as a first approximation .

Conclusions.

The method here described, although not at present an absolute method, is likely to be used with success for the measurement of ionic mobilities, once the geometrical constant of the apparatus has been established (using a liquid of known mobility). There is also some hope of making out of it an absolute method, as a consequence of the considerations made above (see Discussion, b); work is in progress along these lines and for the determination of the mobility of negative ions in liquid helium. Particular attention is being payed to the temperature region close to the lambda-point, where a larger number of measurements(both for positive and negative ions) are in progress.

The limits of the method ($T \gtrsim 1.1$°K in liquid helium, or more generally $\mu \lesssim 2$ cm^2/ volt.sec) suggest using it for other liquids, such as He^3 for example, which has mobilities of the order of 10^{-2} cm^2/ volt.sec.

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Captions to Figures.

Fig. 1 - Plot of the logarithm of the mobility μ of positive ions in superfluid helium versus the inverse of the absolute temperature T.
- time-of-flight method, Meyer and Reif[4],
- time-of-flight method, Careri, Cunsolo, Duprè[5,6],
- o heat flush method, Careri, Scaramuzzi, Thomson[3],
- \+ space charge method, Careri, Cunsolo, Duprè[5].

The solid line is a best fit between the experimental data. Greater weight has been given to the measurements based on time-of-flight methods. Mobilities are in cm^2/ volt.sec., temperatures in degrees Kelvin.

Fig. 2 - Block diagram of the electric currents measuring system.

Fig. 3 - Schematic view of the plane-geometry apparatus; the drawing has a cylindrical symmetry along the vertical axis.
E = emitter,
C = collector,
G = guard electrode.

Fig. 4 - Schematic view of the cylindrical-geometry apparatus; the drawing has a cylindrical symmetry along the vertical axis.
E = emitter,
C = collector,
G = guard electrodes.

Fig. 5 - Typical plot of the electric current i versus the square of the applied voltage V. Currents are in ampere.10^{-13}, voltage in volts.

Fig. 6 - Plot of the logarithm of the mobility μ of positive ions versus the inverse of the absolute temperature T. The experimental results have been taken with the plane-geometry apparatus (see fig.3). The solid line is the same of fig. 1. Mobilities are in cm^2/volt.sec., temperatures in degrees Kelvin.

Fig. 7 - Plot of the logarithm of the mobility μ of positive ions versus the inverse of the absolute temperature T. The experimental data have been taken with the cylindrical-geometry apparatus (see fig. 4). The solid line is the same of fig.1. Mobilities are in cm^2/volt.sec., temperatures in degrees Kelvin.

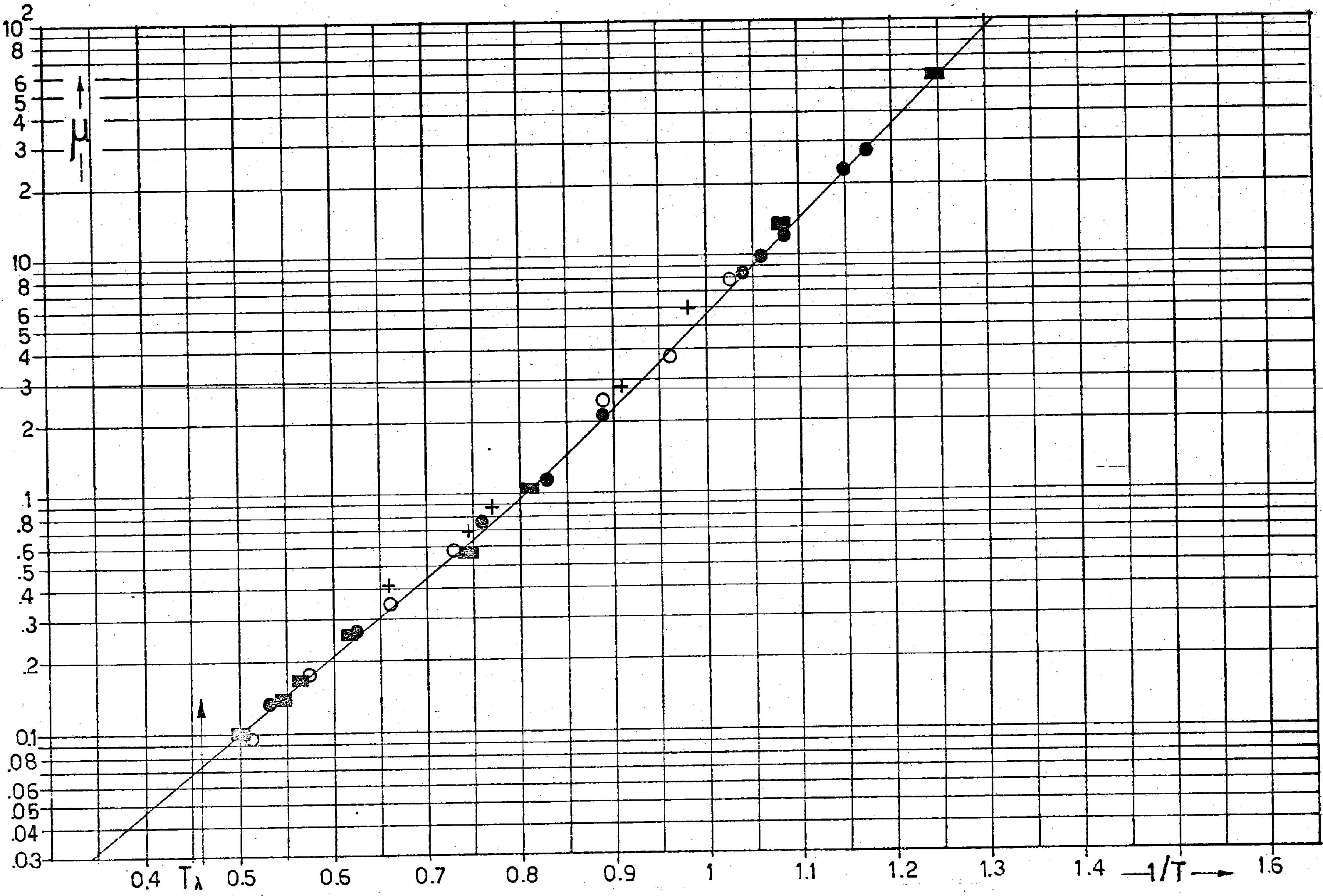
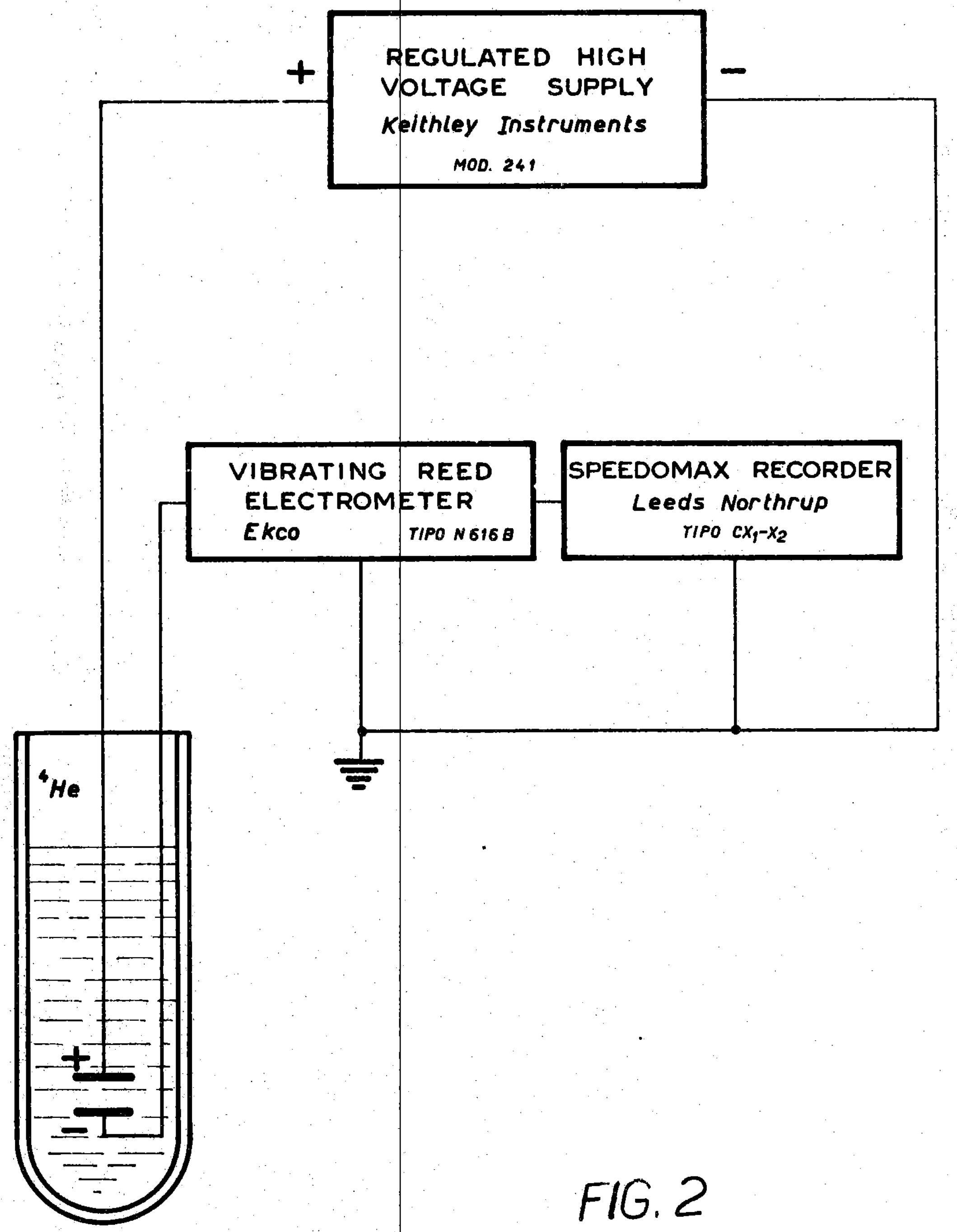


FIG. 1



+
REGULATED HIGH VOLTAGE SUPPLY
Keithley Instruments
MOD. 241
−
VIBRATING REED ELECTROMETER
Ekco
TIPO N 616 B
SPEEDOMAX RECORDER
Leeds Northrup
TIPO CX_1-X_2
^4He
+
−

FIG. 2

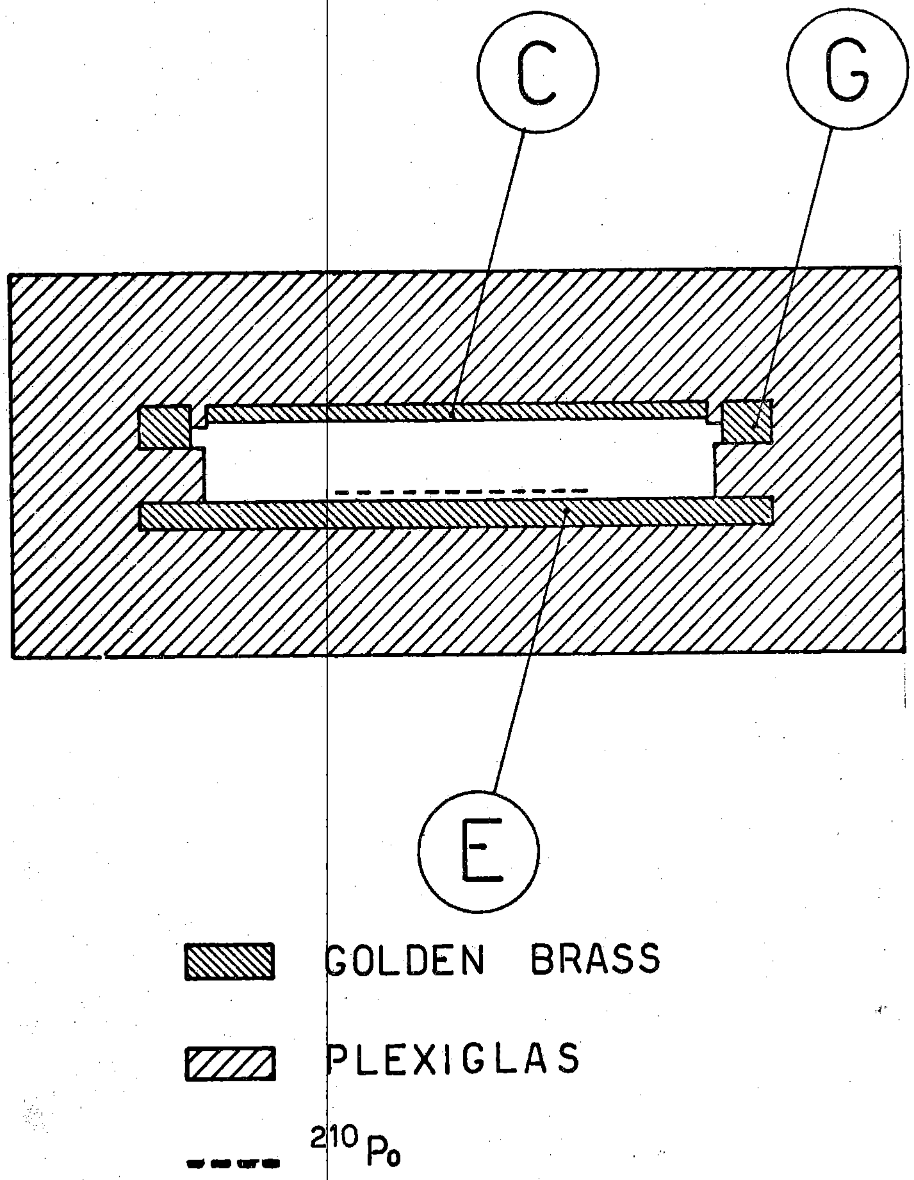
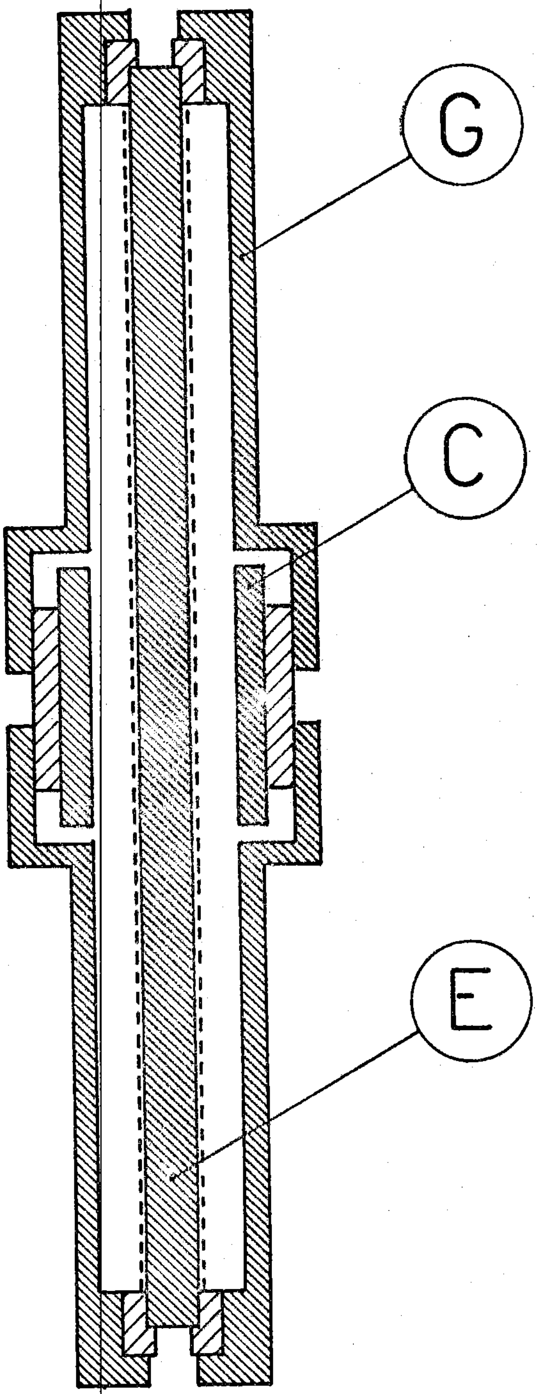


FIG. 3



GOLDEN BRASS

PLEXIGLAS

^{210}Po

FIG.4

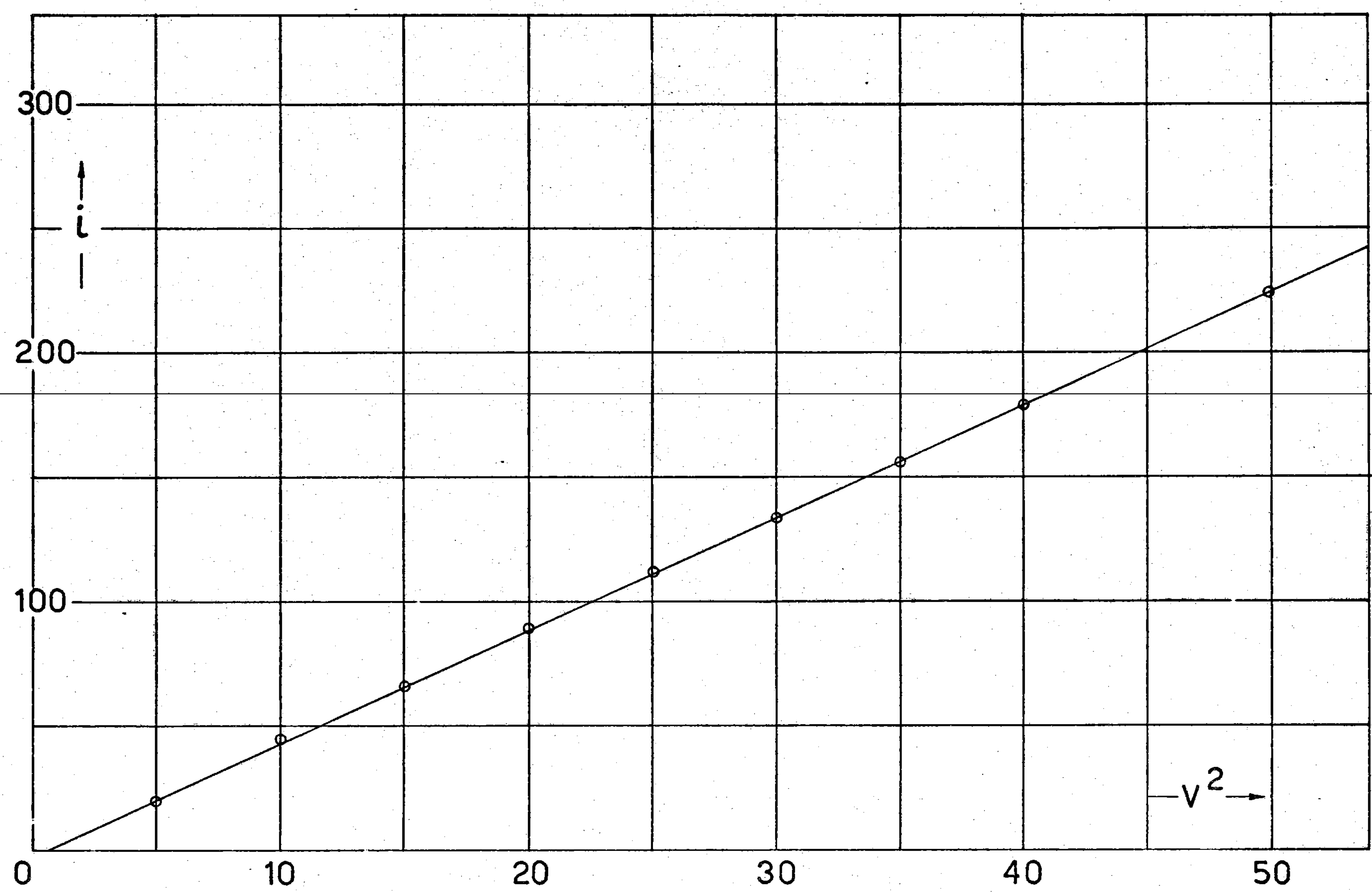
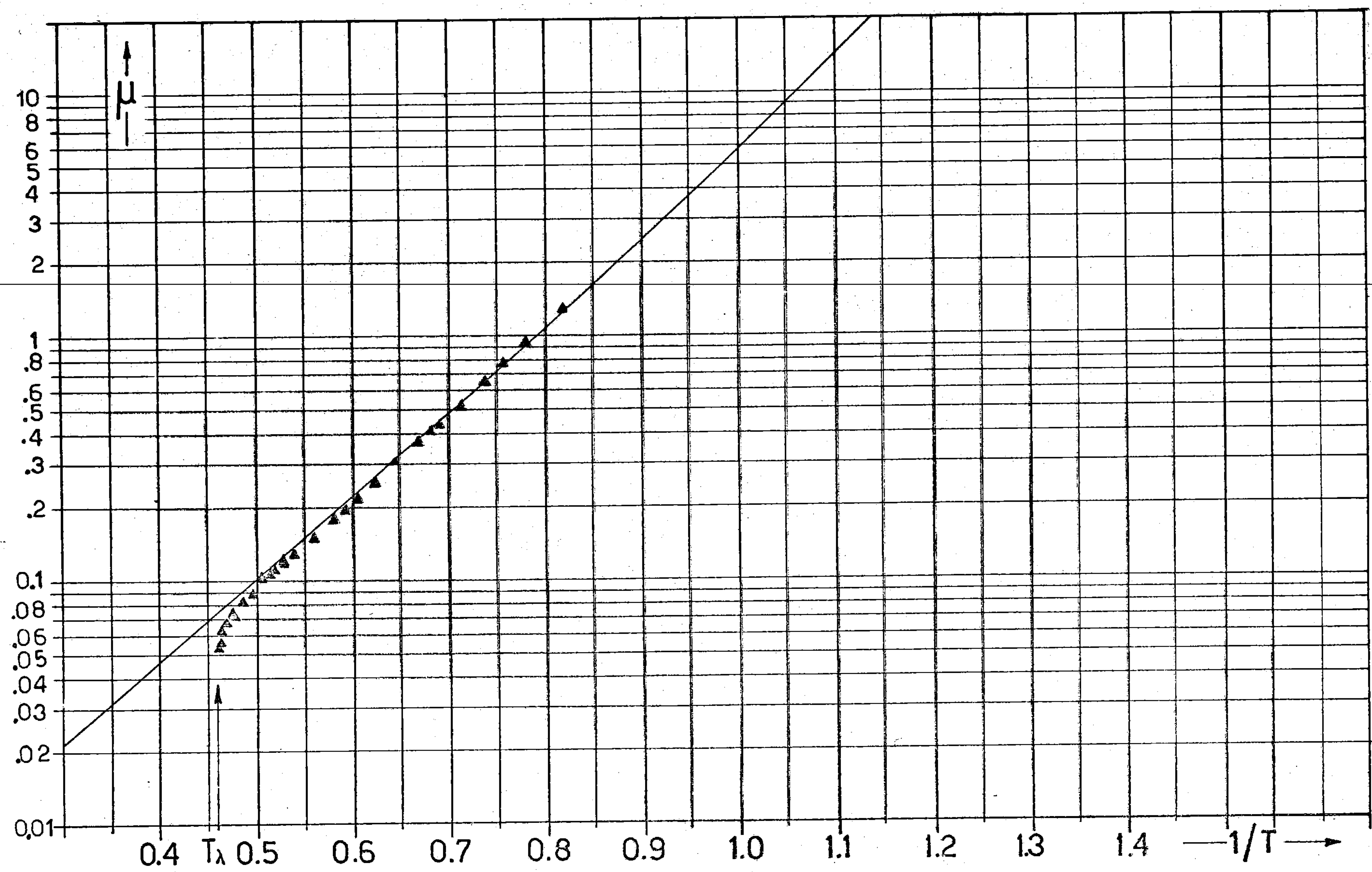


FIG. 5


μ
10
8
6
5
4
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2
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.8
.6
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.4
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0.1
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.05
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.03
.02
0.01
0.4
Tλ
0.5
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1.2
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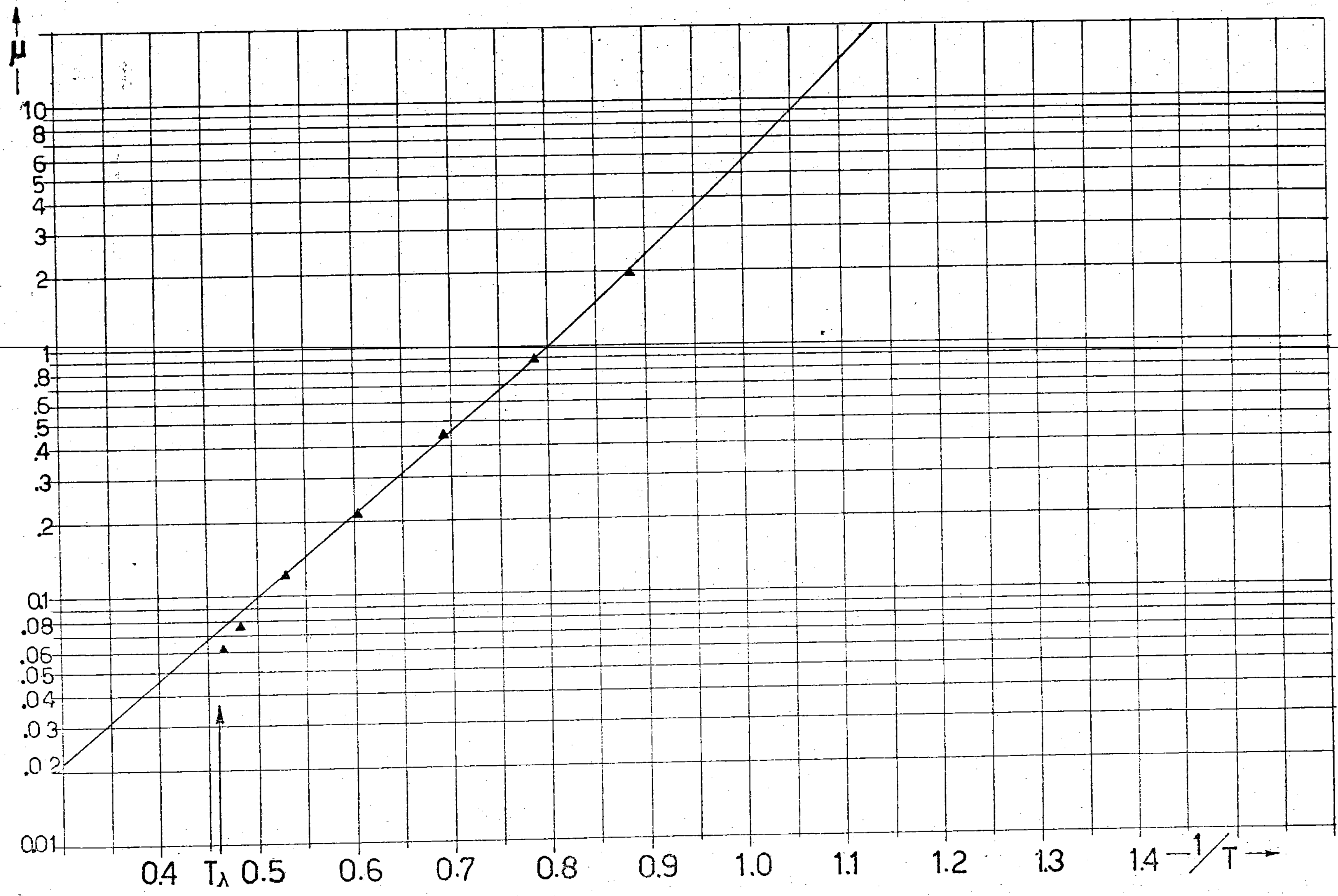
1.4
1/T

FIG. 6


μ
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0.4
Tλ
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0.7
0.8
0.9
1.0
1.1
1.2
1.3
1.4
1/T

FIG. 7