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INTRODUCTION

On the exposure of matter to high energy radiation ionization takes place in the form

$$M \rightsquigarrow M^{+} + e \qquad /1/$$

where M^{+} is the positive molecular ion and e^{-} is the electron. The ejected electrons have a wide spread of energy. The energetic ions are capable of inducing further ionization during their thermalization, thus giving birth to secondary, tertiary, etc. electrons in the irradiated system. In liquids usually more than 90 percent of these electrons are neutralized within about 10^{-11} sec by recombination with their parent ions or with ions in the sample spur [1], and only a minor fraction can escape from the attractive field of the ions. Electrons that do escape become thermalized at a distance from their parent ions at which the thermal energy is higher than the coulombic attraction energy. Until they finally encounter a positive ion, these electrons are free to diffuse at random in the system. The time taken by this diffusion is many orders longer than the time for geminate recombination, and in some favourable cases it may be of the order of seconds. The diffusion of the escaped /or quasi-free/ electrons can be oriented and enhanced by the presence of an external electric field. Thus, an applied field permits the observation of an increase in the electric current in the irradiated sample. Eventually all the quasi-free electrons are neutralized by positive ions crossing their path, so that under continuous irradiation a steady state sets in when the rate of formation of charge carriers equals their rate of recombination.

A steady-state current can be used to evaluate the free ion yield, G_{fi}, but many details in the mechanisms of carrier generation and recombination can be studied only by pulse methods.

In this paper we report the observation of steady-state and transient currents in 3-methylhexane /3-MHx/ induced by ^{60}Co gamma-irradiation photoemission and photoionization. The temperature dependence of charge carrier recombination rate, the induced current, charge carrier mobility and initial current have been measured.

EXPERIMENTAL

Preparation of samples. 3-MHx synthesized in this laboratory was purified first by shaking five times with cc sulphuric acid, neutralized by sodium hydroxide, washed with water, dried over calcium chloride, then distilled from metallic sodium into sealed ampoules. The purified samples were introduced into the equipment shown in Fig. 1. In part I of the apparatus the samples were degassed by thawing-freezing technique up to a pressure of 10^{-5} Torr of the frozen sample. In part II the sample was dehydrated by sodium mirror in 5 stages /a small amount of precondensate was collected and discarded before each step/. In part III the sample was electrolyzed for 4 days using tungsten wire electrodes at an applied voltage of 5000 V. After electrolysis the sample was again distilled before introduction into the measuring cell IV.

The- dc dark conductivity of 3-MHx purified by this method was less than $\sim 10^{-17}$ ohm^{-1} cm^{-1} at room temperature and about 1.5×10^{-18} ohm^{-1} cm^{-1} for the glass at 77^oK.

Irradiation. For irradiation a 0,5 Ci ^{60}Co gamma source and a dose rate of $4,75 \times 10^{11}$ eV cm^{-3} sec^{-1} were used. The ^{60}Co source could be pushed into a tube with its closed end protruding into the box housing the measuring cell and thus the source could be brought to 30 mm distance from the sample.

Conductivity measurement. The conductivity cell, prepared from Pyrex glass is shown in Fig. 2. The 1 mm thick Ag electrodes of 1 cm^2 surface area, were connected to the contacts by 1 mm dia tungsten wires fixed in glass tubes. The spacing of the electrodes was 1 mm.

Currents induced by gamma irradiation were measured with an ORION-KETI, TR-1501 electrometer with a time constant of less than 1 sec for the maximum input resistance of 10^{11} ohm used in the measurements. The insulation resistance of the equipment was about 10^{15} ohm. The temperature dependence of the current could be followed with adequate sensitivity for rates of temperature change, up to 2-3^{o}C/min.

Photo-currents induced by 10 μsec light pulses of an Ar filled discharge tube were measured in quartz cells /shown in [2]/ directly by oscilloscope or at lower current intensities by photographing the output of a Keithley model 640 electrometer displayed on an EMG type TR 4643 oscilloscope and photoed.

The cells and the heat exchanger blocks were mounted into a Faraday cage built from 5 mm thick iron plates; the lead from the heat exchanger was isolated from the box by a teflon plug, while the iron-constantan thermocouples and the power supply were connected through rubber and amphenol connectors, respectively. The 4 mm dia copper rod connecting the electrometer was fixed by means of quartz rings within an iron tube protruding into the Faraday cage. The applied voltage was taken from dry batteries or from stabilized power supplies.

Results and discussion

Steady state currents

The conductivity vs reciprocal temperature curves taken during irradiation are shown in Fig. 3. The current was measured during continuous cooling and spontaneous warming. An inflection is seen on both curves at the temperatures of structural transformations /131^{o}K and around 220^{o}K/, a maximum is seen only in the curve measured during continuous warming.

Charge carrier recombination is a second order process, and thus after irradiation has been stopped the decrease in concentration caused by recombination can be expressed as

$$- \frac{dn}{dt} = k'n^2 \qquad /2/$$

where n is the number of quasi free electrons /= number of positive ions/ per cm^3;

k' is the recombination rate constant, in $cm^3\ sec^{-1}$.

The solution to the differencial equation /1/ gives

$$k' = \frac{1}{t_{1/2}\ n_o} \qquad /3/$$

where $t_{1/2}$ is the half life of ions at an initial concentration of n_o. The k' values obtained from eq. /3/ using the curves of current versus time decay measured after γ irradiation had been stopped are given in table I. The k'/μ values were evaluated from 1/i vs time curves /see Fig. 4./ which are in good agreement with those calculated from [3]

$$\frac{k'}{\mu} = \frac{1{,}6.10^{-19}}{t_{1/2}\ \sigma_o} \qquad /4/$$

The mobility of the charge carriers was measured directly by photo pulse method. The variation of mobility with reciprocal temperature is shown in Fig. 5. The mobilities indicate that the current is due to slowly moving balky ions rather than "dry" electrons, which according to Schmidt and Allen [4] and Hummel [5] have mobilities of the order of $10^2\ cm^2\ volt^{-1}\ sec^{-1}$. The variation of k'/μ with temperature can be seen in Fig. 6.

By approximating with straight lines the two different slopes of conductivity vs reciprocal temperature curves /Fig. 3/ at the lower and upper ends, the activation energy was evaluated as 0.01 eV and 0.03 eV.

Table I

Values of k′ and k′/μ for 3-methylhexane at different temperatures

temp. K^o	k′ $/cm^3 sec^{-1}/$	k′/μ /volt cm/
293		1.93×10^{-5}
283.5	7.3×10^{-9}	1.7×10^{-5}
283,5	8.92×10^{-9}	2.08×10^{-5}
272.0	7.3×10^{-9}	1.95×10^{-5}
258.0	1.3×10^{-8}	3.56×10^{-5}
245	6.55×10^{-9}	2.92×10^{-5}
223	5.5×10^{-9}	3.98×10^{-5}
210	3.63×10^{-9}	3.7×10^{-5}
183	2.7×10^{-9}	7.2×10^{-5}
163	7.4×10^{-10}	5.7×10^{-5}
154	5.0×10^{-10}	7.95×10^{-5}
143	1.55×10^{-10}	7.75×10^{-5}
111		7.9×10^{-5}
98		1.7×10^{-4}
93		1.75×10^{-4}
91		1.6×10^{-4}

It can be seen that practically no activation energy is needed for charge carrier motion. This implies impurity conduction with a quasi-Fermi level coinciding with a c or v band, depending on the nature of the impurity molecules.

The results presented here can be explained in the framework of theories [6,7] put forward for the interpretation of ionic processes taking part in irradiated insulating liquids. Deviations appears near and below the glass transition temperatures.

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FIGURE CAPTIONS

Fig. 1 Equipment for sample preparation

Fig. 2 The conductivity cell

Fig. 3 The variation of electrical conductivity with reciprocal temperature for gamma-irradiated 3-methylhexane

Fig. 4 Reciprocal current versus time curves for ^{60}Co gamma-irradiated 3-methylhexane. E = 16,2 kV/cm. 1-143°K, 2-163°K, 3-210°K, 4-258°K, 5-283°K

Fig. 5 Mobility vs reciprocal temperature as measured by flash-conductivity method.

Fig. 6 The variation of k'/μ with temperature for 3-methylhexane

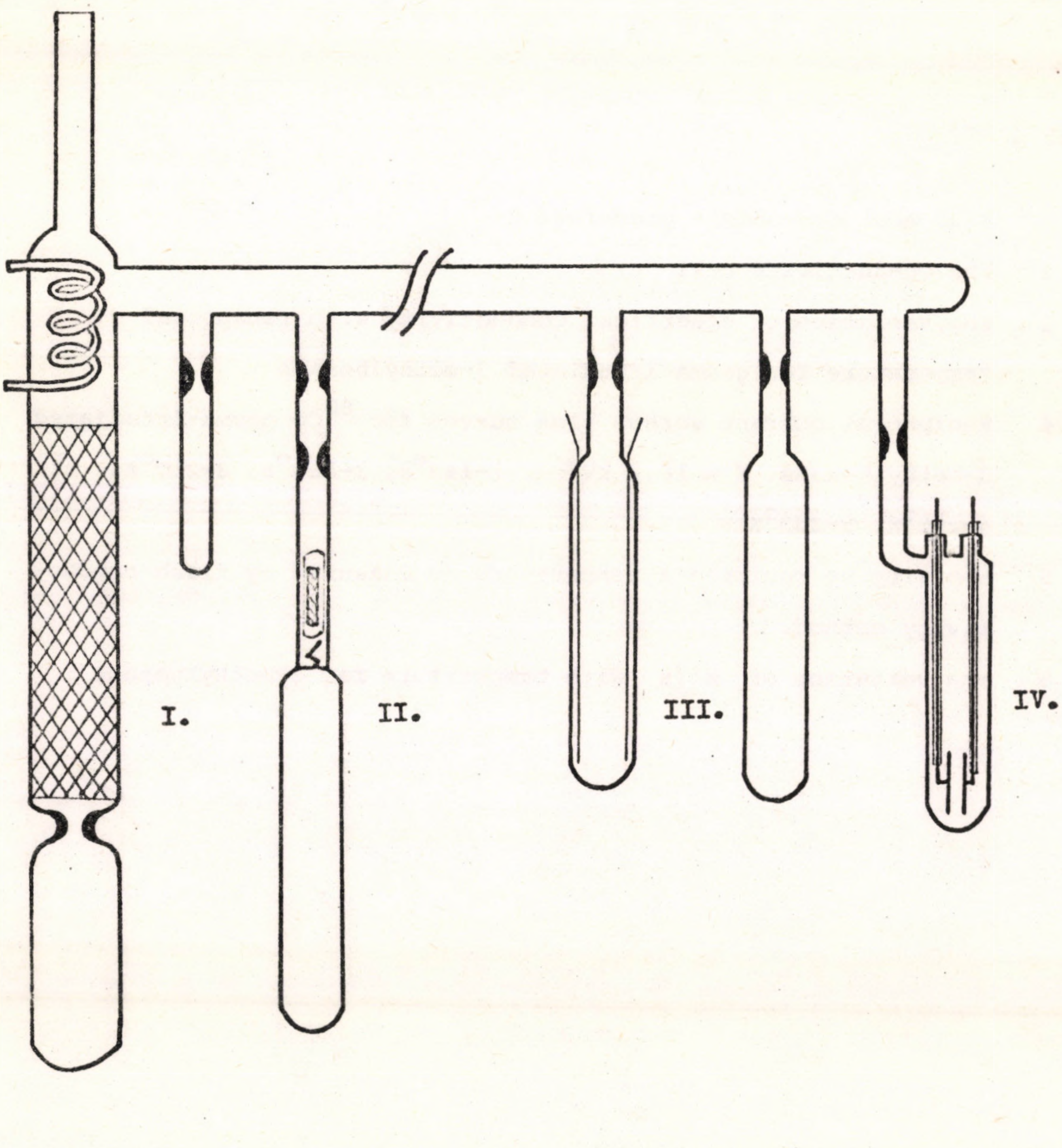

Fig. 1

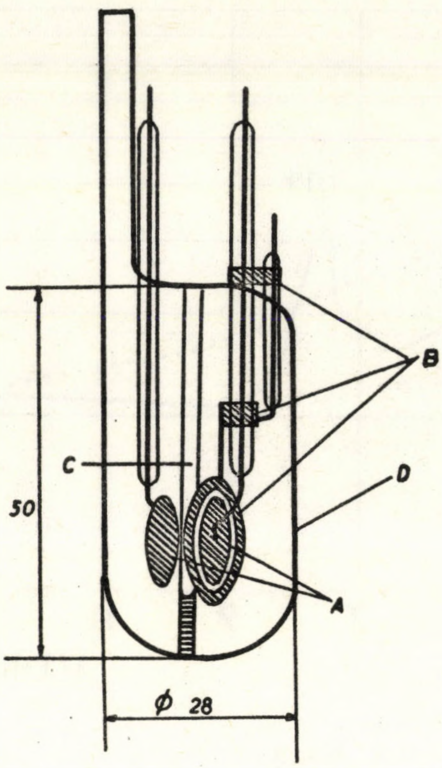

Fig. 2

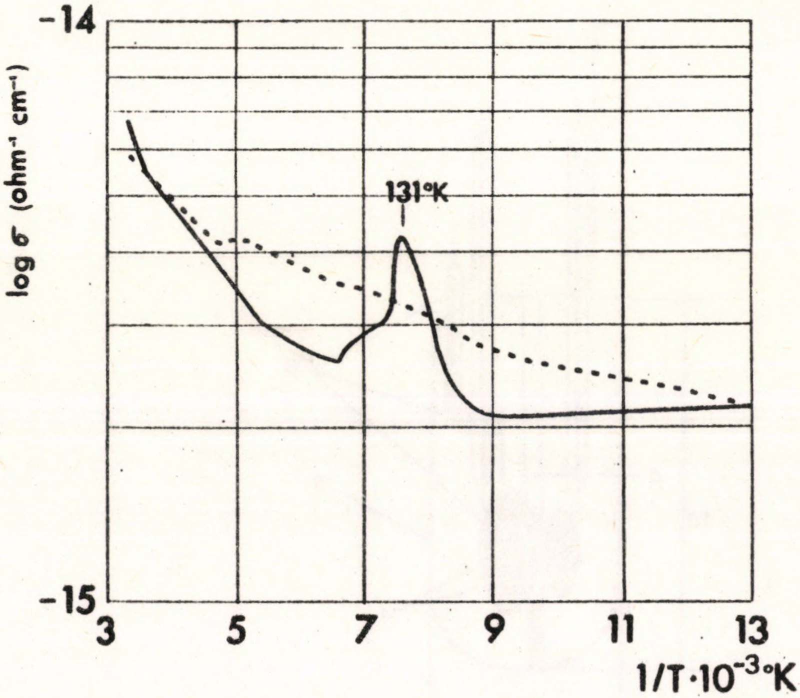

Fig. 3

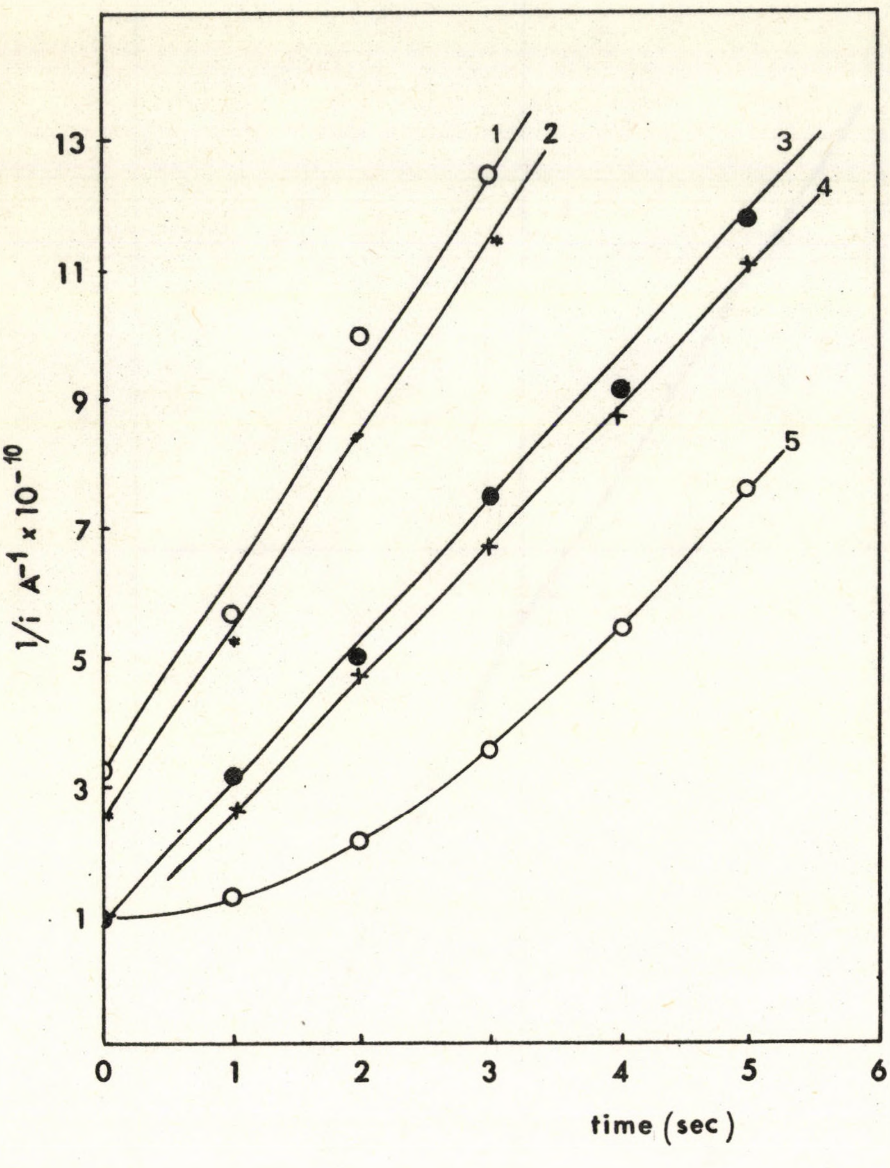

Fig. 4

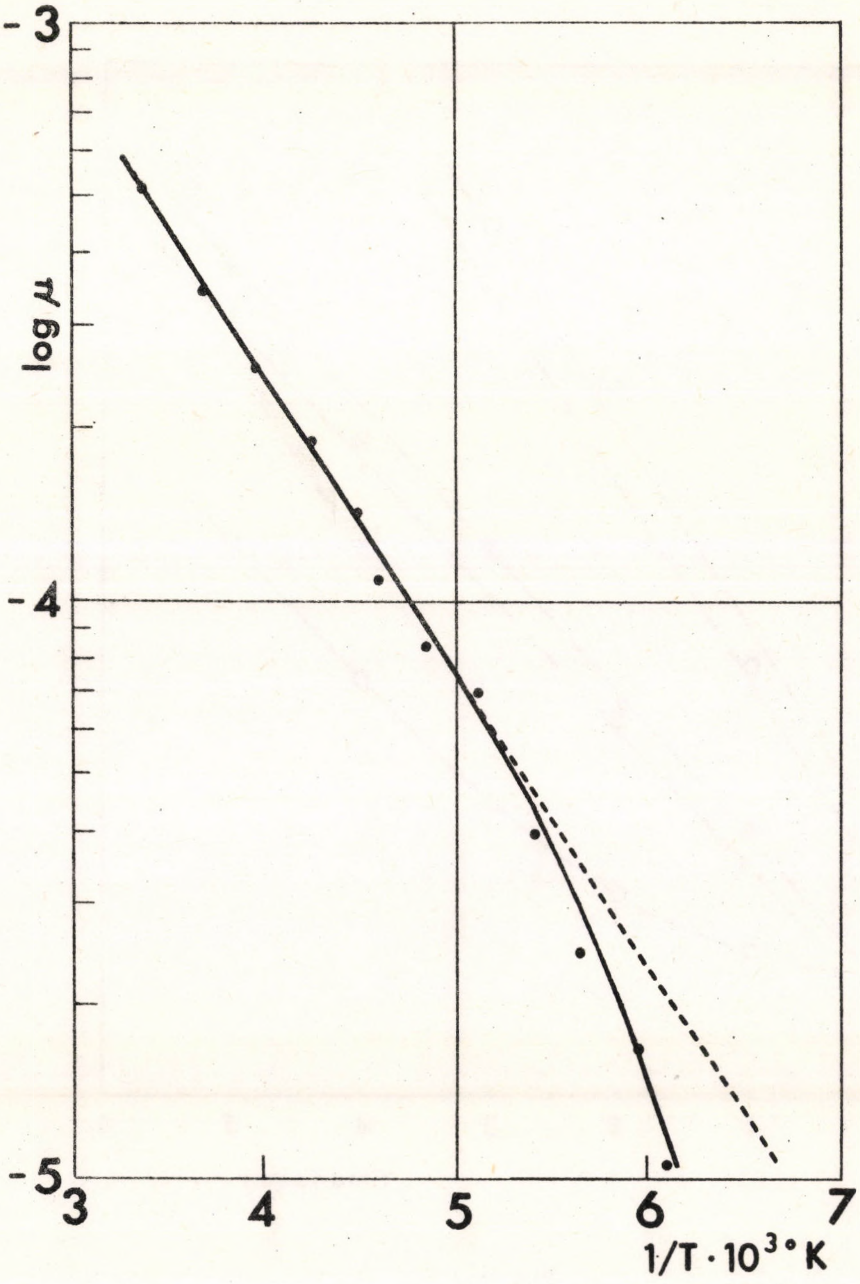

Fig. 5

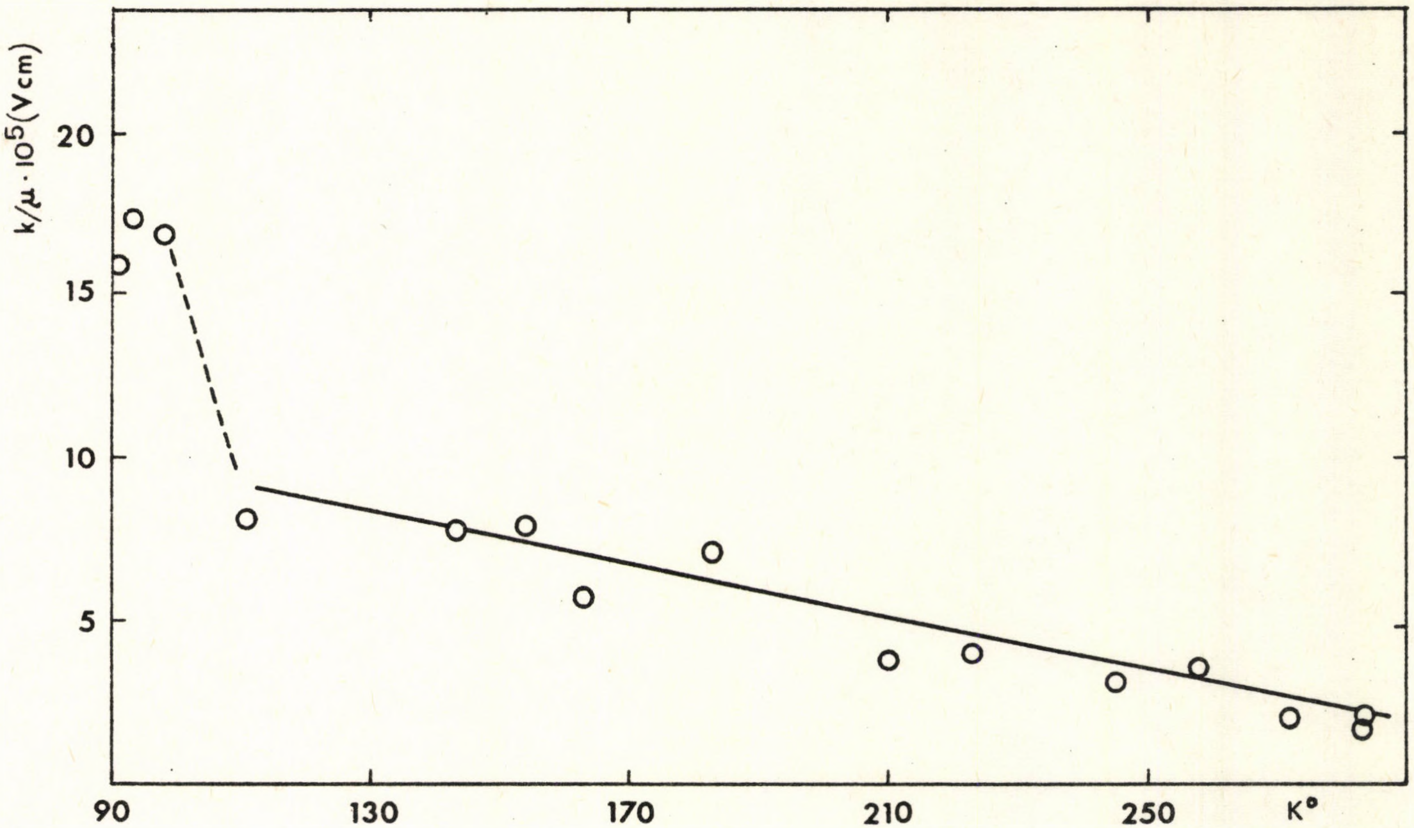

Fig.6

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