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ANALYSIS OF STIMULATED CURRENT TRANSIENTS
IN DIELECTRICS, WITH THE USE OF AN
ON LINE COMPUTER

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Thesis presented for the degree of Doctor of
Philosophy of the City University, London

1975

Abstract

The shape of current transients in solid and liquid insulants obtained by "time of flight" measurements, are often not in accordance with simple theories of the charge transport mechanism. This work is concerned with the application of the techniques of current transient analysis to the problem.

The thesis contains a study of the fundamental relationships between the motion of charge and the resulting current at the electrodes, including the transition from low level to space charge perturbed (S.C.P.) currents. The case involving static space charge is given special study for this has often resulted in the use of fallacious expressions in the past.

An iterative computer simulation of the process has been developed, using a computer in real time, to further aid the analysis of transients. This model can simulate the motion of two carriers of differing mobilities and can allow the possibility of either, or both, of the carriers being converted (or "trapped") into the other. Such a model can be easily used to represent a whole range of conditions and transport mechanisms; examples of which are given. One important application is the use of the model in an adaptive mode to produce the charge injection necessary for a given current transient.

The ionic charge transport mechanism in the liquid hydrocarbon n-hexane is studied in depth. For this, a fresh look at earlier work and results is taken from a current transient analysis point of view. This work includes experimental work undertaken, using U.V. photoinjection, to produce the best possible current transients for various conditions for analysis. To this end, a lot of attention is paid to test cell

design and use has been made of "state of the art" picoammeter techniques to achieve the best possible performance in terms of noise, risetime and sensitivity.

The experimental results allow an investigation of the linearity of, and the effects of temperature, current level, U.V. filtering, ageing etc. on the shape of the current transients. The results are compatible with earlier work and suggest that the "long tail" effect in the transients is due to impurities producing an effective range of carrier mobilities. The transients also apparently give an indication of current levels necessary for the onset of liquid motion.

Table of Contents

1.	INTRODUCTION	2
2.	CURRENT TRANSIENT ANALYSIS	
2.1	Introduction	6
2.2	The relationship between charge motion and current	7
2.3	The effect of diffusion on transients	19
2.4	The application of transient analysis to solid dielectrics	21
2.5	"Long tail" transients in solids	24
2.6	The use of current transients as an internal field probe	30
3.	DIELECTRIC LIQUID STUDIES	
3.1	Introduction	33
3.2	Ultra pure liquids	34
3.2.1	Minday, Schmidt and Davis (1971)	35
3.2.2	Schmidt and Allen (1970)	37
3.2.3	Bakale, Gregg and McCreary (1972)	
3.2.4	Allen and Holroyd (1974)	40
3.3	The electrochemical study of ionic motion in polar liquids	41
3.4	Liquid motion studies on polar liquids	49
3.5	Mobility studies (for all methods of carrier generation)	52
3.6	Methods of carrier generation	
3.6.1	Introduction	
3.6.2	Radiation	
3.6.3	High field injection	55
3.6.4	U.V. photo injection	
3.7	Liquid motion studies on non-polar liquids	57
3.8	U.V. mobility studies and current transient analysis	62
3.9	Current transients and stress distributions in liquids	81
4.	REAL TIME COMPUTING	
4.1	Introduction	84 89
4.2	The application of real time computing	90
4.3	The background to the development of the computer model	94
4.4	Description of the model and associated software	108
4.5	Simulation tests	113
4.6	Application of computer model	119
4.6.1	Introduction	
4.6.2	Application of model to stress analysis	
4.6.3	The production of S.C.P. transients	123
4.6.4	Adaptive use of model	
5.	EXPERIMENTAL METHOD	
5.1	Introduction	132
5.2	U.V. sources	134
5.3	Test cell design factors	137
5.4	Description of test cell	139
5.5	Photocathode film deposition	142
5.6	Liquid preparation	145
5.6.1	Introduction	
5.6.2	Purification	
5.6.3	Liquid degassing method	147
5.7	Picoammeter - design techniques	151
5.7.1	Introduction	
5.7.2	Risetime	154
5.7.3	Noise	157
5.7.4	Sensitivity	
5.7.5	Risetime measurement	158
5.7.6	Performance figures	160

5.8	Measuring apparatus	
5.8.1	Introduction	
5.8.2	Picoammeter head	
5.8.3	Control box and associated equipment	163
5.8.4	Performance of the picoammeter	164
5.8.5	Processing of current transients	
5.9	High voltage supplies	166
6.	RESULTS	
6.1	Introduction	168
6.2	Background to the results	169
6.3	Summary of relevant data	171
6.4	Current transients	172
6.4.1	Amplifier distortion	
6.4.2	Effect of different liquid samples	
6.4.3	Effect of time	174
6.4.4	Effect of voltage	
6.4.5	Effect of current magnitude	175
6.4.6	Effect of filtering	
6.4.7	Forward and reverse transients	
6.4.8	Effect of temperature	176
6.4.9	Linearity of transients	
7.	DISCUSSION	
7.1	Discussion of the experimental current transients	207
7.1.1	Variation between liquid samples	
7.1.2	Bulk ionisation	
7.1.3	Linearity of the transport mechanism	208
7.1.4	Near S.C.L. current transients	
7.1.5	Effect of temperature	
7.1.6	Evaluation of the range of mobilities	211
7.2	Comparison with previous results	212
7.3	A recent paper on the tail effect in solids	214
8.	SUGGESTIONS FOR FURTHER WORK	216
9.	CONCLUSIONS	217

APPENDIX A	218
APPENDIX B	221
APPENDIX C	223
APPENDIX D	226
APPENDIX E	231
APPENDIX F	237
APPENDIX G	241
ACKNOWLEDGEMENTS	244
REFERENCES	245

CHAPTER 1

INTRODUCTION

1. INTRODUCTION

The conduction mechanism of induced charge carriers in amorphous solids and liquid insulants is not at present well understood. This thesis is a study of the principles and application of the technique of current transient analysis, which can provide insight into the nature of these processes.

Such studies are of importance because of the potential engineering value of these materials. For example, knowledge of the properties of carrier conduction in solids, due to photoinjection is applied in the common office photocopier.

The importance of an understanding of the properties of insulating liquids cannot be overstated in an age concerned with energy conservation; for example, the problems and costs of energy transmission could be eased if it was known how to improve the breakdown performance of the various hydrocarbon oils.

The work in this thesis concentrates on the liquid phase and includes an experimental study of photoconduction in the hydrocarbon n-hexane. Nevertheless, much of the work will be relevant to solid dielectrics as both phases produce similar problems.

The central problem concerns the shape of the stimulated current transients; these do not accord with the present simple theories. The shape due to a pulse injection of a layer of charge should be rectangular. Instead of an abrupt cessation of current, however, the transients exhibit a long decay tail. Whereas the workers in the solid phase have sought to explain in terms of the conduction mechanism, liquids researchers have tended to treat this as unfortunate side effect of their measurements. The reason for this is partly historical.

The insulating oils used in industry are a mixture of various oils. If these oils were sufficiently pure, they would exhibit no intrinsic conduction at all. In practice, they are so impure as to prevent meaningful experimental work because of the extreme inconsistencies between different workers' results. They sought to overcome this by choosing a particular hydrocarbon (n-hexane) and sufficiently purify it, to produce meaningful results which could then be applied to the industrial oils. One property which was subject to a lot of attention, was the mobility of induced charge carriers. This is the constant of proportionality between the velocity of the carriers and the applied electric field. Such a study, it was hoped, would give an insight into the conduction mechanism, and hence into the breakdown phenomenon.

Unfortunately, actual measurements produced widely different values for such a fundamental property. Various theories were put forward to explain this, such as the effect of liquid motion which would enhance the results. These measurements produced the distorted transients mentioned earlier, but as the experimentors were more interested in producing a single mobility figure, and as the current measurement problems were severe, this was neglected.

This all changed dramatically in 1969, when work on ultra purified liquid revealed the presence of a carrier of speed about a 1,000 times faster than the earlier measurements. It was soon realised that this new carrier was the free one, and the slower carriers were ions.

It is extremely difficult to purify the liquids sufficiently well, and as yet this has only been achieved in a few laboratories; so work on the less pure liquids is still of major importance, because such liquids are much closer to the oils used in practice.

The use of current transient analysis to investigate the distorted curves is a novel approach to liquids, although it has aroused much interest in the solids field. The work of this thesis attacks this problem in two ways.

The first is to reperform experimental measurements using the best possible current measuring techniques in order to produce far better transients than have been obtained previously. Methods will be adopted to eliminate some of the forms of distortion already suggested.

Secondly, an adaptable iterative computer model will be developed to aid the interpretation of the experimental transients. For unless the conduction mechanism is simple, it is extremely difficult to predict what transient shapes will be produced. This is especially true for solid dielectrics.

It might, at this stage, be useful to note that the calculations in this thesis were made using the following values for the relative permittivity(ϵ_r) and viscosity (η) of n hexane at 20°C.

$$\epsilon_r = 1.89$$

$$\eta = 3.26 \times 10^{-4} \text{ P.}$$

CHAPTER 2

CURRENT TRANSIENT ANALYSIS

2.1 INTRODUCTION

Current transient analysis is the study of the motion of carriers in materials, so as to provide insight into the mechanism of carrier transport. The method involves enhancing the population of carriers in the material under an applied field, measuring the resulting current and then relating this to the physical processes which have occurred.

This process can be thought of in terms of having a black box to which a stimulus is applied producing an output. From such an input and output it is designed to predict what is happening inside the black box.

Fundamental to such investigations is the relationship between the motion of a charged carrier and the current it produces in an external circuit. The purpose of this chapter is to study this in depth.

In general this technique has not been widely applied to research in liquids, so a certain number of examples will be given for results from the solid phase. The major difference between the two states is that it is possible for mobile charge to get rigidly trapped in a solid, forming fixed space charge. But in liquids, if a charge is captured, it will merely form a slower moving carrier. However, some of the problems in solids and their interpretation will have an equivalence to those in liquids. This is not to say that stationary space charge does not occur in liquids, but if it does, it is a dynamic equilibrium rather than the static equilibrium of solids.

The chapter includes a section on the theory of the use of a current transient to map the internal field distribution of a medium, which in turn could be used to give the space charge distribution.

2.2 THE RELATIONSHIP BETWEEN CHARGE MOTION AND CURRENT

To perform current transient analysis, charge carriers have to be generated as a function of time and of known spatial position. The function of time is usually either a pulse, step or a square waveform. It is normal to generate the carriers in a layer which is very thin compared to the width of the medium, but sometimes they are generated throughout the medium.

The normal electrode configuration is to sandwich the dielectric between two plane parallel conducting electrodes which should be sufficiently large compared to the cross sectional area of the dielectric to represent two infinite parallel planes. If this condition is fulfilled it is possible to use the expression given by Shockley (1938) relating the motion of a charged carrier to the resulting current i , in an external circuit

$$i = \frac{q v}{d} \quad 2.1$$

where q is the charge of the carrier

v is the velocity of the carrier

and d is the electrode separation

Nothing yet has been stated as to the cause of motion; it is irrelevant to the validity of the above expression.

Now the technique of current transient analysis involves the application of a potential difference to the electrodes which produces an electric field in the gap. This field applies a force to the free charge, which causes it to move. As the motion is sufficiently slow in all practical cases, the problem can be treated as an electrostatic one (which is a condition for equ. 2.1 to hold). Hence the force F on the charge due to a field E is given by

$$F = q E \quad 2.2$$

Now in most cases the resistance to motion is proportional to velocity v , so that for a given species of carrier v is directly related to the field E by

$$v = \mu E \quad 2.3$$

The constant of proportionality is known as the mobility of that carrier.

Thus equ. 2.1 now becomes

$$i = \frac{q}{d} \mu E \quad 2.4$$

The relationship between the field E and the applied voltage depends on the quantity of charge between the electrodes, because if sufficient charge is present it will distort the stress distribution from its constant level which occurs in the absence of space charge.

If the quantity of charge is so low that the field can be assumed to be unaltered then

$$E = \frac{V}{d} \quad 2.5$$

where V is the applied voltage.

This gives from 2.4

$$i = \frac{q \mu V}{d^2} \quad 2.6$$

This expression, which applies in the absence of space charge effects, is commonly applied to the determination of the mobility of the carriers and is the fundamental relationship of current transient analysis. If all the carriers have the same mobility equ. 2.6 shows that the external current at any instant is directly proportional to the number of free carriers, irrespective of their position.

The time taken for a carrier to traverse the gap d is known as the transit time τ given by

$$\tau = \frac{d}{v} \quad 2.7$$

$$= \frac{d^2}{\mu V} \quad 2.8$$

Thus if a method is found to measure the transit time then it is possible to calculate the mobility of the carriers knowing the applied voltage and the gap width. This is the basis of "time of flight" measurements.

It is now possible to show the characteristics of the current waveforms due to the different methods of carrier generation discussed earlier. The various waveforms are shown in fig. 2.1 all plotted against time.

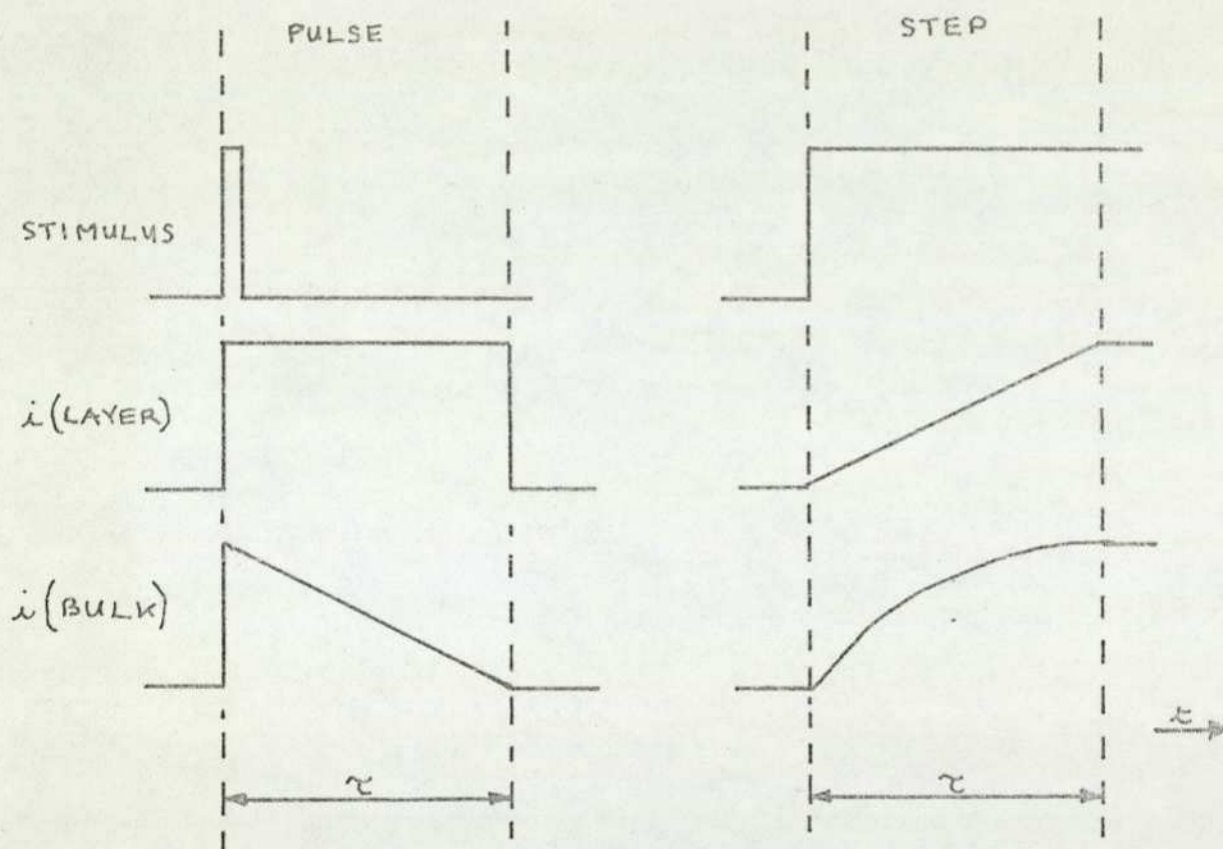


Fig. 2.1 Charge injection rates and their resulting current transients.

The waveforms for a pulse injection of charge in a layer near one electrode show the response to be rectangular in shape. This is because all the carriers take the transit time to reach the other electrode and when they all reach it the current abruptly ceases.

Now as space charge effects are assumed to be negligible, the relationship between injection and current is linear and the principle of superposition applies. So a step injection can be considered to be the integral of pulse injection, hence the resulting current is the integral of the rectangular shape, a ramp which levels off after the transit time. In physical terms what happens is that the number of carriers in the gap linearly increases up to the transit time, after which it remains constant because as many carriers are leaving the dielectric as are being created.

The corresponding cases for carriers being generated throughout the bulk of the dielectric can similarly be deduced. For the case of pulse injection, carriers generated near the collector will be removed almost immediately, but carriers on the opposite side of the gap will take the transit time to be removed. Hence the number of carriers linearly decreases with time and the current is a decreasing ramp.

The case of step injection is more complicated, but the current has been shown by Hudson (1946) to be

$$i = \frac{2k}{\tau} t \left(1 - \frac{t}{2\tau} \right) \quad 2.9$$

for values of time t of $0 \leq t \leq \tau$ (i.e. up to the transit time).

Where k is the final value of the current after the transit time. Such a method giving a parabolic relationship with time is seldom used in practice because it is not easy to interpret the current shape in terms of the transit time or other effects. It has importance for the experimental work in this thesis, as such a curve can be due to the undesirable effect of bulk ionisation in photo-electric injection measurements. Belmont (1973) erroneously stated the effect would be to produce a constant (pedestal) current; this shows the importance of being clear about these fundamental concepts.

Although most experimental work is carried out in the regime free of space charge effects, it is important to appreciate how space charge

effects can modify the current transients. Not only to be able to differentiate these effects from other causes, and appreciate when they occur, but because valuable information can be gained from measurements on space charged perturbed (S.C.P.) transients.

Fig. 2.2 shows how a large amount of charge alters the field from the linear distribution(dotted), in the absence of charge, to the non-linear distribution.

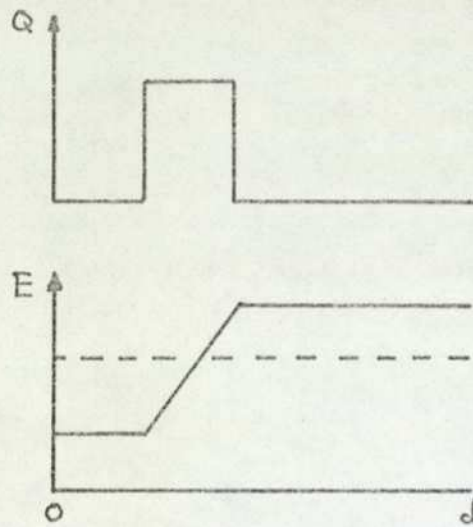


Fig. 2.2 The effect of space charge on the stress distribution.

The one dimensional Poisson's law gives the relationship between the charge and stress distribution

$$\frac{dE}{dx} = + \frac{\rho}{\epsilon} \quad 2.10$$

It follows that one side of the charge is under the influence of a stronger field than the other side. This causes the layer of charge to spread out, and the front edge to traverse the gap in a time shorter than τ , while the rear edge takes longer. Thus making it complicated to predict the resulting current transient.

One of the first papers on this was by Many and Rakavy (1962), who considered step injected space charge limited (S.C.L.) currents. Now S.C.L. transients are the limiting case of S.C.P. transients and are the situation when the maximum amount of charge possible is injected into the dielectric from one electrode. The injection mechanism is assumed to be ohmic, and hence introduce instantaneously sufficient charge to bring the stress at the injecting electrode down to zero, thus stopping the injection of further charge.

The analytical solution was derived from three equations:-

- (a) the law of conduction

$$J(x,t) = q\mu n(x,t) E(x,t) \quad 2.11$$

- (b) the continuity equation

$$J(x,t) = -q \frac{\partial n(x,t)}{\partial t} \quad 2.12$$

- (c) Poisson's law

$$\frac{\partial E(x,t)}{\partial x} = \frac{q}{\epsilon} n(x,t) \quad 2.13$$

where J is the conduction current density

x the distance across the gap

n is the number of carriers

and (x,t) indicates a quantity is a function of time and distance

They also normalised the variables into dimensionless numbers, which is now a standard practise as follows:-

- (a) distance as a fraction of the gap width L

$$x = \frac{x'}{L} \quad 2.14$$

- (b) time in terms of the transit time (in the absence of space charge effects)

$$t = t' \frac{\mu V}{L^2} \quad 2.15$$

(c) similarly the field

$$E = E' \cdot \frac{L}{V} \quad 2.16$$

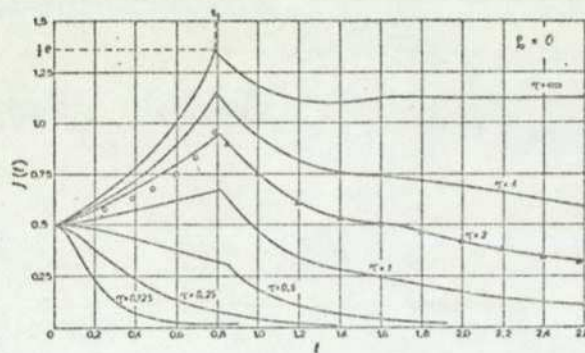
(d) the charge in terms of that quantity of charge, if evenly distributed across the gap, would reduce the cathode stress to zero

$$Q = q_n \cdot \frac{L^2}{\epsilon V} \quad 2.17$$

(e) current in terms of the current the above charge would cause if under unit field

$$J = J \cdot \frac{L^3}{\epsilon \mu V^2} \quad 2.18$$

The resulting current transient is shown in fig. 2.3



The time dependence of the SCLC density for insulating crystals characterized by various trapping times, and $E_0 = 0$.

Fig. 2.3 S.C.L. transients (after Many and Rakavy)

The current has an initial value of .5 because at the instant of charge injection, the charge's front edge is under the influence of unit field while the rear edge is at zero field strength. As this bulk of charge moves away from the injecting electrode the current increases up to a peak value of about 4/3 at a time of $t = .787$.

At this point of time, the front edge of the charge has just reached the collecting electrode. From then on the current decreases slightly to level out at the final steady value given by Shockley and Prim (1953) of

$$J = \frac{q}{8} \frac{\epsilon \mu V^2}{L^3} \quad 2.19$$

giving a normalised value of 1.125.

To give some idea of the typical current levels involved in slow carrier studies in liquid n. hexane, the S.C.L. current density for a gap of 1 cm., a cross sectional area of 1 cm^2 , a voltage of 1Kv and a mobility of $10^{-7} \text{ m}^2/\text{v}.\text{sec}$ is $I = 186 \text{ pA}$ (i.e. $1.86 \times 10^{-10} \text{ A}$)

Many and Rakavy's analysis was limited to step injection S.C.L. transients. Papadakis (1967) extended the subject to S.C.P. transients caused by pulsed charge injection. Instead of starting from the basic equations after Many and Rakavy, Papadakis based his method on starting with the spatial position of the charge to work out the field distribution. From this he worked out the subsequent charge motion and hence the current. Fig. 2.4 shows the resulting transients for various injection levels.

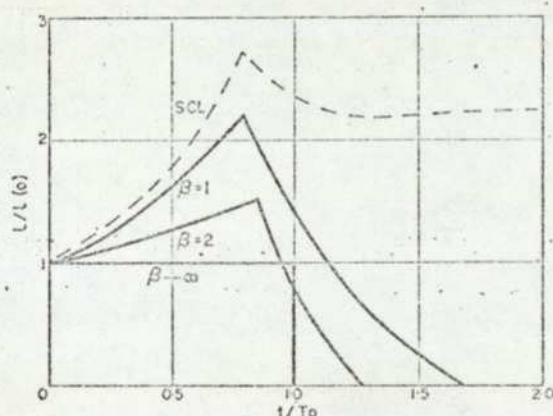


Fig. 2.4 Pulse S.C.P. transients (after Papadakis).

The injection levels are expressed in terms of a parameter β given by

$$\beta = \frac{E_0}{\epsilon_0} \quad 2.20$$

where E_0 is the space charge free field and ϵ_0 is the difference in field strength across the charge layer at a time $t=0$.

Thus β can have a minimum value of 1 (associated with maximum space charge distortion).

For β tended to infinity the current is the expected rectangular shape which would be expected in the absence of space charge effects. As β approaches 1 and space charge effects become more dominant the transients exhibit more of a peak, the time to reach the point of inflection changes from 1 and tends towards the S.C.L. value of .787, and the curve exhibits more of a tail. The dotted line corresponds to the S.C.L. current. The curves give a good illustration of how space charge effects alter the time of flight measurement.

A further development by Batra et al (1970) was to provide the relationship between the internal stress distribution and time.

A good summary of the transition from transient to steady state currents was provided by Mort and Scher (1971) for both small and large signals.

Fig. 2.5 shows the transition from small signal to S.C.P. currents, extending the results of Papadakis,

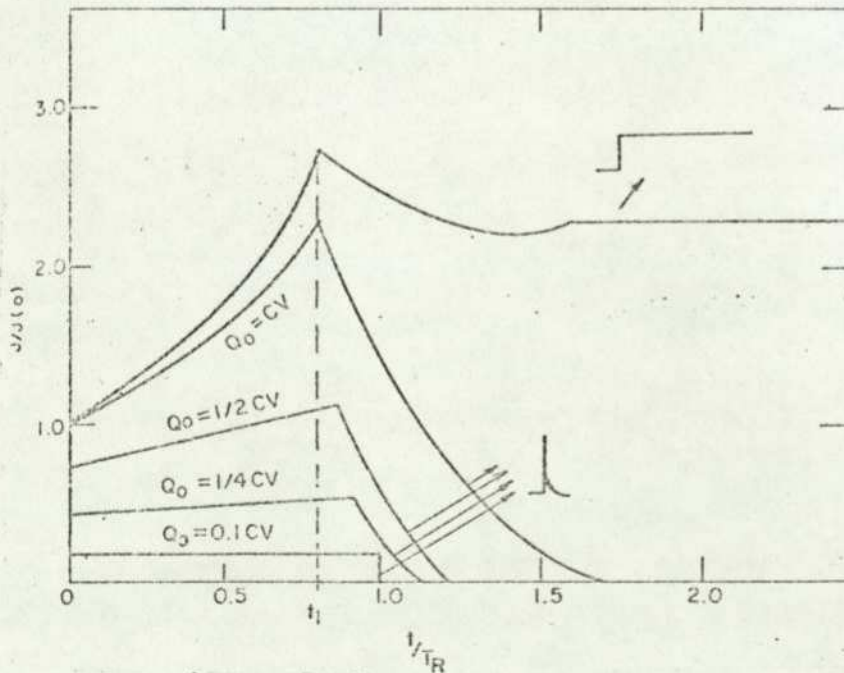
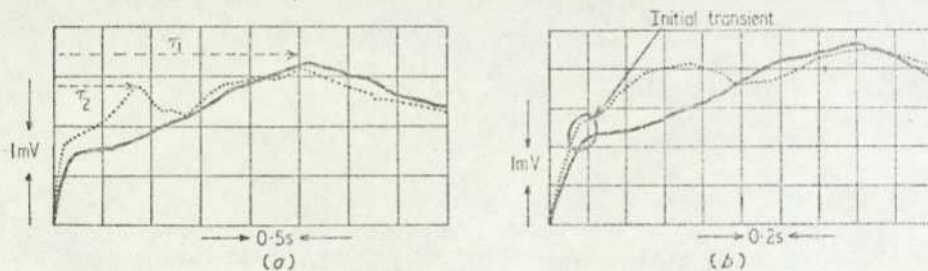


Fig. 2.5 The transition from small signal to S.C.P. transients.

Here the injection level is expressed in terms of CV, which is the maximum charge which can be injected under pulse conditions. The curves reveal that the transients get distorted for injection levels in excess of 1/10 of the maximum possible.

Such space charge distortion has not always been fully appreciated. Thus Allen and Hummel (1966) at first thought their measured distorted transients to be due to space charge effects until they realised that the charge levels involved were too small for that to be so. Taylor (1972) used the transients from step injection of charge shown in fig. 2.6 to calculate the mobilities of carriers in liquid n.-hexane and further to postulate the existence of two -ve charge carriers.



Collector current transients recorded from the storage oscilloscope. The polarity of the oscilloscope input was reversed for the negative transient to enable both waveforms to be displayed on the same axes. — positive carriers, ... negative carriers. Injection voltages: (a) 3.5 kV (—) and 3.0 kV (...); (b) 5.5 kV (—) and 4.75 kV (...).

Fig. 2.6 Step injection transients of Taylor (1972).

Now the currents approach a value of 100nA, huge currents, yet Taylor makes no mention of possible space charge distortion. It cannot be said without knowing more about the cell whether space charge effects would have occurred, but the curves show the hallmarks of S.C.L. current i.e. an initial value corresponding to .5 and a peak of about 4/3.

Two other relationships concerning charge motion and current are contained in appendices A and B.

Appendix A concerns the relationship between the displacement current density produced at a distance from a moving charge. It is included because it is a very fundamental relationship and it is demonstrated that it can be applied to show that a charge moving in free space does not create a displacement current. It is only when electrodes are present that a displacement current is produced.

Appendix B gives a modified form of equ. 2.1, to be applied when the medium has not constant permittivity throughout. An application of this would be to the work of Buttle (1969) and Hewish and Brignell (1972) where the medium is a sandwich of gas and liquid. Both these relationships were derived by J.E. Brignell and the proofs in the appendices are the author's independent verification of them.

Before concluding this section, the situation where both pulse and bulk injection can occur has to be considered. When photo-injecting charges into a dielectric there is always the possibility of carriers being created in the bulk of the medium. The process of bulk ionisation will produce carriers of either sign in equal amounts and the shape of the resulting transients will depend on the relative mobilities of the three carriers produced.

If the three mobilities are equal then it is easily seen that the total current for pulse injection will be the sum of the two waveforms shown in fig. 2.7

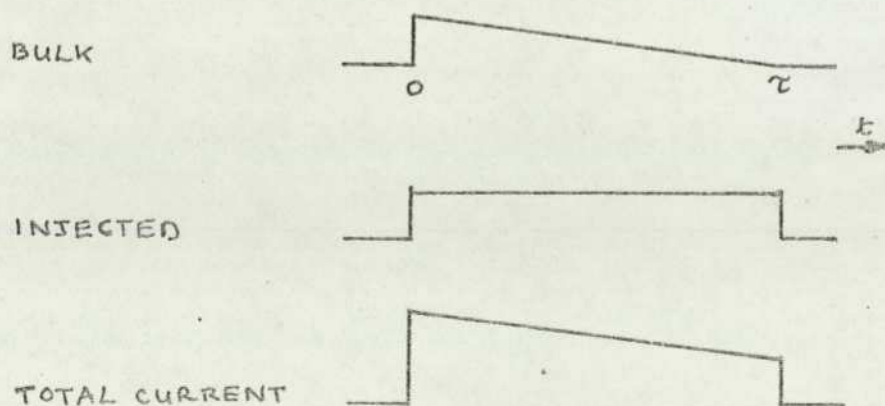


Fig. 2.7 Bulk and injection generated transients (equal mobilities).

The curves for the case of step injection will not be considered because the shape of the resulting transients (which would be the integral of the pulse transient) would be hard to interpret.

The situation when one of the bulk generated carriers has the same mobility as the injected carrier may be split into two.

(a) the third carrier is slower than the other two; shown in fig. 2.8(a)

(b) the third carrier is faster than the other two; shown in fig. 2.8(b)

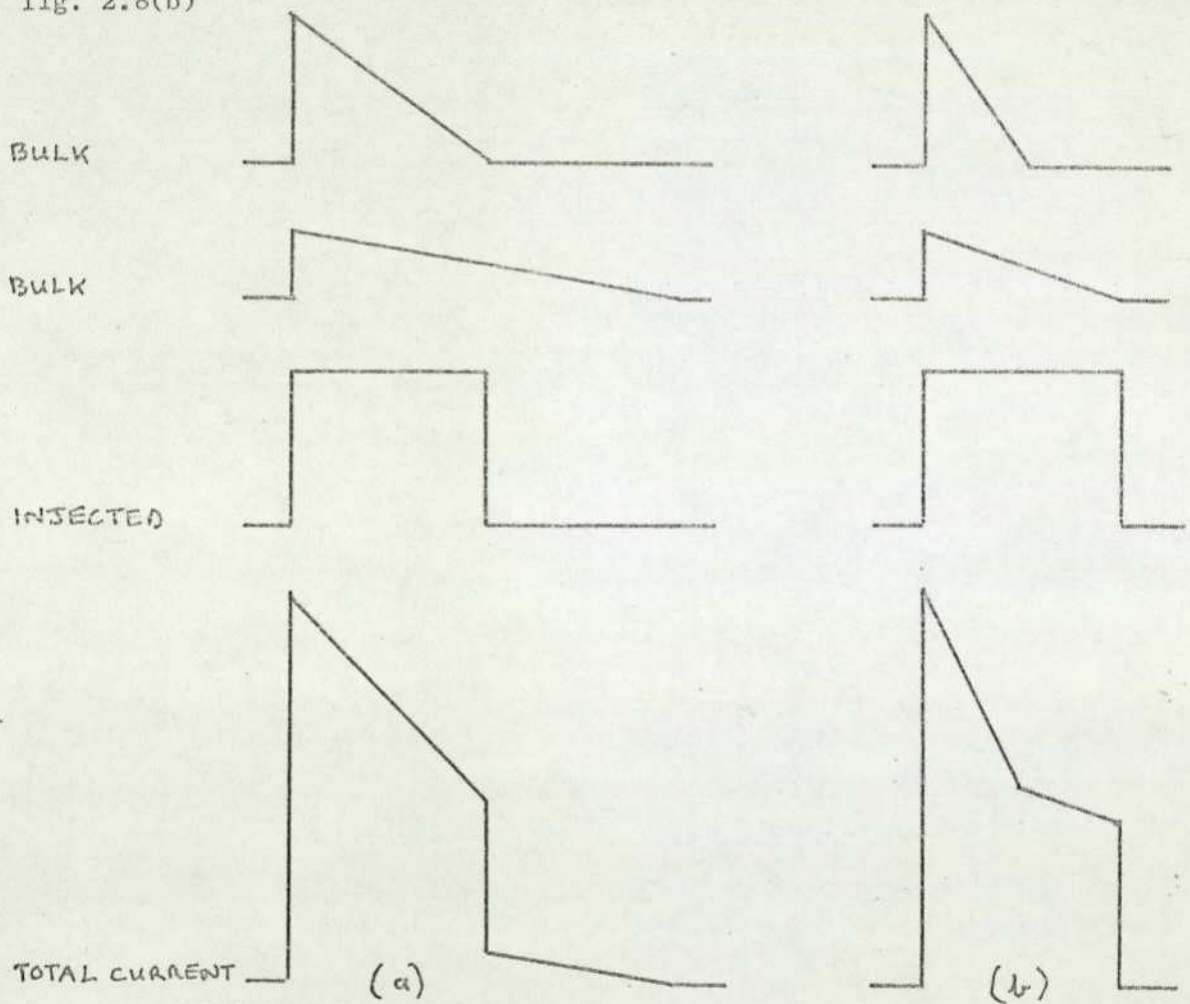


Fig. 2.8 Bulk and injection generated transients (unequal mobilities).

It is still possible to analyse the total current into its respective components. But if in practise the transients become rounded then their interpretation becomes very much harder. Factors which could cause such rounding must be understood in order to assess their effect in practice.

2.3 THE EFFECT OF DIFFUSION ON TRANSIENTS

Diffusion is the major theoretical explanation employed to explain the rounding of transients. There are others of an experimental nature which will be discussed subsequently.

Diffusion is the tendency of charge to spread from regions of higher to lower concentration. This is illustrated in fig. 2.9

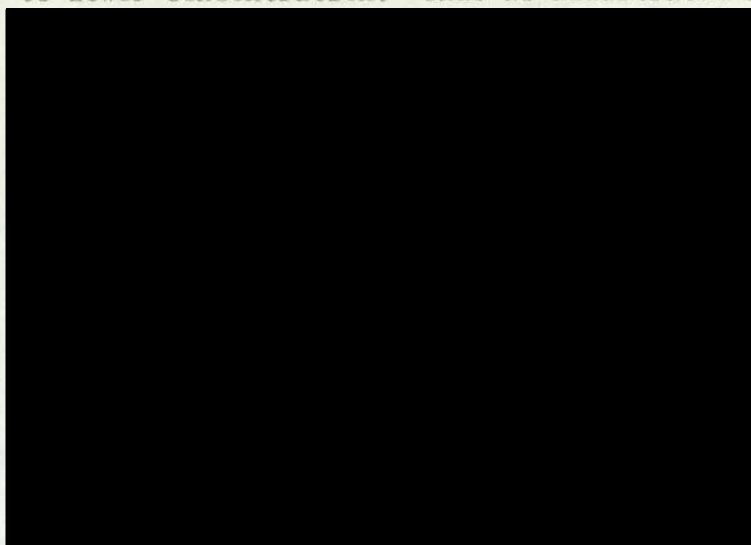


Fig. 2.9 The effect of diffusion

The original rectangular shape of the pulse of charge gradually changes as the layer traverses the gap. The effect on the current transient is also shown, the dotted line indicates the ideal shape in the absence of diffusion.

The mean square distance of displacement has been shown by Einstein to be

$$\bar{x}^2 = 2 D t \quad 2.21$$

where D is the diffusion constant and is related to the mobility by

$$D = \frac{\mu k T}{q} \quad 2.22$$

Thus the distance is independent of the field strength and only related to time. Hence the effects of diffusion will be more apparent

at lower field strengths, which imply longer transit times. A measure of the effect of diffusion is to compare the ratio of the R.M.S. diffusion distance to that distance moved by the charge due to the applied voltage V . This has been shown by Buttle (1969) to be

$$V \approx \sqrt{\frac{500}{V}} \% \quad 2.23$$

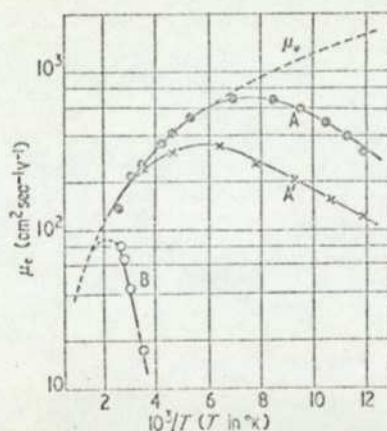
So for a voltage as low as 100v the above distortion is about 2% showing that its effect, for higher voltages, on the transient will be very slight.

This conclusion does not apply necessarily in the space charge affected regime, where very low field can occur near the injecting electrodes, or with grids (Hewish and Brignell 1972). This former case has been studied by Lampert and Edelman (1964) and by Schilling and Schachter (1967).

2.4 THE APPLICATION OF TRANSIENT ANALYSIS TO SOLID DIELECTRICS

Some insight into the usefulness of current transient analysis can be gained from its application to solid dielectrics.

A characteristic feature of charge transport in solids is that the carriers (either holes or electrons) can be trapped. It is convenient to classify these traps into deep and shallow types according to the energy needed to release the carrier. For the case of shallow traps the charge gets trapped and released several times during its motion across the dielectric. Thus the mobility is more a measure of the time spent in traps than of the charge motion, and it is usually found that there is a complicated relationship between mobility and temperature. Fig. 2.10 shows the results of such measurements on CdS (from Spear 1969).



Temperature dependence of the electron drift mobility in CdS specimens. Solid curves have been calculated from eq. (9). Crystal A: $\epsilon_t = 0.043$ eV, $N_t = 3.5 \times 10^{15} \text{ m}^{-3}$; crystal A': $\epsilon_t = 0.030$ eV, $N_t \approx 10^{17} \text{ cm}^{-3}$; crystal B: $\epsilon_t = 0.16$ eV, $N_t = 5 \times 10^{16} \text{ cm}^{-3}$. μ_0 denotes the lattice mobility.

Fig. 2.10 Temperature dependence of electron drift mobility in CdS (after Spear)

The dotted line shows how the mobility varies against temperature in the absence of traps. The points on the graph are of actual

measurements and the solid line is a plot of the following temperature of the mobility μ

$$\mu = \frac{\mu_0}{\left[1 + K \exp\left(\frac{\xi}{KT}\right) \right]} \quad 2.24$$

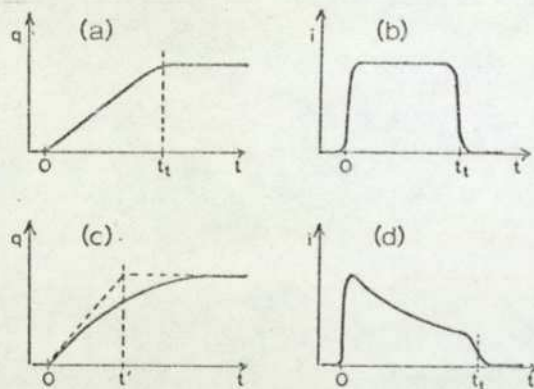
where μ_0 is related to the trap free mobility

ξ is the trap depth (in eV)

and K is a constant.

This shows that at high temperature the traps have little effect on the mobility measurements because the carriers if trapped are quickly released. From such measurements it is possible to estimate the carrier mobility, the trap density and the trap depth.

When deep trapping occurs during charge transit, the charges are not released by the transit time (by definition). Fig. 2.11 shows the effect this has on pulse transients.



Typical pulse shapes observed in drift mobility experiments when the carriers are generated close to the top electrode, $\delta \ll d$. (a) integrated signal, $CR \gg t_t$, (b) current pulse, $CR \ll t_t$. In both (a) and (b) deep trapping is absent. (c) and (d) show corresponding pulse shapes when the drifting carriers interact with deep centres.

Fig. 2.11 The effect of deep trapping on charge and current transients (after Spear)

Curves (a) and (b) are the usual transients measured in the V and I modes respectively in the absence of traps. Curves (c) and (d) show the corresponding waveforms when deep trapping occurs. The measurement in the V mode (i.e. by integrating the current waveform) is curved and

it is very hard to establish the transit time. It is much easier to determine this from the current transient and also from its decay to obtain information on the trapping time.

To observe these effects at least some of the carriers must cross the sample. Often this means the use of very thin samples and hence fast transit times. A review of such experimental methods has been provided by Spear (1969).

The relevance of this study of solids to field of liquids is that it is possible to have trapping phenomena occurring in liquids. The fundamental difference is that a carrier, if trapped, does not become fixed but forms a larger and slower moving carrier. However, the considerable amount of work done on trapping in solids will prove useful in the development on a computer model to be explained later.

2.5 "LONG TAIL" TRANSIENTS IN SOLIDS

It will be shown in the review on liquid dielectrics that the current transients exhibit a long tail after the apparent transit time of the carriers. This however is not unique to liquids, and often occurs in experimental work on solids. Let us therefore look at this effect in solids and at the various explanations put forward, to see whether they have any relevance to liquids.

One of the first examples of this was given by Kepler (1960) who measured the drift mobilities of electrons and holes in anthracene crystals. He used the pulsed layer method and a typical curve obtained with either carrier is shown in fig. 2.12. Here the initial rise of the transient is limited by the risetime of the measuring equipment. The flash duration was negligible compared to the transit time. The curve exhibits a long tail which Kepler was unable to account for. To investigate the problem, he tried interposing insulating mylar sheets between the electrodes and the sample, but without effect. This showed that the tail was not a consequence of electrode-sample interface effects. He was further able to prove that surface currents along the outside of the crystal were not responsible either. Space charge effects were eliminated by showing that when high injection rates were used it was possible to control such effects by varying the field strength but without effect on the tail.

In spite of this tail, Kepler was still able to perform mobility measurements by using the sudden drop in current level as indicating the transit time. He thus obtained a field independent mobility for both holes and electrons of about $10^{-2} \text{ m}^2/\text{V}\cdot\text{sec}$. Such results suggest that the tail effect is not an anomaly but a characteristic of the transport mechanism.



Fig. 2.12 Time of flight measurements in anthracene (after Kepler).

Another example is that of Tabak (1970) who performed time of flight measurements on vitreous selenium over a wide temperature range. Measurements were made in the V (integrated I) mode at high temperatures, in the normal I mode at low temperatures and in both modes at intermediate temperatures. Fig. 2.13 shows such a measurement in the Vmode at 15°C .

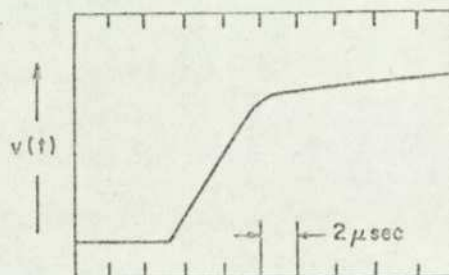


Fig. 2.13 Integrated current transients (V mode) in selenium at 15°C (after Tabak)

This curve shows an abrupt change of slope clearly indicating the transit time but the subsequent slope is not zero indicating that the current has not returned to zero.

Fig. 2.14 shows measurements in both modes at -95°C . The time T_r indicates the transit time obtained from the change of slope in the I mode. It is interesting to see that the integrating process (V mode) obscures this point and also the subsequent tail.

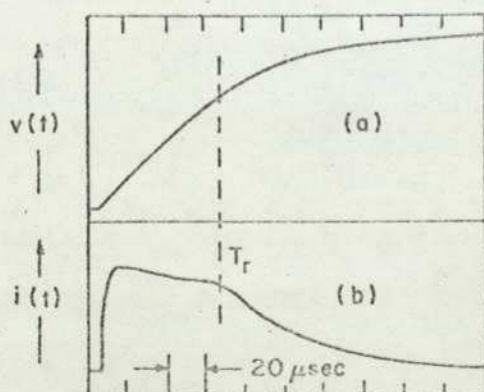


Fig. 2.14 Transients in selenium at -95°C
(a) V mode (b) I mode (after Tabak)

For comparison fig. 2.15 shows two measurements in the I mode on the same time scale but (a) is at -52°C and (b) is at -89°C and the field strengths have been adjusted to give roughly the same transit time.

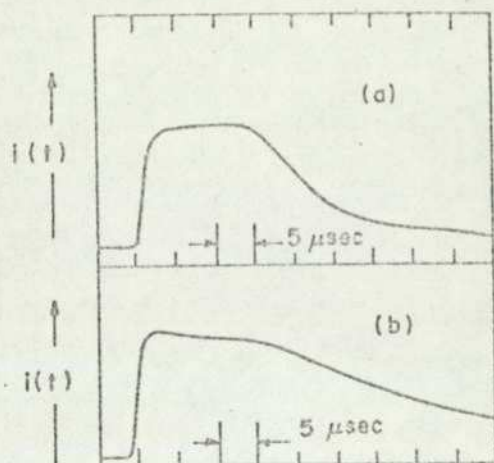


Fig. 2.15 Current transients in selenium (after Tabak)
 (a) at -52°C , $V = 140\text{V}$
 (b) at -89°C , $V = 700\text{V}$

This clearly shows how the tail becomes more pronounced at lower temperatures. Tabak was able to measure field independent mobilities for temperatures above -70°C ; but not below this, indicating a different form of transport mechanism at low temperatures. He suggested that the cause was an electric field and temperature dependent release rate from shallow traps.

Silver et al (1971) published an alternative explanation of Tabak's results. They put forward a model which explained the tail effect and the apparent field dependent mobility by including surface, as well as bulk trapping. In effect surface trapping would be equivalent to a delay in carrier generation.

To illustrate the effects on transients, they used a simplified Monte Carlo method to generate the transient shown in fig. 2.16.

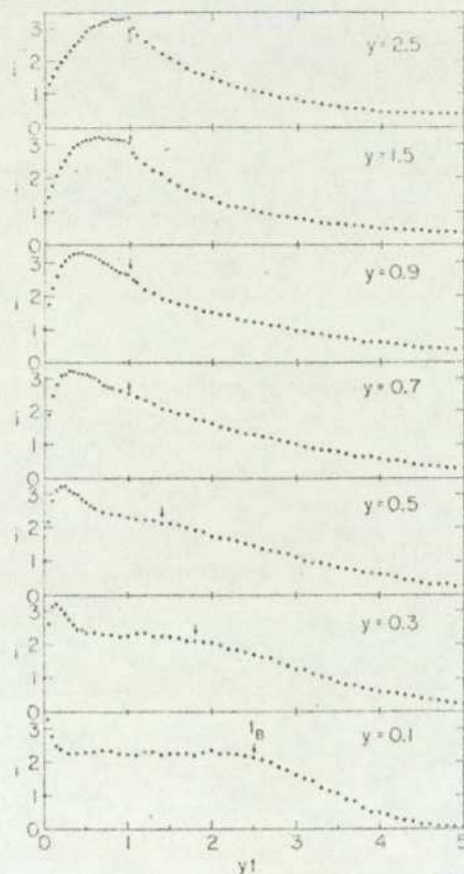


Fig. 2.16 Predicted current transients when both surface and bulk trapping occurs (after Silver et al).

The parameter γ is analogous to the applied voltage. The time scale is normalised in terms of the transit time.

The effect of this is to disguise the real transit time (which is unity on the time scale) and give an apparently longer one. Although it does give the tail effect, it should be realised that surface trapping effects both the beginning and end of the transient, thus some of the curves have not got the usual square shaped front edge.

Scharfe (1970) reported on charge transport of holes in vitreous As_2Se_3 . Fig. 2.17 shows the current transient due to a pulsed time of flight measurement. The curve exhibits a long tail and a point of inflexion. Now Scharfe chose to interpret the curve in terms of carriers with a distribution of mobilities, where the break point represents the transit time of the fastest carrier.

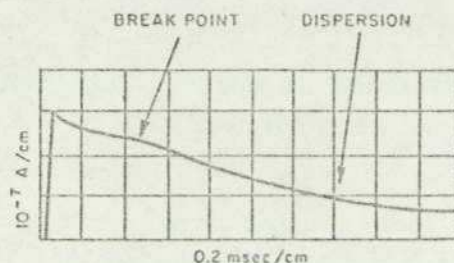


Fig. 2.17 Transients in As_2Se_3 (after Scharfe)

In physical terms Scharfe explained that the mobilities are controlled by shallow traps; the range of mobilities being due to a distribution of the release times. Fig. 2.18 shows the transient for the same conditions as fig. 2.17 except that the light intensity is two orders of magnitude higher.

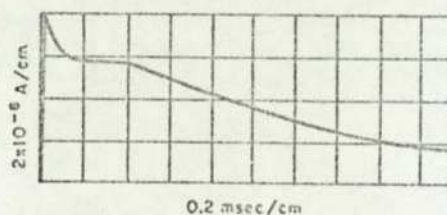


Fig. 2.18 Transient in As_2Se_3 due to high light intensity (after Scharfe).

The mobility of the fastest carriers, as calculated from the break point, was not found to be independent of field strength for a given sample. In fact for the same field strength there was found to be a fivefold variation in mobility in different thickness samples.

The wavelength of the light was varied and the effect on the transients is shown in fig. 2.19.

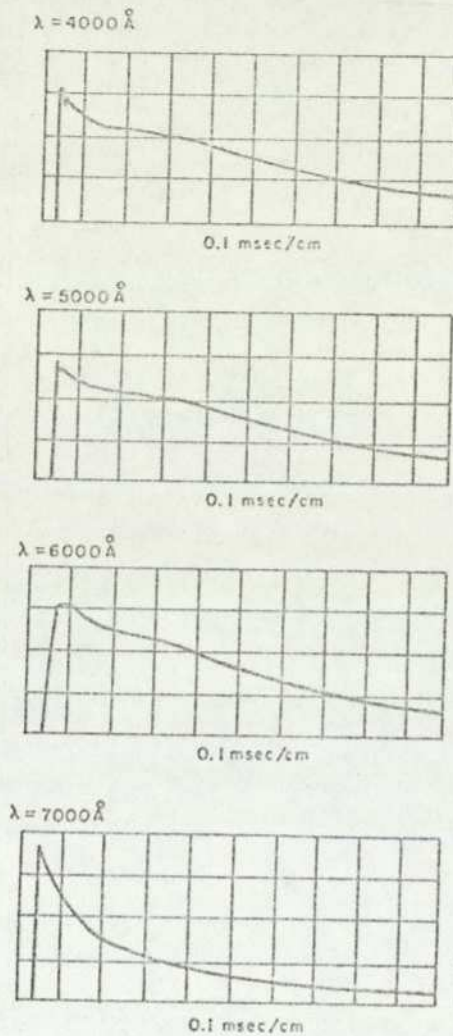


Fig. 2.19 Effect of wavelength (absorption) on transients (after Scharfe)

Now for wavelengths between $4,000 \text{ Å}$ and $6,500 \text{ Å}$ the light hardly penetrates the material and hence all the carriers are generated in a thin layer. As the wavelength approaches $7,000 \text{ Å}$ the light penetrates the bulk of material. This is reflected in the transients and as the wavelength increases, the transients change from being crudely rectangular to that of a decreasing ramp. This agrees well with the theory discussed in section 2.2.

2.6 THE USE OF CURRENT TRANSIENTS AS AN INTERNAL FIELD PROBE

An initial aim of this study was to investigate the internal field distribution in liquids by the use of a charge probe. The idea behind this is to perform a pulsed time of flight measurement on the liquid dielectric and to relate the shape of the current transient to the internal field. Fundamental to this is the relationship between the motion of the charge q , the field strength $E(x)$ at that point in the medium and the resulting current i .

Two papers were published concerning the field distribution in liquids by this method, Bloor and Morant (1968) and Jackym et al (1971). They both used the same formula:-

$$i = \frac{q E(x) v}{V} \quad 2.25$$

where v is the carrier velocity

then using equ. 2.3 gives

$$i = \frac{q \mu E(x)^2}{V} \quad 2.26$$

Now Brignell and Buttle (1971) gave the following formula:-

$$i = \frac{q v}{d} \quad 2.27$$

giving

$$i = \frac{q \mu E(x)}{d} \quad 2.28$$

These authors were unaware of the contradiction when they published and J.E. Brignell and this author published a letter (Brignell and Evison 1973) which resolved this controversy and showed equ. 2.28 to be correct. This letter is reproduced in appendix C.

After this letter was published the authors received a communication from J.B. Gunn which cited a paper he had published to resolve the same dispute, but in the semiconductor field. This spurred this author to do a thorough literature search on the problem, and this revealed that this same problem had occurred in several independent fields of research.

More serious was the fact that people seemed to have had great difficulty in accepting equ. 2.28 to be correct and had published successive papers raising new methods to prove equ. 2 to be correct. However, each time a reply would be published reasserting the validity of equ. 2.28.

It thus seems likely that sooner or later someone will try to disprove this work. To forestall this appendix D contains a brief summary of most of the published work on this controversy, indicating some of the erroneous theories and their subsequent demise.

CHAPTER 3

DIELECTRIC LIQUID STUDIES

3.1 INTRODUCTION

This chapter is not intended to be a general review of the work on dielectric liquids, for there are already several available. For example general reviews are given by Buttle (1969) and Bloor (1970); on liquid motion effects by Essex (1968) and Taylor (1972); and on the stress distribution in liquids by Pollard (1972).

Instead this chapter will have the following aims:

- (a) to review the very recent work on ultra pure liquids
- (b) to look at those areas which have been relatively neglected by workers in the field,
- (c) to review past results in terms of current transient analysis.

It will become apparent from this that the theory of charge transport in liquids is still very inadequate, a comment which applies to both fast, or "free", carrier transport in ultra pure liquids and to slow, or ionic, transport in less rigorously purified liquids. The reason in the former case is that the experimental results indicate a rather complicated transport mechanism, whereas for slow carriers, it is because of the large amount of experimental work, which is often contradictory, but which has been carried out without an appreciation of the implications of the fast carrier studies. For, most of such work was performed before 1969, when the existence of fast carriers was first revealed, and since then most workers have focused their attentions on this new development.

3.2 ULTRA PURE LIQUIDS

It is only comparatively recently that experimental studies have revealed the existence of carriers whose mobility were several orders of magnitude greater than had previously been measured. It was soon appreciated that these fast carriers were characteristic of the motion of an untrapped or "free" electron, as opposed to the slow carriers which were then shown to be due to charge scavenging molecules permanently capturing the free electrons. This mechanism is similar to that of charge transport in the liquidified noble gases, such as Argon, which had been understood for sometime.

The reason it took some time to identify the corresponding situation in the liquid hydrocarbons was due to the difficulty of their purification. It seems that it is comparatively easy to rid Argon of charge scavenging impurities, of which nitrogen and oxygen are the most important. However, for liquid hydrocarbons it is much more difficult and research workers tended to presume that they had purified their liquids sufficiently well so as to show that hydrocarbons did not exhibit the fast carriers of the noble gases.

But as Minday et al (1971) points out, it only requires an impurity concentration of one part in one hundred million to scavenge all the electrons in normal experimental conditions. To put this in perspective, the usual practice of drying liquids by silica gel, results in a water content of about one part in a million, and it can thus be appreciated why fast carriers were not normally observed. It is, however, possible to observe fast carriers in other than ultra pure liquids by the use of fast response measuring equipment after the fashion of Bakale et al (1972) whose equipment had a risetime of the order of 10nS.

In fact, to obtain a sufficient purity for an injected electron to have a good chance of traversing the electrode gap without capture, it

is necessary to go to extreme measures. This involves a bakeable apparatus to achieve an ultra high vacuum of the order of 10^{-9} torr to get a "clean" system, and the use of molecular sieves and chemisorbtion techniques to purify the liquid.

The relevance of this to a study of slow carrier transport is due to the fact that these fast carriers can be converted into slow carriers by the controlled addition of charge scavenger impurities. Thus information is provided on slow carrier transport, free of the major problem associated with the work on less pure liquids. This problem is that work is always liable to the criticism that there might be some odd impurity present which produces different results from other researchers.

3.2.1. Minday, Schmidt and Davis (1971)

This paper reported a study of fast carriers generated in hydrocarbon liquids by photoelectric injection. It gave results and put forward theories on the carrier and the photoelectric injection mechanism.

A purification system with an integral measuring cell was used, thus enabling measurements to be performed at various stages of liquid purity. Hence as the purification proceeded, the conductivity fell from 10^{-13} to $10^{-19} \Omega^{-1} \text{cm}^{-1}$ and the photoelectric current density rose from 10^{-11} to 10^{-8} A/cm^2 .

Fig. 3.1 shows the current transient in hexane obtained from the application of a step waveform of U.V. light.

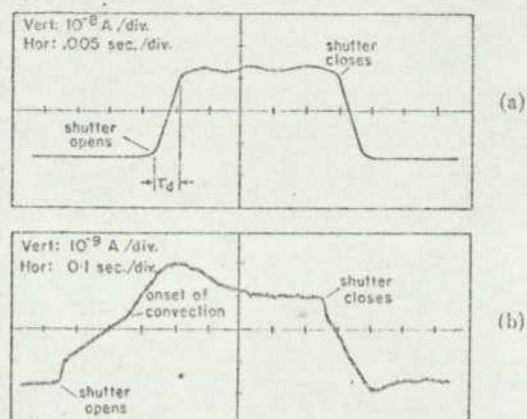


Fig. 3.1 Transients in hexane (after Minday et al)
(a) ultra pure (b) less pure

The upper curve, for an ultra pure liquid, is the expected ramp shape and τ_D , the transit time, indicates a well defined fast carrier mobility of $.07 \text{ cm}^2/\text{V. sec.}$

The lower curve is for a less pure liquid where ions are the predominant charge carrier. On opening the shutter, the current rises in a steep ramp due to the transit of electrons across the gap which elude capture. After this the current rises with a ramp of reduced slope due to the ionic carriers, and it is possible to make a crude estimate of their mobility of $8 \times 10^{-4} \text{ cm}^2/\text{V. sec.}$

It is possible to make an estimate of the S.C.L. current for these conditions to be about 2,800pA. In fact the current rises to peak of 3,800pA indicating the possibility of both space charge and liquid motion effects. The off transient is much less distorted and produces a mobility figure of $17 \times 10^{-8} \text{ m}^2/\text{V. sec.}$ indicating that something such as liquid motion is taking place and enhancing the apparent mobility results.

The effect of adding either oxygen or water to a liquid which exhibited fast carriers produces slow carriers with a mobility of about $10 \times 10^{-8} \text{ m}^2/\text{V. sec.}$. It appears that the addition of the such charge scavengers produce results indistinguishable from a liquid in the early stages of purity.

From the results, the following model for the photoelectric injection process was put forward. This model involves a cathode image barrier which the photoelectric electrons have to surmount if they are to escape, and not return to the cathode. The distance between this barrier and the cathode is a function of the field strength; for a field of 6Kv the distance is $250 \text{ \AA}$ and for 250V it is $1500 \text{ \AA}$. To overcome this barrier the carriers need sufficient energy, but if they are thermalised, i.e. slowed down by scattering processes, then they will not escape. The authors found from their measurements that the mean distance before scattering was $250 \text{ \AA}$. These results imply that the injection mechanism is instantaneous, and that it should be impossible for charge to be slowly emitted from the cathode after the U.V. has stopped.

3.2.2 Schmidt and Allen (1970)

Confirmation of the above results is provided by the work of Schmidt and Allen (1970) who produced carriers by X rays. Although the liquids were not so pure, a mobility of $.09 \text{ cm}^2/\text{V}\cdot\text{sec}$ was reported which is close to the previous result of $.07 \text{ cm}^2/\text{V}\cdot\text{sec}$. It was also found that the addition of oxygen or sulphur dioxide changed the fast carriers into slow ones.

3.2.3 Bakale, Gregg and McCreary (1972)

Further information on impurity trapping was provided by Bakale et al (1972). The liquids used were not purified to the extent mentioned earlier and hence to study the effect of trapping measurements were made in the nanosecond time scale. A parallel electrode test cell was used and the carriers were generated by a 10 n sec X ray pulse. Fig. 3.2 shows how the current decays with time as the fast carriers are trapped to form the slower ions.

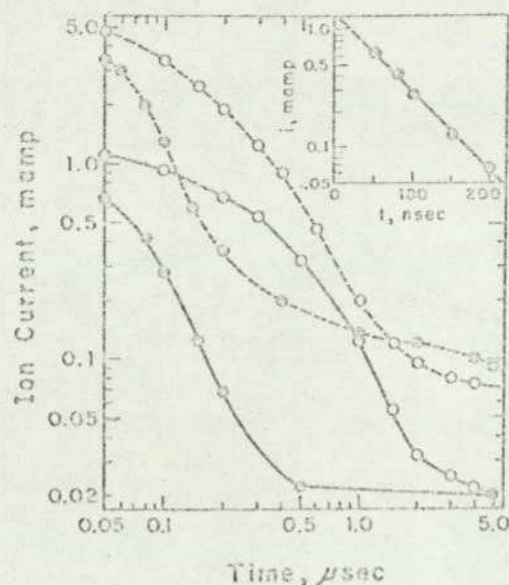


Fig. 3.2 Decay curve of fast to slow carriers in hexane (after Bakale et al)

The solid curves are for the addition to the liquid of a small quantity of biphenyl, a charge scavenging molecule. As expected the curves show it takes a reduced time for the fast carriers to be converted into the slower ions. Hence, by adding different impurities, it is possible to get a measure of their relative trapping abilities.

As the decay curve of current against time indicates an approximate exponential relationship, it is convenient to measure an effective half life time constant for the electron and relate this to the quantity of scavenger added. The expression used is

$$k_e = k_t + k_s [s] = \ln 2 \left(\frac{t_1}{2} \right)$$

where k_e is the effective pseudo first order rate constant of electron attachment which is the sum of k_t , due to the impurities already present in the hexane and k_s , due to the solute. Of course k_s is a function of the quantity of solute added.

Fig. 3.3 shows a graph of such a relationship for the scavengers CCl_4 and biphenyl.

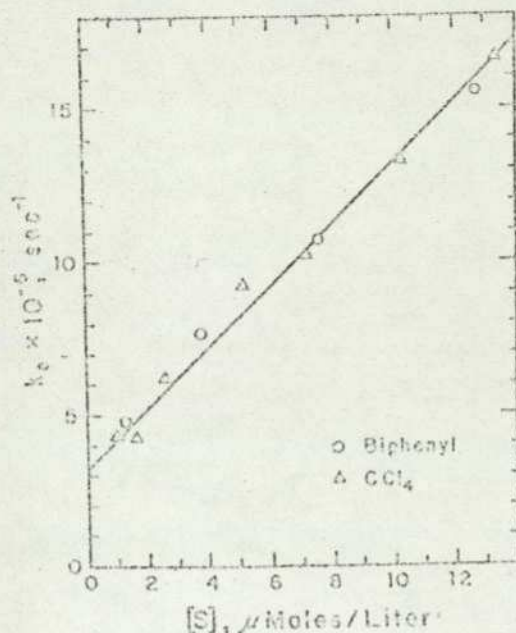


Fig. 3.3 Variation with the quantity of scavenger added of free carrier lifetimes in hexane (after Bakale et al).

It shows that these are equally effective charge scavengers. It also gives information on the scavengers already present in the hexane, before the solute was added. If such impurities are as efficient at scavenging as CCl_4 , then their concentration must be of the order of $3 \mu\text{Moles/litre}$.

The value of K_s obtained from the slopes of such graphs are as follows:

Solute	$K_s \left(\frac{1}{\text{M Sec}} \right)$
CCl_4	10^{12}
Biphenyl	10^{12}
I_2	$2 \cdot 10^{11}$
H_2O	$> 10^9$
Benzene	$< 10^9$
$\text{O}_2, \text{CO}_2, \text{N}_2\text{O}, \text{SF}_6$	$> 2 \cdot 10^8$

The reason for the lower limit on O_2 , CO_2 etc. is that saturated solutions were used, i.e. concentrations of the order of 1 Mole/litre. Such high concentrations scavenge the electrons so quickly that it was found difficult to resolve the decay half lives. So the values for these molecules are very much the lower limit.

These results do show that several molecules are very proficient at charge scavenging. Oxygen is probably the most important of these and it thus appears likely to be the actual scavenger in most experiments on air saturated liquids. There is also a good chance that less thorough methods of degassing will leave a sufficient oxygen concentration to form the slow carrier. But it is also clear that water and benzene, two major impurities of hexane, are comparatively unimportant in trapping electrons when in competition with other impurities.

3.2.4. Allen and Holroyd (1974)

Similar results to Bakale et al (1972) were obtained by Allen and Holroyd (1974). A major difference was that Allen and Holroyd were adding impurities to ultra pure liquids. They also used X rays and gave the theory of a method for determining the mobilities of not only the -ve ion due to the scavenger, but also of the +ve ion due to the host liquid. Experimental results by this method were promised for a later paper.

3.3 THE ELECTROCHEMICAL STUDY OF IONIC MOTION IN POLAR LIQUIDS

The work on ultra pure liquids has proved that the slow carriers in less pure liquids are ions. It is hence desirable to look at Electrochemistry to get an understanding of the motion of ions in liquids. This will give an appreciation of the ion-host liquid molecule interaction and hence a background to criticize the various carrier models. It also gives the true mobility results which will be compared with apparent mobility figures in the section on liquid motion.

Electrochemistry is mainly about the chemical study of conducting solutions. So the only liquids which can be studied are those which will dissolve salts to form ions. This ability is related to its dielectric constant, hence water, with a relative permittivity of 80, is very suitable. Other problems arise with low permittivity liquids which serves to rule out the use of liquid hydrocarbons. But there are valuable results on polar liquids such as nitrobenzene.

The basic problem is to relate the current I , due to an applied voltage V across the cell, to the concentration of the solute. If the cell has a cross sectional area A , a gap width of l , then the specific conductivity κ is given by

$$\kappa = \frac{I \cdot l}{V A}$$

Now obviously this will vary with the concentration of the salt, so it is preferable to use the molar conductivity Λ_m . This is the conductivity per molecule, defined as

$$\Lambda_m = \frac{\kappa}{c}$$

where c is the concentration in gram moles of solute per unit volume.

Finally, for the purpose of comparing ions with varying units of charge, the equivalent conductivity Λ is used,

$$\Lambda = \frac{\kappa}{c z}$$

where z is the number of units of charge.

For a given electrolyte this should be a constant, but in fact it is found to vary slightly with the concentration because of ion-ion interactions. This can be overcome by defining a limiting value of Λ , i.e. Λ_0 the equivalent conductivity at infinite dilution. This is determined by extrapolating a curve of Λ against the log of c .

Now Λ_0 is due to the sum of the contributions of the +ve ions (cations) and -ve ions (anions); the question is how to separate them. For any one cation there is a whole range of anions according to the salts available. The proportion of the total current carried by one ion is referred to as the transport number. The method used is to determine the transport number for one ion and then to determine the conductivities of the other ions from family of salts containing that ion by simple subtraction of the known ions conductivity from the sum of the two.

The equivalent conductivity is also known as the ionic mobility because of the relationship between the conductivity and the mobility is

$$\mu = \frac{\Lambda_0}{F}$$

where F is Faraday's constant (96,790).

A table of such mobility figures for various ions in different liquids is as follows:

	η	ϵ	<u>Cations</u>				<u>Anions</u>			
			Li	Na	K	H	Cl	Br	I	Pi
Water	.89	80	4.0	5.2	7.6		7.9	8.1	8.0	3.2
Methanol	.55	33	4.1	4.7	5.4		5.4	5.9	6.5	
Ethanol	1.1	24	1.6			6.4	2.3			2.6
radius ($\text{\AA}$)			.8	1.0	1.3		1.8	1.9	2.2	

where η is the viscosity (millipoise)

and ϵ is the relative permittivity.

Such figures give information on the relative importance of the two major forces which retard the motion of the ions. One is Stokes' type friction which will be greater for ions of large size and for liquids of high viscosity.

The other force is due to the ion-solute molecule interaction, which will be related to the permittivity of the liquid. This attraction between the charged ion and the host liquid molecule tends to increase the effective size of the ion. This is known as ion solvation (or hydration in the case of water). However, this is not totally a function of the permittivity as the above table shows. For the solvation effect increases in the order of water, methanol and then ethanol; the reverse order to the magnitudes of the permittivities thus showing how complicated it all is.

Stokes' law gives the relationship between the mobility of a particle (of radius r) and the liquid viscosity

$$\mu = \frac{e}{6\pi r \eta}$$

It is not widely appreciated that this relationship is only valid if the ion molecule attraction, discussed above, is negligible and if the ion size is very much larger than the liquid molecule.

Now if only the former applies than extra frictional forces arise due to a tendency for the solvent molecules to move with the ion. This is quite distinct from the polarisation phenomenon. Boyd (1963) and Zwanzig (1970) have derived an expression to account for this of the form

$$F = \frac{k}{r^3}$$

where F is the frictional force
and r is the ion radius.

This force must be added to that of Stokes' of

$$F_s = 6\pi\eta r$$

to get the total retarding force. Now this extra term becomes more significant for smaller sized ions.

An illustration of this is provided by the results of Jansen and Yeager (1973) in fig. 3.4 for ions in propylene carbonate.

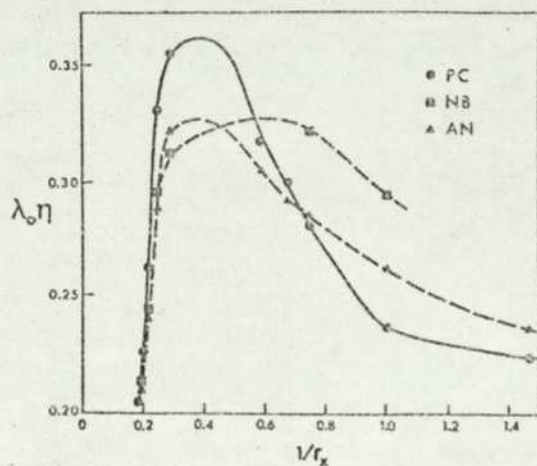


Fig. 3.4 Conductivity - viscosity products against ionic radi for various solutes (after Jansen and Yeager).

Also shown on the graph are curves for Boyd and Zwarzig's model, the two curves relate to two different values of K ; one for a slipping condition (B) and the other for sticking (A). Stokes' law (which would give a straight line through the origin) only fits the larger ions and it is Boyd and Zwanzig's model which correctly predicts a maximum in the curve.

The relevance of these studies is that they reveal how complicated is the charged ion-host molecule interaction. It has however, been assumed by several authors that such forces on an ion are negligible

compared with Stokes' friction. For example Le Blanc (1959) measured the activation energy of slow carriers in hexane to be .11eV. He argued that if the process was ionic then Stokes Walden's law would apply. This states that the product of all the mobility of the carriers and the liquid viscosity is independent of temperature, and is subject to the same constraints as Stokes' law. Now for hexane, the viscosity activation energy is .08eV, owing to the difference between the two, Le Blanc argued that the transport mechanism was not ionic and he went on to discuss a hopping electron model.

Secker (1970) has suggested that a perspective on this can be gained by comparing the measurements of the self diffusion coefficient of a hexane molecule to that of the diffusion coefficient of a charged carrier. This should give a measure of the extra forces due to the charge of the carrier.

Douglass and McCall (1958) used a spin echo method to determine this self diffusion coefficient (D) in various hydrocarbon liquids. For hexane (at 25°C) this was found to be $4.21 \times 10^{-5} \text{ cm}^2/\text{sec}$. Stokes Einstein's relationship can be used to relate this to the mobility

$$\frac{\mu}{D} = \frac{e}{kT}$$

This gives an effective "mobility" of $16.8 \times 10^{-4} \text{ cm}^2/\text{V}\cdot\text{sec}$ for the uncharged hexane molecule. It is interesting to compare this with a mobility calculated from Stokes' law. This hexane molecule is cylindrical with a length of 10°A and a diameter of 5°A . If this is represented by a sphere of 7°A diameter then it gives a mobility of $7.3 \times 10^{-4} \text{ cm}^2/\text{V}\cdot\text{sec}$.

Douglass and McCall also measured the activation energy of the self diffusion coefficient to be $2.07 \pm .13 \text{ Kcal/mole}$ (.09eV); compared with that for the viscosity of 1.78 Kcal/mole (.08eV).

The authors went into this difference to some depth but came to the conclusion that much more experimental and theoretical work was needed before the mechanism of molecular motion could be established.

Now this conclusion was reached over the motion of uncharged molecules; it thus can be appreciated that the understanding of charged ionic carriers is even less understood. So any models (such as Secker's 1970) must be regarded as interesting attempts to gain an understanding of the processes involved, but it must be realised that such theories are a gross simplification of a very complex process.

3.4 LIQUID MOTION STUDIES ON POLAR LIQUIDS

Although nitrobenzene is a polar liquid, research on this liquid is of interest to workers on non-polar liquids (such as hexane). This is because it is possible to use the Kerr effect to measure the stress distribution across the liquid optically. Also, permselective membranes have recently been developed which have made purification and ion injection much easier. Since the mobility of various ions in nitrobenzene is known from conductivity measurements, it is possible to get a measure of the effect of liquid motion on mobility studies.

The research has principally been carried out by French workers who have tried to tie up "cellular" type liquid motion theories with their experimental measurements. Although they have used large injection rates some interesting results have emerged.

An analysis of the steady state injection rates necessary for the onset of cellular type motion has been given by Atten and Moreau (1970). The theory has been translated into the experimental variables of voltage and current by Hewish and Brignell (1972) and this is shown in fig. 3.5

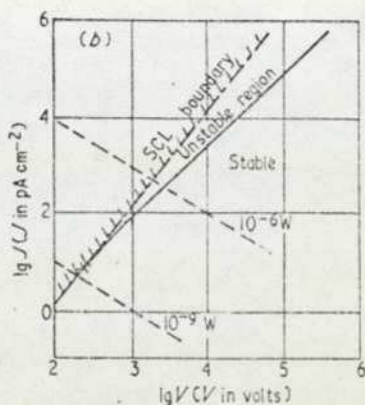


Fig. 3.5 The stability criterion with the S.C.L. condition (after Hewish and Brignell).

The actual figures are based on an assumed carrier mobility of $7.5 \times 10^{-8} \text{ m}^2/\text{V}\cdot\text{sec}$ but the important point is that the current needed for instability to occur is very close to the S.C.L. current, at any particular voltage. Further, there is a critical value of voltage below which motion should be impossible to start; this is 208V for a non polar liquid and about 16V for a polar liquid. This model has been experimentally verified for polar liquids by Fillipini, Lacroix and Tobazeon (1970).

Allen and Gosse (1969) performed measurements with near S.C.L. currents in nitrobenzene. Known ions were injected and the mobility was determined from the current transient, and a knowledge of the electric field given by the Kerr effect. Not surprisingly, the mobilities were enhanced by an order of magnitude from the true values. But what was interesting was that these apparent mobilities were found to be independent of the field strength or the electrode gap distance. This is illustrated in fig. 3.6 for protons.

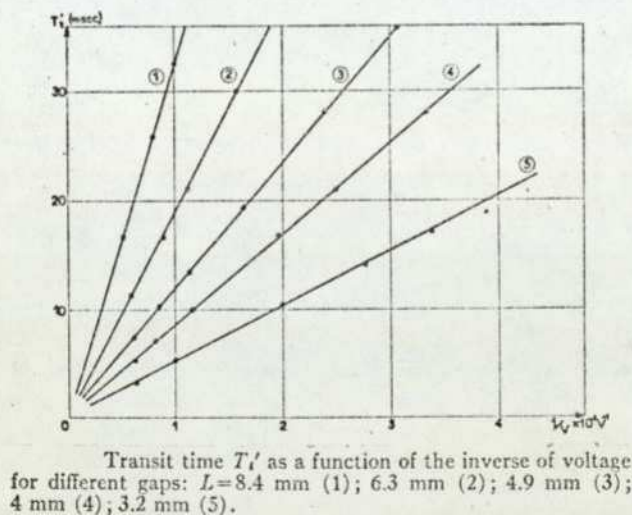


Fig. 3.6 (after Atten and Gosse).

Thus authors such as Belmont (1973) who perform mobility measurements with quite large currents (i.e. greater than 10,000 pA) in hexane, are wrong to argue that because their results are independent

of field strength and gap width then it must mean that no liquid motion is taking place.

Other results reported in the paper are listed below:

<u>Solvent</u>	<u>η</u>	<u>ρ</u> (25°C)	<u>ϵ</u> (25°C)	<u>μ_e</u> ($\times 10^8$)	<u>μ_t</u> ($\times 10^8$)
Ethanol	1.1(25°C)	.78	24.3	21	2.3
Nitrobenzene	1.0(30°C)	1.2	35	20	2.4
Propylene Carbonate	1.6(30°C)	1.2	69(23°C)	20	.9
Acetonitrile	.33(30°C)	.78	37.5(20°C)	27	10

where η is the viscosity (centipoise)

ρ is the density (g/cm^3)

ϵ is the dielectric constant

μ_e experimental mobility

μ_t true mobility

The above mobility figures relate to a C_2^- ion (except for the figures for propylene carbonate).

The true mobility results show some correlation with the liquid viscosities. It has been shown by the method of dimensional analysis (Felici 1969) that the fluid velocity is related to $\sqrt{\epsilon/\rho}$ but such a relationship is not reflected in the above results.

Hopfinger and Gosse (1971) observed electrohydrodynamic (E.H.D.) motion by the use of a Schlieren apparatus. Under similar experimental conditions to those of Atten and Gosse they found that after the onset of charge injection, the liquid put into motion and a turbulent region crossed the gap with a velocity related to $\sqrt{\epsilon/\rho}$, which for nitrobenzene gives a mobility of $55 \times 10^{-8} \text{ cm}^2/\text{Vsec}$. Just before this turbulent region has crossed the gap, the injected space charge starts moving across at constant speed. The primary mechanism of charge transport was found to be this self generated turbulence.

It is however possible to arrive at the true mobility, experimentally, by making use of the fact the apparent mobility is enhanced less during steady state than under transient conditions (i.e. switch on or switch off of injection). This was applied by Barret and Gaspard (1974).

When the authors applied a step injection of protons into nitrobenzene they arrived at a mobility of $17 \times 10^{-8} \text{ cm}^2/\text{V}.\text{Sec.}$ To measure the mobility under steady state conditions they made use of the Kerr effect to measure the stress difference across the two electrodes ($E_c - E_e$) and used Poissons law to calculate the charge ρ

$$\rho = \frac{\epsilon}{l} (E_c - E_e)$$

where l is the gap width

Then using the measured current density J it is possible to calculate the apparent mobility from

$$J = \rho \mu E$$

The results of such calculations are shown in fig. 3.7, a graph of apparent mobility against the charge ρ for various field strengths.

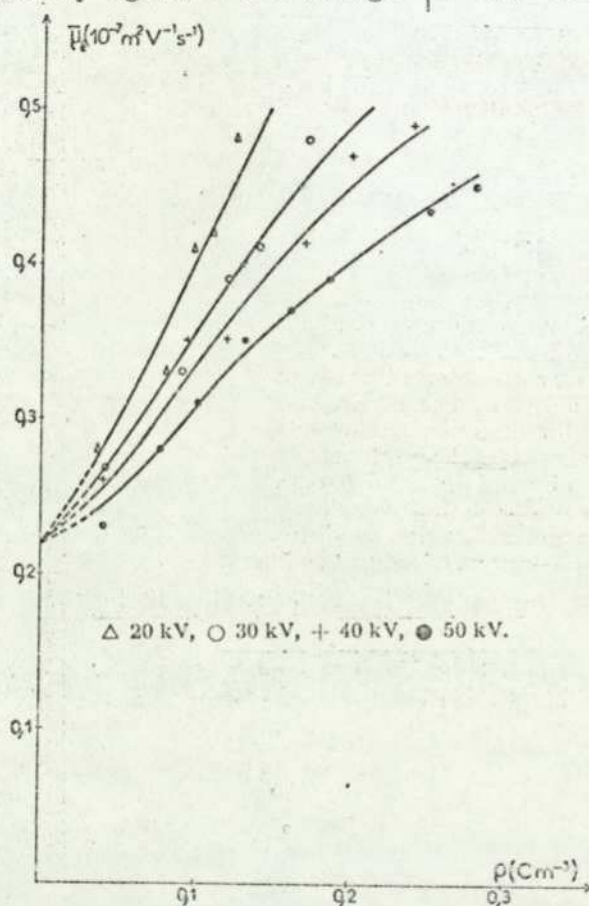


Fig. 3.7 E.H.D. mobility against current density (after Barret and Gaspard).

It can be seen that the curves when extrapolated back to zero charge give a mobility of $2.2 \times 10^{-8} \text{ m}^2/\text{V}.\text{sec}$ which compares well with the value from conductivity measurements of $2.3 \times 10^{-8} \text{ cm}^2/\text{V}.\text{sec}$. This method of extrapolating back for zero liquid motion enhancement has been applied to studies on hexane by Essex and Secker (1969) and Taylor (1972) and will be discussed in section 3.7.

3.5 MOBILITY STUDIES (FOR ALL METHODS OF CARRIER GENERATION)

The previous sections in this chapter have provided a background to slow carrier studies; in this section actual measurements in hexane will be discussed. Most of these experiments have been designed to measure the mobility of the slow carrier; the tacit assumption being that there is a single carrier mechanism which will produce a well defined mobility. So it has been assumed that once the experimental difficulties were overcome, then everyone would arrive at a single universal mobility. These experimental difficulties include liquid purification, ultra low current measurement, charge injection methods and also possible liquid motion effects.

A list of such results follows:

<u>Author</u>	<u>Method</u>	<u>Mobility</u> <u>($\times 10^8$)$m^2/V.sec$</u>	<u>Activation</u> <u>Energy (eV)</u>
Le Blanc (1959)	U.V.	11	.14
Gzowshi & Terlecki (1959)	R	13	
Chong & Inuishi (1960)	U.V.	10	.16
Terlecki (1962)	U.V.	9.8	
Gzowski (1962)	R	9.8	.78
Allen & Hummel (1963)	R	7.8	
	R	1.8	
" " (1966)	R	13	.125
Secker & Lewis (1965)	R		
Schmidt (1968)	R	9.1	
Tewari & Freeman (1968)	R	11.1	
Essex & Secker (1968)	H.F.	2.7	
" " (1969)	H.F.	1.5	<u>0</u>
Gray & Lewis (1969)	R	2.5	

<u>Author</u>	<u>Method</u>	<u>Mobility</u> <u>($\times 10^8$)$m^2/V.sec.$</u>	<u>Activation</u> <u>Energy (eV)</u>
Bloor (1979)	U.V.	7.6	
Buttle (1969)	U.V.	7.5	
"		2.5	
Taylor (1972)	H.F.	7	
"		1	
Belmont & Secker (1972)	R	11	.8
"		5.8	.8
Hewish & Brignell (1972)	U.V.	7.5	

where U.V. signifies ultra violet photo injection

where R signifies radiation injection

and H.F. signifies high field injection.

These include a wide variety of injection techniques, purification methods and mobility determination methods. Clearly the most outstanding feature is the wide variation in the results for both mobility and even more disturbing in the activation energy.

It might well be wondered as to the reason for each experimenter adding yet another result to the list without adequately explaining why his result is any better than before. The answer is that there are many parameters which could affect the result, but which cannot be accurately assessed. Hence it is easy for each author to find fault with earlier results and yet perhaps be less critical about his own work.

When it is known from the work of Bakale et al (1972) that a number of molecules is capable of electron scavenging, it is arguable whether there would be one discrete carrier in an experiment or that the same type of carrier is involved in different experiments. Another possible explanation is the effects of liquid motion.

3.6 METHODS OF CARRIER GENERATION

3.6.1 Introduction

If a hydrocarbon is sufficiently pure and free of polar impurities, it will exhibit a conductivity of the order of $10^{-20} \Omega^{-1} \text{cm}^{-1}$. This limit on the conductivity has been shown to be due to cosmic radiation inducing carriers in the bulk of the liquid. Hence all conductivity in hexane is produced by factors which are not inherent to the liquid.

To study carriers in hexane, it is necessary to produce them by one of several methods. Each method has its own virtues and these are discussed here.

3.6.2 Radiation

Radiation can be used to produce carriers throughout the liquid or in a thin layer. The problem with the latter method is the difficulty of collimating the beam to be narrow in proportion to the gap width. There is some uncertainty as to the nature of the carriers produced, especially with regard to the positive carrier, which could be a hexane ion or a fragment of the molecule. It is interesting to note that workers using ultra pure liquids have found that a preliminary exposure to radiation has reduced the effects of impurities (Schmidt and Allen 1970) though the reason for this is not fully understood. Both Belmont (1974) and Allen and Hummell (1966) have found that it is possible to get low mobility values if the liquid is not sufficiently pure. However, neither could find out what substance was removed by purification to account for this. Schmidt (1968) achieved similar results to these workers with liquid straight out of the bottle, so this phenomena is hard to explain.

With X rays, it is possible to generate extremely large currents i.e. Belmont (1973) used currents in excess of 10,000pA, regardless

of the purity of the liquid. Hence compared with the work using U.V. injection, there is more uncertainty concerning the purity of the liquid, the nature of the carriers produced, and the current magnitudes employed.

3.6.3 High field injection

It is possible to generate carriers of either sign by what is known as high field injection. It was at first thought that the negative carriers were caused by the field emission of electrons due to the intense field at the tip of the injection system (razor blades are often used). However Hughes (1970) has shown that such currents are dependent on a water concentration of greater than 25 p.p.m. Presumably, carriers are produced by the disassociation of water molecules. Thus it is possible to generate currents in excess of 10^{-8} A in rather impure liquids. Because of the uncertainty over the nature of the carriers, which it seems likely will be different from other injection methods this method will not be considered further.

3.6.4 U.V. photo injection

This method was first used by Le Blanc (1959) in mobility measurements. It was further investigated by Morant (1960) and by Swan (1961) who suggested the method of back illumination of the photocathode.

The work of Minday et al (1971) has shown that the photo current increases with the amount of degassing and purification of the liquid. Bloor (1970) found that a fivefold improvement occurred between an air saturated liquid and the same liquid degassed to 10^{-6} torr.

An advantage of this technique is that it does not degrade the liquid (a problem with X rays) but there is a problem of the possibility of bulk generation of carriers. To reduce this, it is necessary to purify the liquid (usually by silicon gel) and to filter the U.V. to remove the lower wavelengths (Pugh 1968). Since the current magnitudes

are low (the order of pA), and the liquid has to be reasonably pure, this is a very suitable method for making comparisons between the current transients produced by different experimentors.

3.7 LIQUID MOTION STUDIES ON NON POLAR LIQUIDS

The work discussed in section 3.4 suggested that liquid motion only occurred at near S.C.L. currents. However, several authors have argued that most experimentally determined mobilities have been enhanced by liquid motion and that the true value lies around $2.5 \times 10^{-8} \text{ m}^2/\text{V}\cdot\text{sec}$. Certainly this would be a very nice explanation for the spread of results.

It has been known for some time that liquid motion can be a consequence of charge injection, and it would be reasonable to expect it to be influenced by the test cell geometry. The crucial point is the level of current at which motion occurs. Most of the evidence for liquid motion has come indirectly from variation in apparent mobility with current. An example of this is fig. 3.8, from Gray and Lewis (1969) showing how the apparent mobility with the current and also the measuring technique. It is impossible to derive a consistent value for the mobility unless the method is used of extrapolating the curve back to zero current.

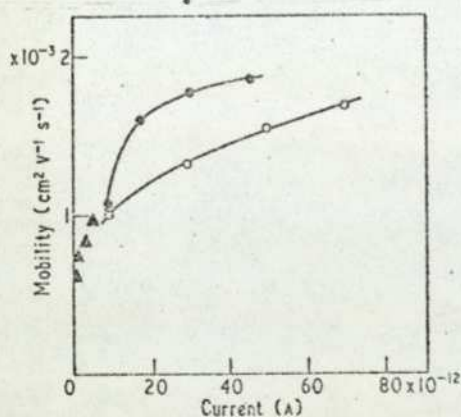


Fig. 3.8 (after Gray and Lewis)

In complete contrast to this, several workers have made no mention of such effects. Buttle (1969) tried to solve this problem by making mobility measurements over a wide range of input power. A graph of his results is shown in fig. 3.9 and indicates an independence of mobility with input power.

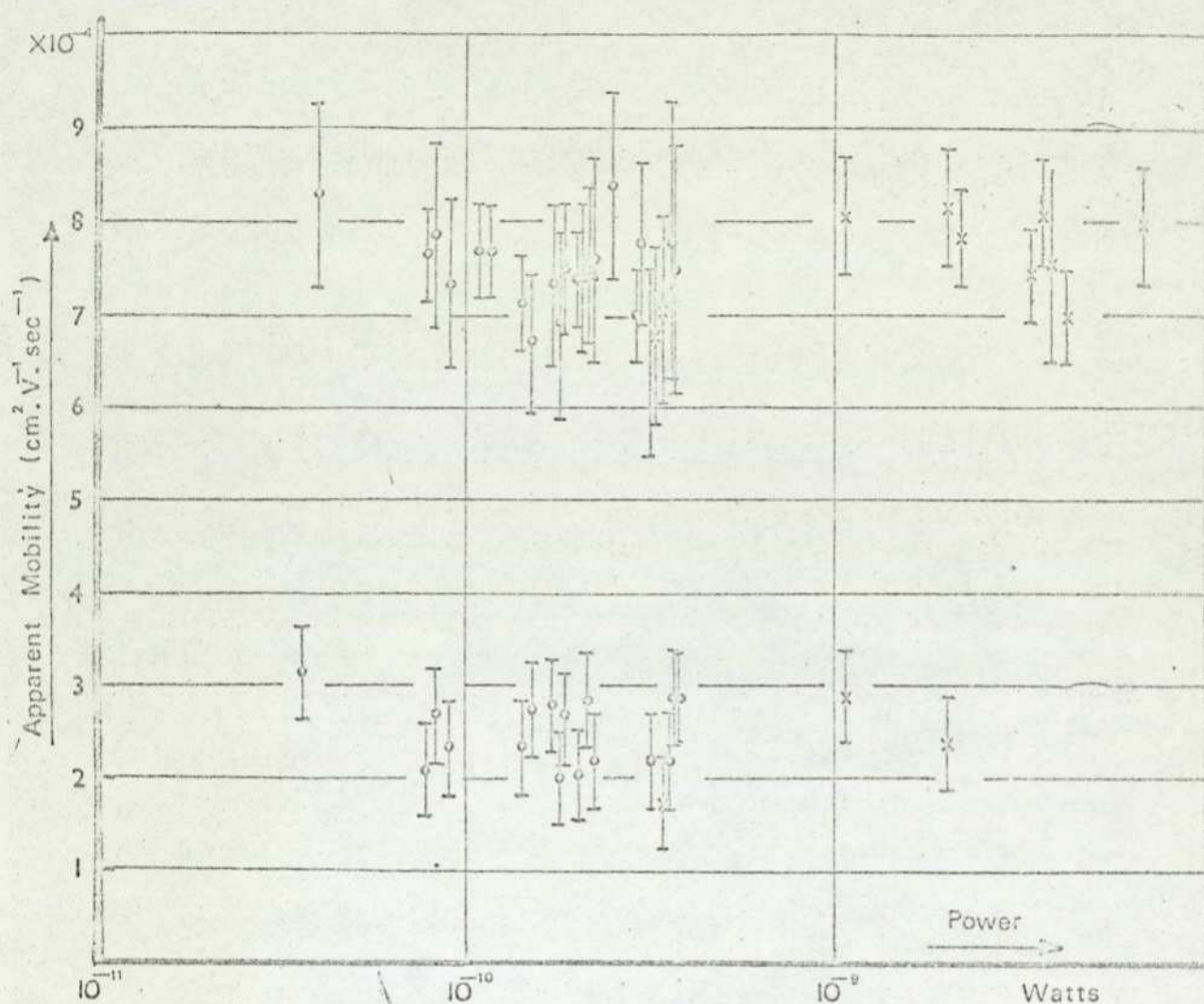
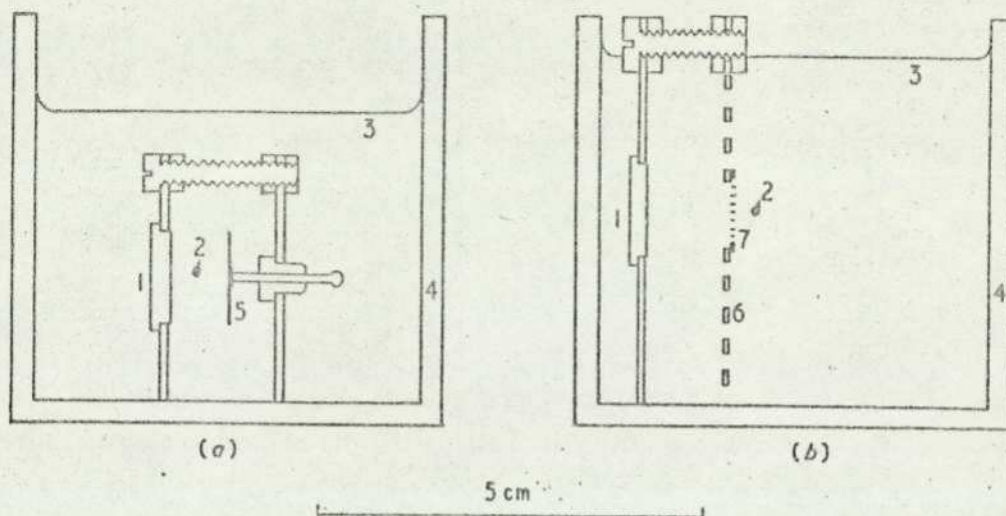


Fig. 3.9 Apparent mobility against power (after Buttle)

However his measurements sometimes indicated two distinct mobilities of 7.5 and $2.5 \times 10^{-8} \text{ m}^2/\text{V}.\text{sec.}$ with the latter only occasionally apparent. This question of current transient interpretation might be a key to this problem and Buttle's results will be discussed further in section 3.8.

Concrete evidence on liquid motion is given in the observations reported by Gray and Lewis (1965). They used a beta particle source mounted in one electrode, which produced a current of about 60pA

for a field of 1KV/cm. The two different electrode systems are shown in fig. 3.10.



Test cell configurations. 1, source; 2, dye blob; 3, hexane level; 4, glass enclosure; 5, collector; 6, collector-grid electrode; 7, special grid insert to provide minimum obstruction to flow.

Fig. 3.10 Grays test cells

The liquid motion was detected by introducing a blob of dye between the electrodes. With no source present, the dye did not move or spread out on application of the field. With the source present, the dye moved and its motion was recorded by film. The use of the second electrode system, with a grid electrode, eliminated the possibility that dye molecules were attached to the ions because when the dye was introduced outside the gap it still moved.

Essex and Secker (1969) and Taylor (1972) have made measurements indicating liquid motion and have claimed to have eliminated the effect to arrive at a mobility figure of about $2 \times 10^{-8} \text{ m}^2/\text{V}\cdot\text{sec}$. But there are some disquietening features to their work. They both used razor blade type injection to produce currents in excess of 10,000pA and used an extrapolation technique (similar to Barret and Gaspard's). Fig. 3.11 shows the method as applied by Essex and Secker; but instead of using the current as a base, the obscure injection diode potential is used.

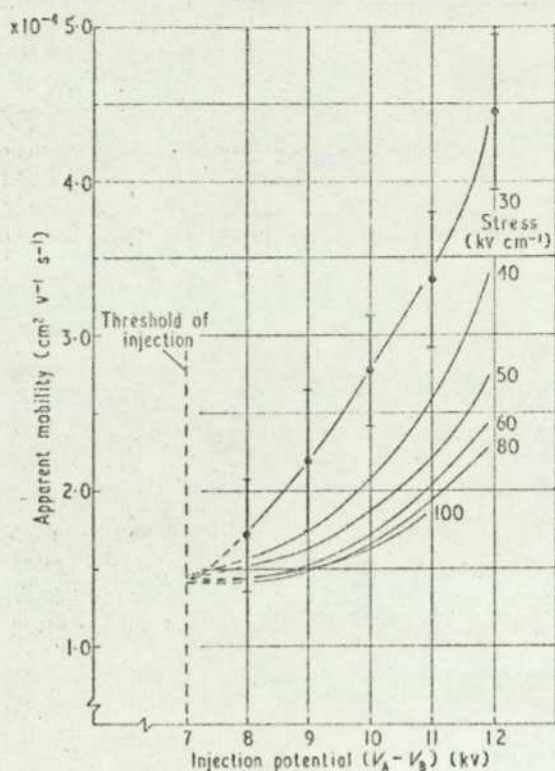


Fig. 3.11 Apparent mobility against injection potential (after Essex and Secker).

Now although this extrapolation technique worked correctly for Barret and Gaspard, it should be remembered that these authors' measurements were under steady state conditions which should produce a lower enhancement of the apparent mobility than the transient method.

Essex measured the mobility of a number of hydrocarbons by his method with the striking result that he obtained the same mobility result. He also performed measurements at different temperatures but again found no effect on the mobility.

It would seem that the justification of the method is that it produces curves from which it appears possible to eliminate liquid motion effects. A study of Taylor's thesis reveals that the transit times, from which he derives his mobility results, are very dependent on the current level, and are quite independent of stress. Fig. 3.12 shows a curve of transit time against current. The symbols

indicate the various field strengths. Clearly more work is needed to verify the results of this method.

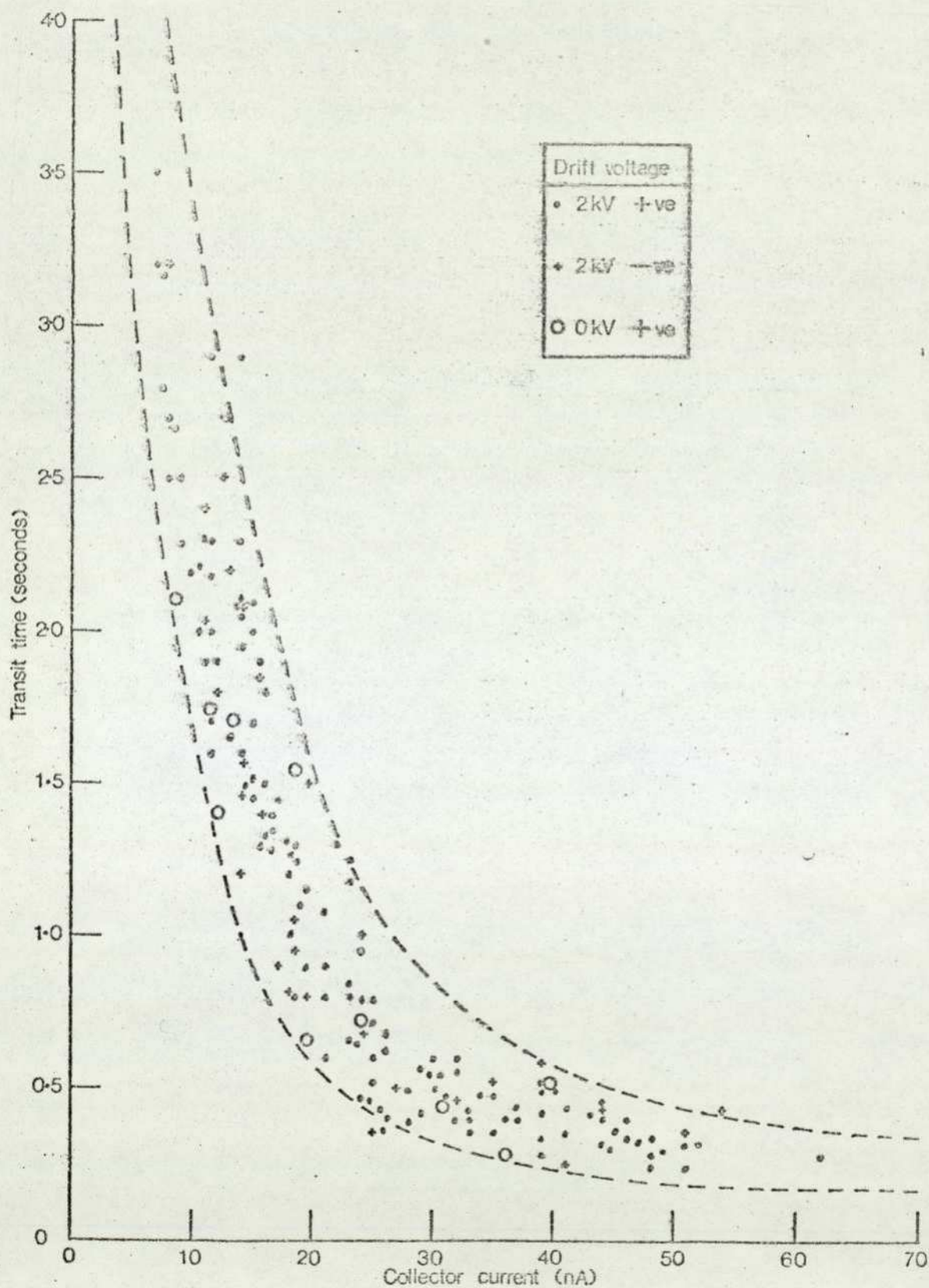


Fig. 3.12 Variation of ion transit time with collector current for various drift gap voltages (after Taylor).

3.8 U.V. MOBILITY STUDIES AND CURRENT TRANSIENT ANALYSIS

It has been shown that experimental mobility experiments have not been altogether helpful. Therefore, this section will be devoted to a study of the current transients obtained, so see if further insight can be gained from them. The emphasis will be on transients obtained by photoinjection in preference to those by X rays and high field injection because the former method implies purer liquids, lower currents, a well-defined layer of charge and less uncertainty over the nature of the carriers.

Le Blanc (1959) used well-purified and degassed hexane and filtered the U.V., thus minimising the effects of bulk ionisation. Two test cells were employed, having different methods of photocathode illumination. The first, in which most of the measurements were carried out, employed illumination from the side at an oblique angle, this is illustrated in fig. 3.13.

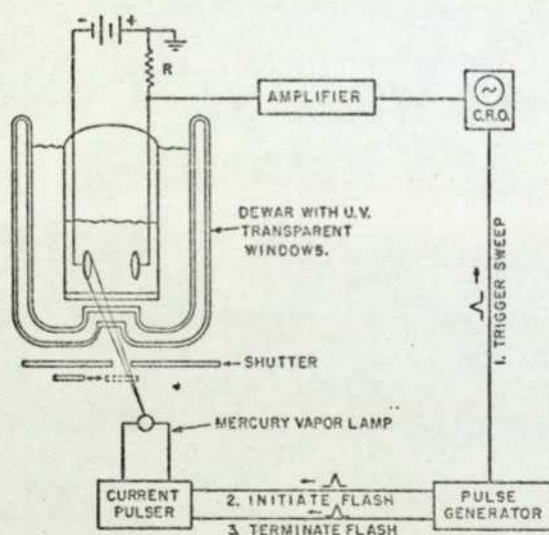


Fig. 3.13 Le Blanc's apparatus

In the second cell, the photocathode was illuminated through several small holes in the adjacent anode. Fig. 3.14 shows the 50mS

light pulse and the resulting current transient.

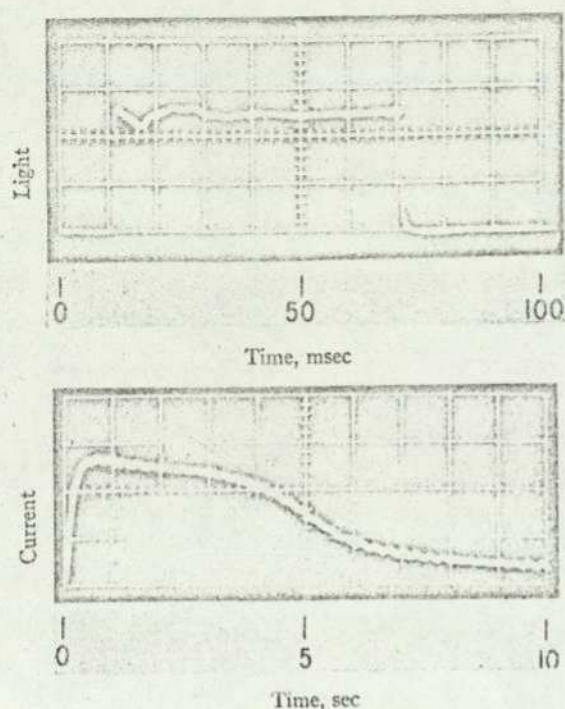


Fig. 3.14 Light pulse and resulting current transient (after Le Blanc)

This is far from the ideal rectangular shape and has been ascribed to the effects of diffusion, liquid motion, and carriers generated away from the centre axis of the electrodes and hence moving in non uniform fields.

Diffusion would produce a slope in the trailing edge of the curve but it is clear from the previous discussion in section 2.3 that diffusion would not have such a marked effect. The possibility of bulk ionisation causing this slope is also unlikely because of the purification employed and by the nearly horizontal top of the curve (c.f. section 2.2). The possibility of liquid motion is hard to assess, but it seems unlikely, for similar transients were obtained for both cells and it would be reasonable to expect such effects to be more

marked for the cell with numerous holes in the collector. A similar argument applies to possibility of carriers being produced near the edges of the electrodes and Le Blanc does state that he made some effort to collimate the U.V.

Having ruled out the factors of experimental technique, it would seem that the shape of the transient is a consequence of the liquid itself. Le Blanc states that he used several liquid samples over a period of months, so the transients cannot be attributed to a fresh sample. As no mention was made of how or if the transient altered with field strength or current level, no further conclusions can be drawn.

Chong and Inuishi (1960) performed measurements on a close gapped cell to determine if the mobility changed at high stress. They went up to 500 kV/cm but found no variation. The shape of the integrated current transient due to a $4\mu\text{s}$ light flash is shown in fig. 3.15.

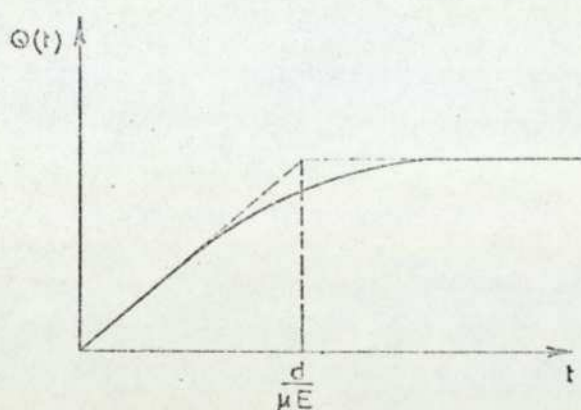


Fig. 3.15 Charge transient (after Chong and Inuishi).

It is hard to draw any conclusions from such curves, except to note that the curves show a deviation from the ideal ramp; the curvature being analogous to the sloping tail of Le Blanc.

Perhaps the best investigation of the transient shape has been provided by Bloor (1970). He was seeking a possible change of mobility, with the amount of liquid degassing, for fields up to 110KV/cm. He used conventional methods to purify his liquid and was able to degas it to 10^{-6} torr. He pulsed the U.V. light onto a deposited photocathode (using a flash tube), and measured the resulting transient by the integrating method. Fig. 3.16 shows the type of transient he obtained.

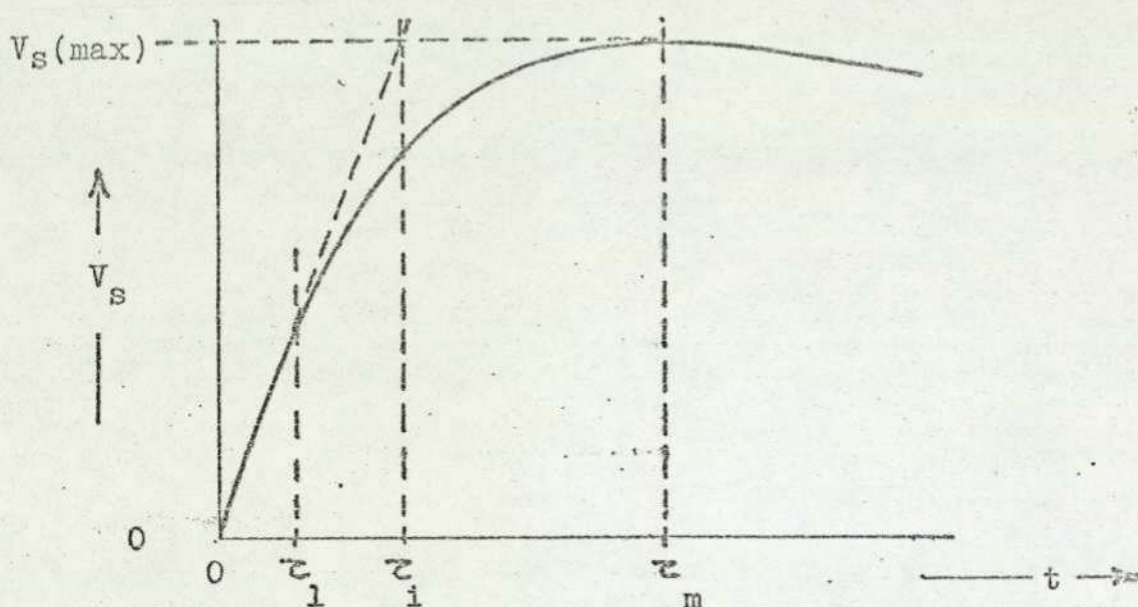


Fig. 3.16 Illustration of Bloor's method of transient analysis.

It reveals why Bloor was interested in the transient shape, for it is not easy to define the carrier transient time and hence the mobility, from such a curve. He considered three ways of estimating the transit time and these are illustrated on the curve as z_l , z_i , and z_m . z_i is found from the intersection of a line drawn through the linear initial rise of the curve and a line through the maximum of the curve ($V_s \text{ max.}$). z_l is derived from the point of departure from linearity. z_m is the time at which the current has fallen to zero, i.e. when the charge stops increasing.

In fact four test cells were used, designated A, B, C and D. The reason for this was that improvements were made to improve curve linearity (by superior photocathode illumination), to incorporate a variable gap and to enable measurements to be made at higher stresses. Fig. 3.17 illustrates the difference in curve shapes (using the same time scale) for cells A and B; showing B to be more linear.

Signal magnitude, V_s/V_{max} .

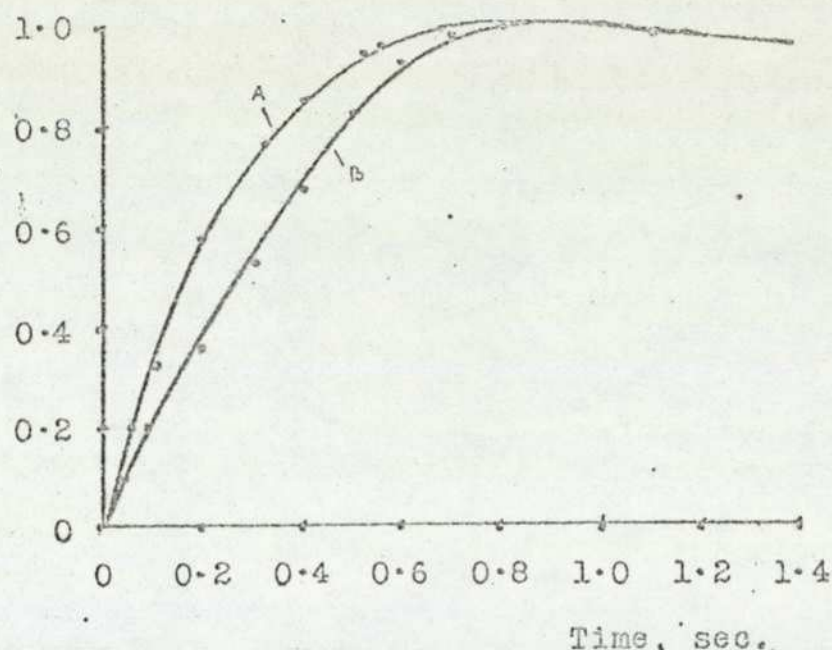


Fig. 3.17 Comparison of transients. Lower curve cell B; upper curve cell A. (after Bloor).

They both produce the Z_m transit time giving a mobility of

$$\mu_m = 3.4 \times 10^{-8} \text{ m}^2/\text{V}.\text{sec.}$$

Also Z_i was estimated for cell B giving a mobility of

$$Z_i = 11 \times 10^{-8} \text{ m}^2/\text{V}.\text{sec.}$$

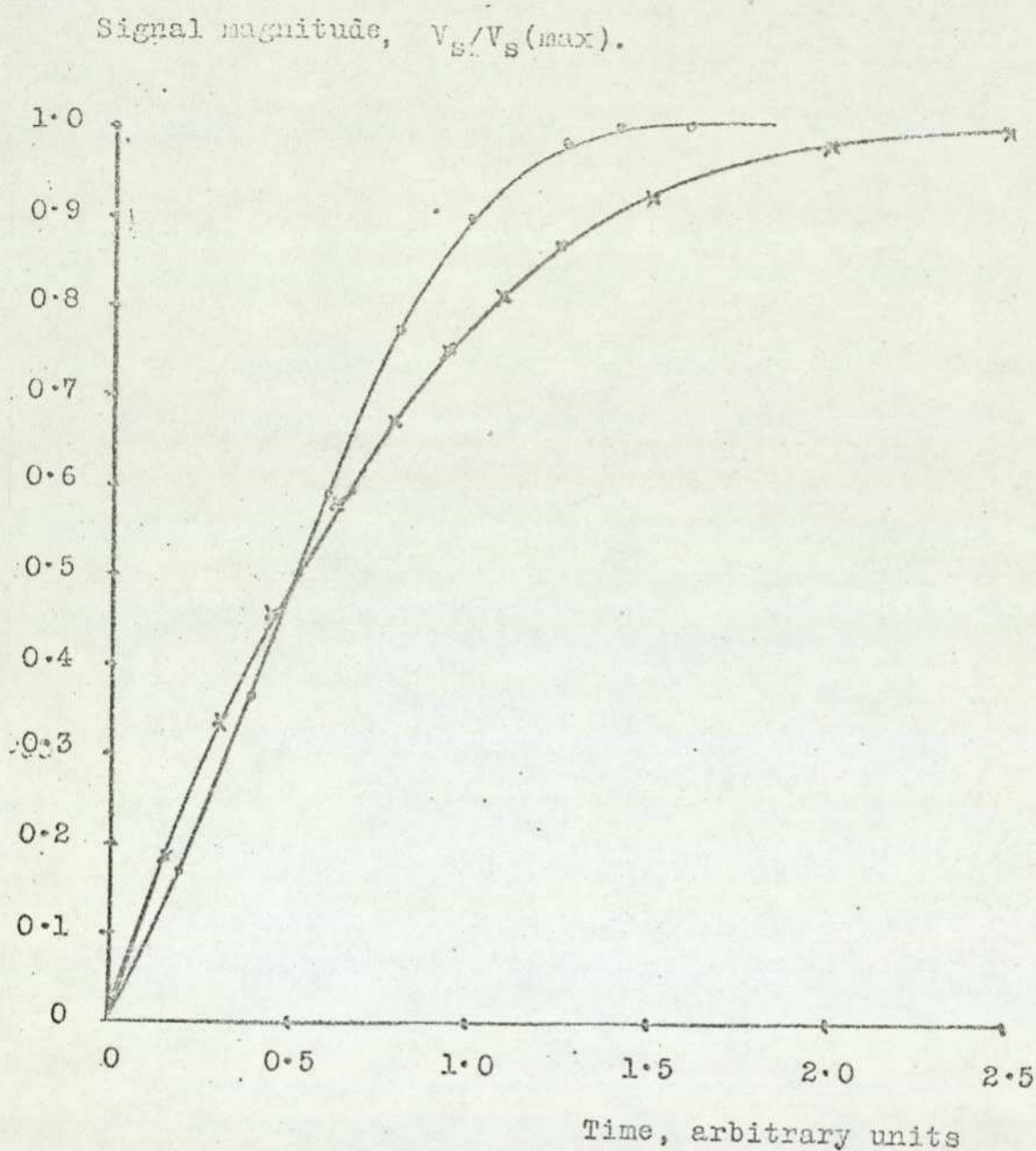
Fig. 3.18 shows a comparison of the transients in cells B and C.

It is important to note that the time scale is in terms of Z_i for each transient, hence it is a different scale for each because the cells gave different mobility figures. For cell C the mobilities were

$$\mu_e = 10 \times 10^{-8} \text{ m}^2/\text{V}.\text{sec.}$$

$$\mu_i = 7 \times 10^{-8} \text{ m}^2/\text{V}.\text{sec.}$$

$$\mu_m = 5 \times 10^{-8} \text{ m}^2/\text{V}.\text{sec.}$$



Comparison of transient shapes obtained
with cell 'B' (×) and cell 'C' (•).

Fig. 3.18 (after Bloor)

The reason for the improved linearity of cell C was that it's design, having back illumination of the photocathode, overcame the problems of illumination from an oblique angle which occurred in cell B.

Cells D1 and D2 had adjustable gaps and produced mobility figures independent of gap width and stress of

$$\begin{aligned}\mu_e &= 10 \times 10^{-8} \text{ m}^2/\text{V}\cdot\text{sec.} \\ \mu_i &= 7 \times 10^{-8} \text{ m}^2/\text{V}\cdot\text{sec.}\end{aligned}$$

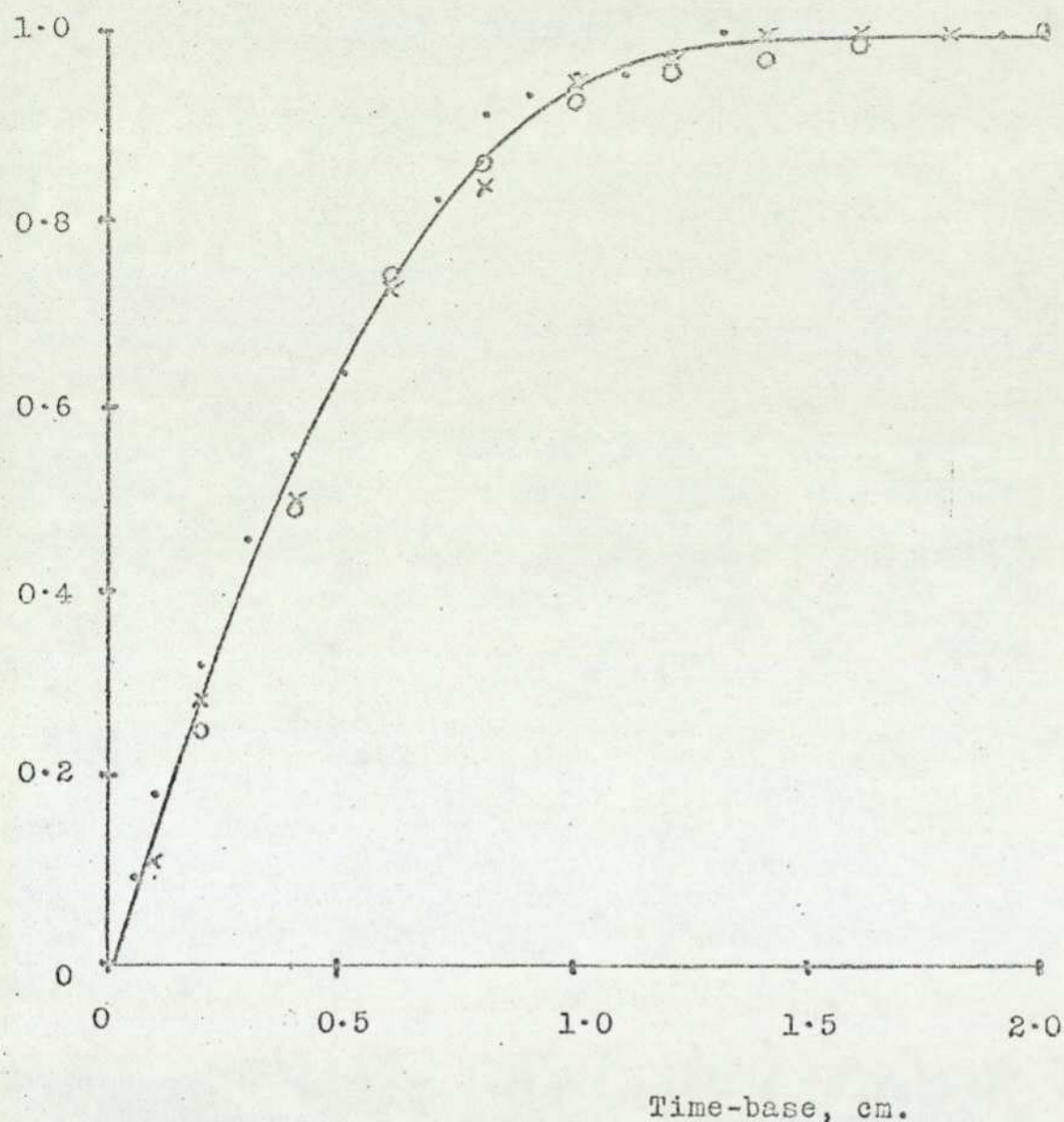
Fig. 3.19 shows the transients on a normalised time scale for gaps of 1 mm. to 4 mm. This shows that the gap distance has very little effect on the curve shape. Bloor also investigated the effect of masking the photocathode so as to only illuminate a central portion. Fig. 3.20 shows the result, of this for various mask apertures. Again there is no effect on the transient.

The effect of liquid degassing can be gauged from fig. 3.21. The amplitudes have been normalised because the effect of air in the liquid is to reduce the photocurrent by an order of magnitude. This is a significant result for an oxygen ion is widely presumed to be the charge carrier, but there is only a marginal change on the transient.

Overall, the curves depart significantly from the ideal ramp and Bloor sought to explain this. Bulk ionisation was ruled out by measurements using forward and reverse bias on the cells. Bloor considered the possibility of static space charge causing field non-linearity which would produce a distorted transient. This was ruled out because such an effect would be expected to vary with the level of stress, but this did not happen in practice. This will be considered further in section 3.9.

The effects of axial and radial diffusion were considered and

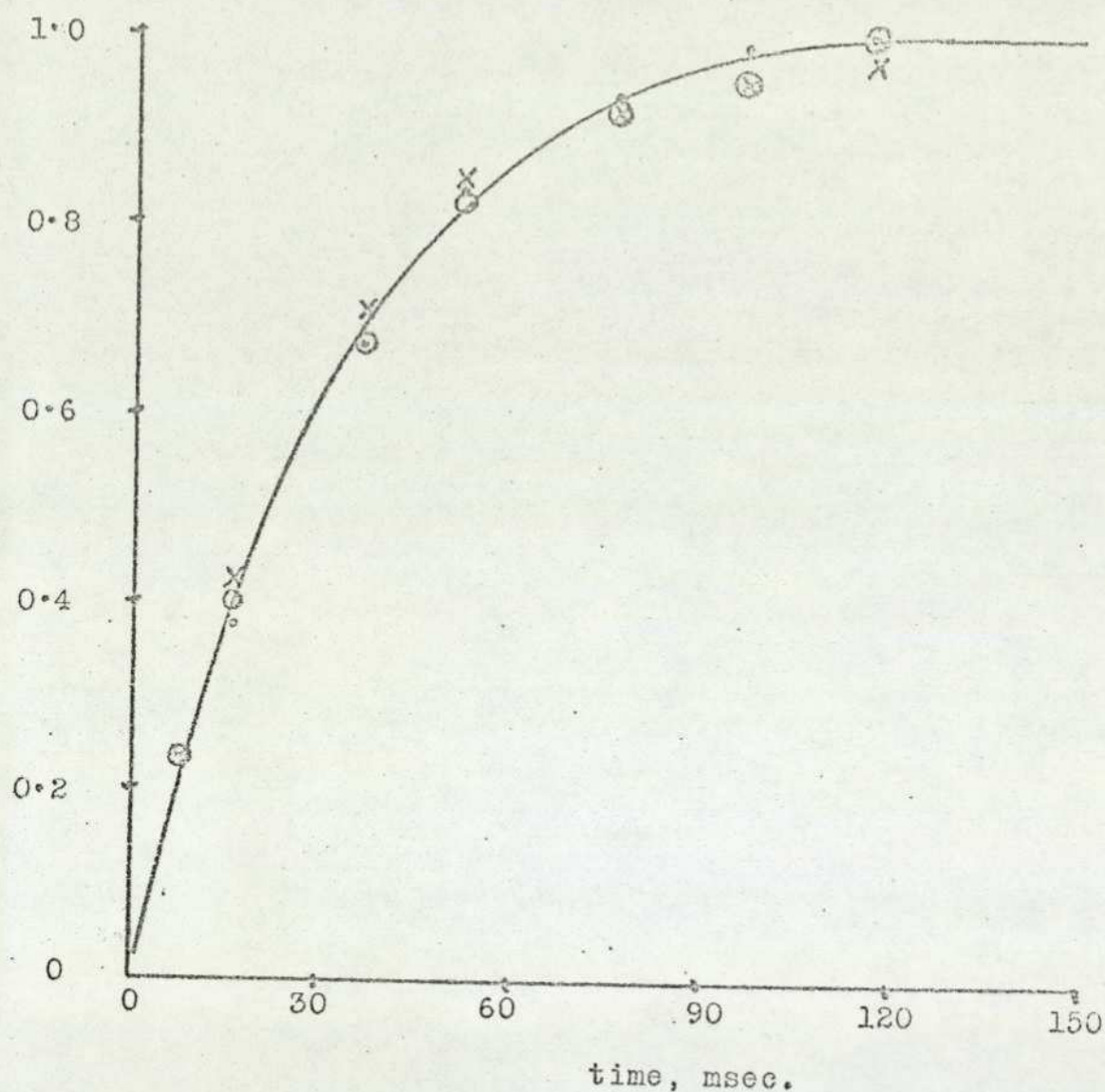
Signal voltage (normalized)



Integrated current-transients for an applied field of 10 KV/cm. Nominal electrode separations: 4 mm. (•); 2 mm. (×); and 1 mm. (○). Time-base scales: 40 msec/cm.; 20 msec/cm.; and 10 msec/cm. respectively.

Fig. 3.19 (after Bloor)

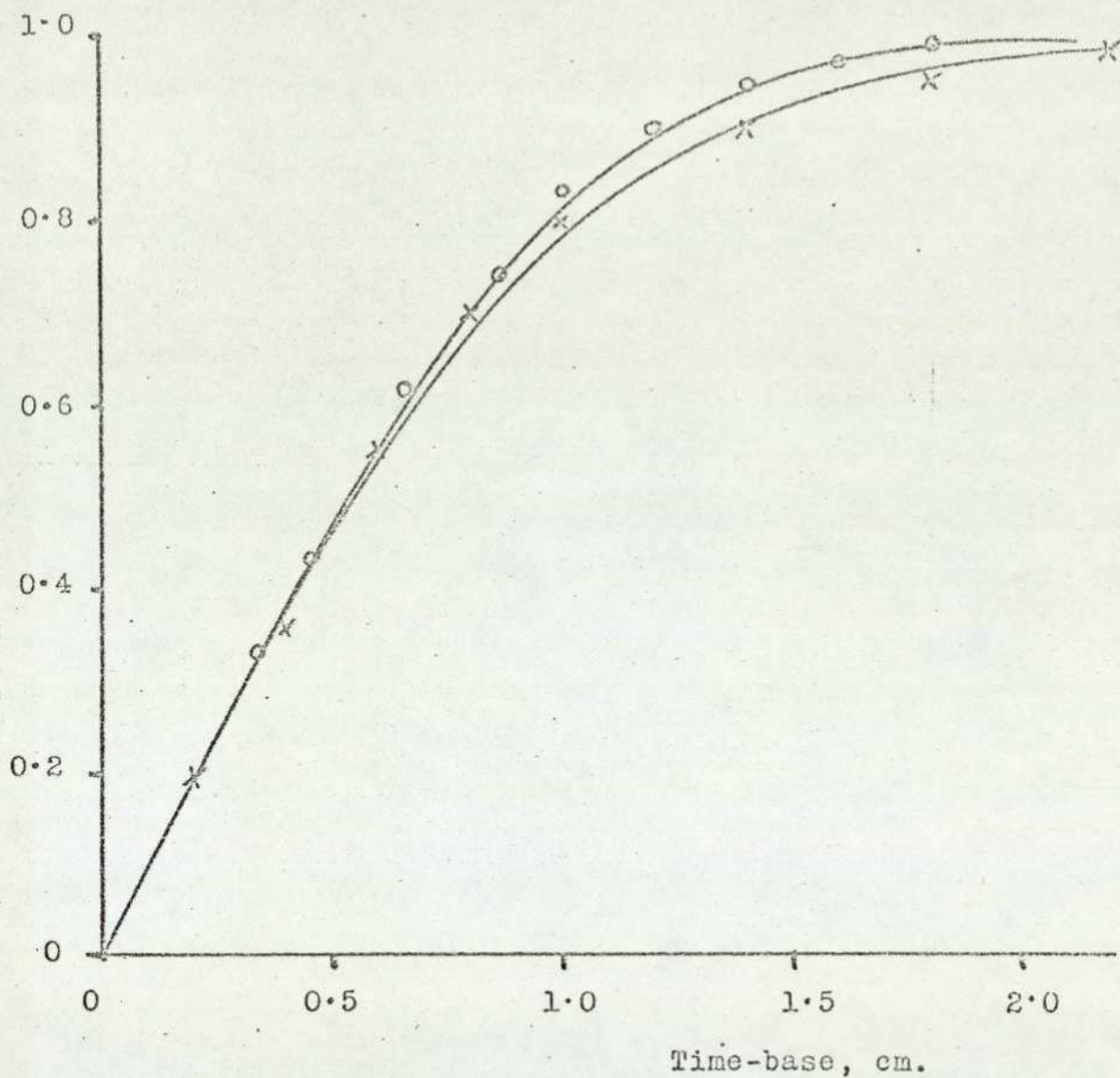
Signal magnitude (normalized)



Effect of mask aperture diameter on the shape of the integrated transient. Aperture diameters : 8 mm (•); 4 mm (X); and 2 mm (O).

Fig. 3.20 (after Bloor)

Signal voltage, $V_S/V_S(\text{max})$.



Transients observed using a) freshly degassed hexane ($\circ$) and b) the same sample in equilibrium with an air pressure of 200 torr ($\times$).

Fig. 3.21 (after Bloor)

were shown to be insignificant. Bloor finally concluded that liquid motion was responsible and used a qualitative model to argue this. This experimental technique prevented him from seeking to establish a relationship between curve shape and current level which would have been helpful in evaluating this conclusion. However fig. 3.22

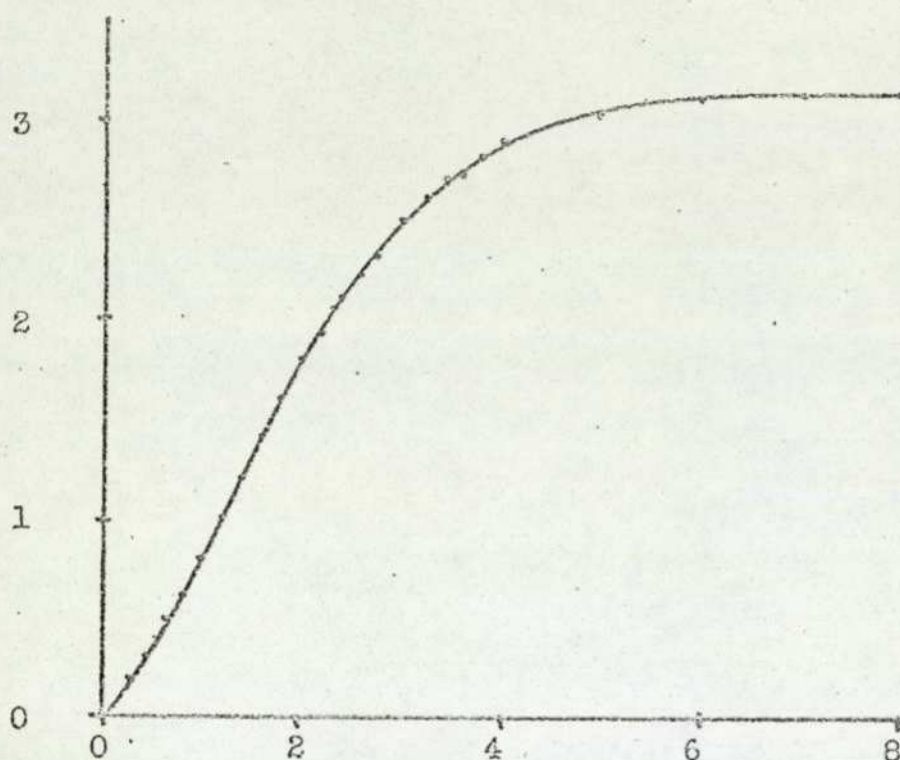


Fig. 3.22 (after Bloor)

shows a transient obtained with an extra sensitive photocathode, which Bloor used to support his hypothesis. It shows an initial concave curvature indicating an increase in current magnitude, and hence carrier velocity, during their transit.

Such an explanation is not so easy to accept when it has shown that gap distance produced little effect. Interestingly, when Bloor considered the implication of such motion on his mobility results, he decided that Z_l indicated the mobility of the carriers. It would

seem more reasonable to consider Z_{ℓ} to be due the sum of liquid and ion motion, and hence Z_m to be more representative of ion mobility.

Additional information is provided by Rahman (1970) who used the test cell shown in fig. 3.23 but used the same apparatus as Bloor.

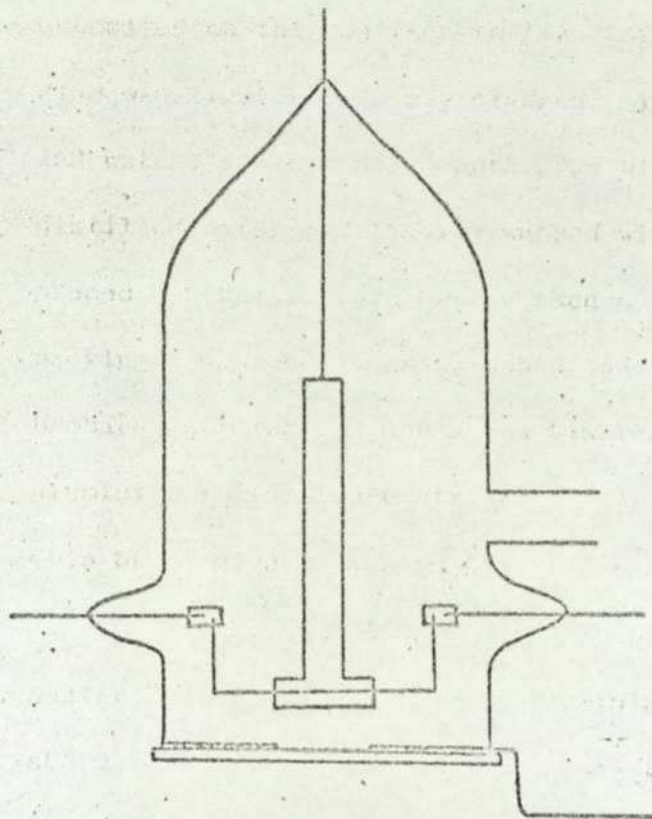


Fig. 3.23 Rahman's test cell

A photocathode was deposited onto a quartz disc (which formed the base of the cell), and this was illuminated through the disc from underneath. It was possible to obtain transients for either polarity of applied voltage, by photo injection from the deposited film or from the solid aluminium electrode. The deposited film produced a two or three times greater current and the transients are shown in fig. 3.24. The deposited film transients (curve A) is more linear but exhibits slight concave curvature. Rahman used the Z_{ℓ} method of transit time estimation to arrive at mobilities of

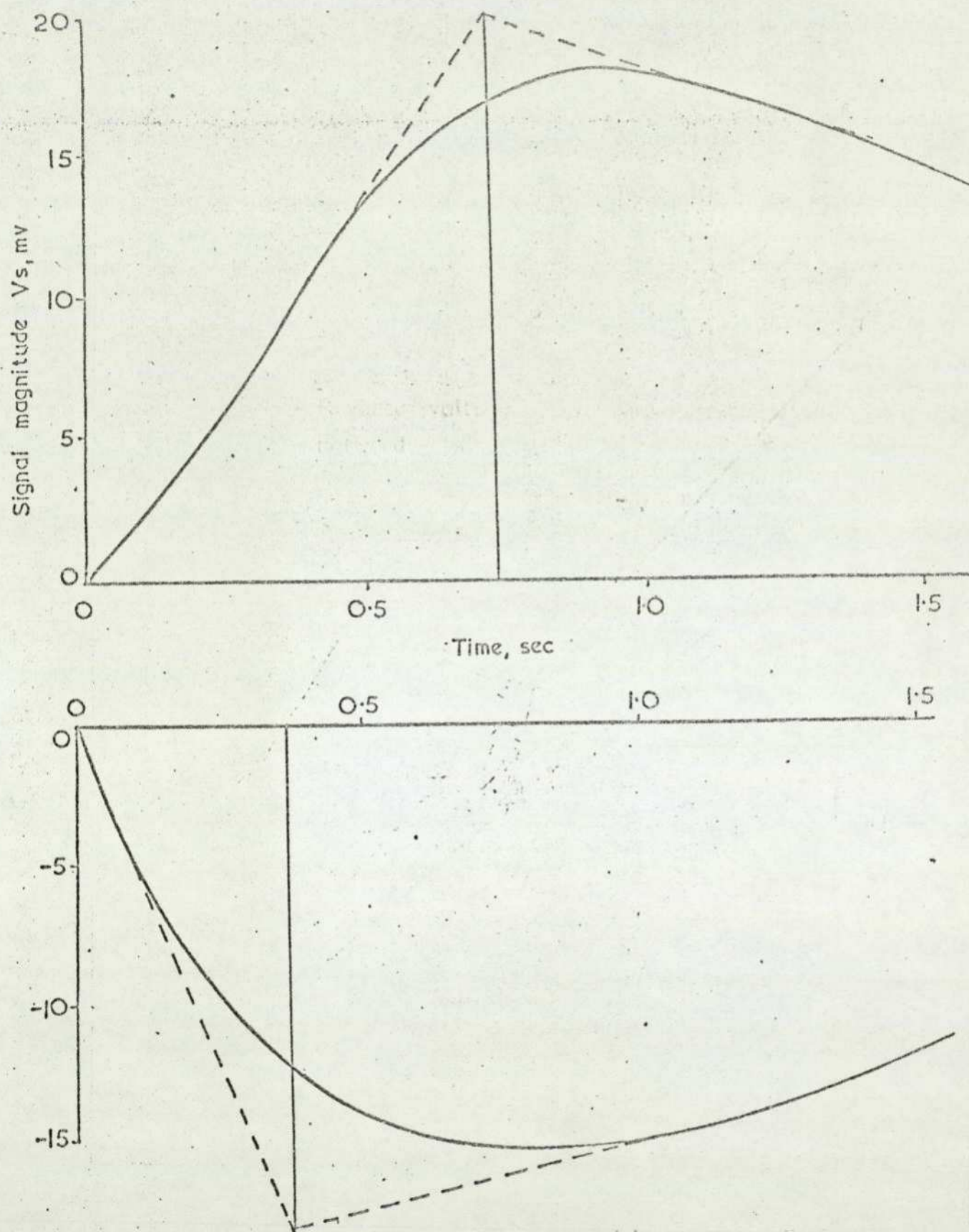


Fig. 3.24 (after Rahman)

$$6.3 \times 10^{-8} \text{ m}^2/\text{V}\cdot\text{sec.}$$

$$8.2 \times 10^{-8} \text{ m}^2/\text{V}\cdot\text{sec.}$$

for the film and solid electrode respectively. Both of these mobilities was shown to be independent of field strength and filtering the U.V. also had no effect.

Now Rahman was not principally interested in the mobilities of the carriers and did not pursue this interesting anomaly. It is unfortunate that both Rahman and Bloor used the integrating method as this tends to obscure information which can be gained from transients.

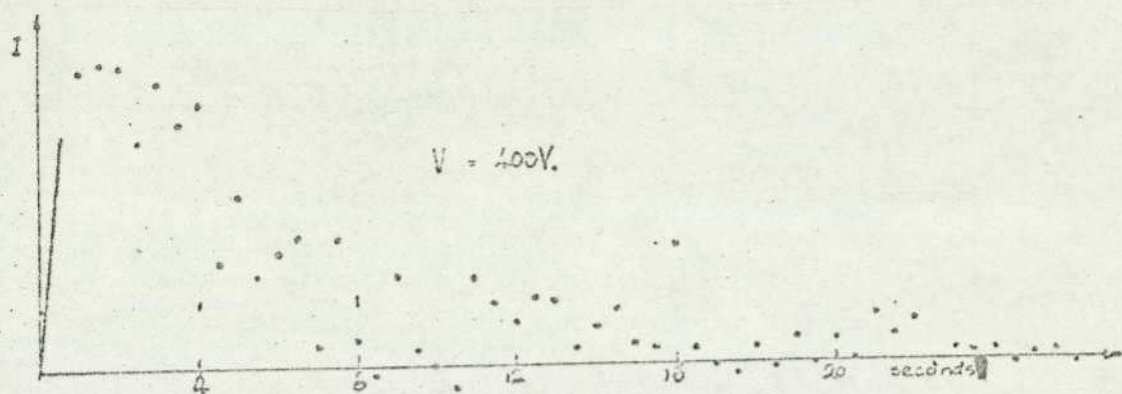
Buttle (1969) overcame the problem of noise by the use of a digital correlation technique to obtain the impulse response of the system. This method is essentially an averaging of several transients. Fig. 3.25 shows the impulse transients and fig. 3.26 gives the integrals of these curves. The current transients are given by ~~bars~~^{dots} and not a line, to indicate the sampling employed (which tends to mask the noise in the waveform). This bears a resemblance to the transient obtained by Le Blanc (fig. 3.14), and the charge transient resembles those of Bloor and Rahman.

Buttle explained the tail on his transients in terms of diffusion, and the effects of charge moving in the non uniform field around the peripheries of the electrodes. However, he also argued that the charge transients have two slopes and that these correspond to two distinct charge carriers with mobilities of

$$7.5 \times 10^{-8} \text{ m}^2/\text{V}\cdot\text{sec.}$$

$$\text{and } 2.5 \times 10^{-8} \text{ m}^2/\text{V}\cdot\text{sec.}$$

He stated that the slower carrier was less easily discernible than the faster carrier and it was sometimes buried in noise. As this carrier was more apparent at lower voltages and currents, Buttle argued



(1)

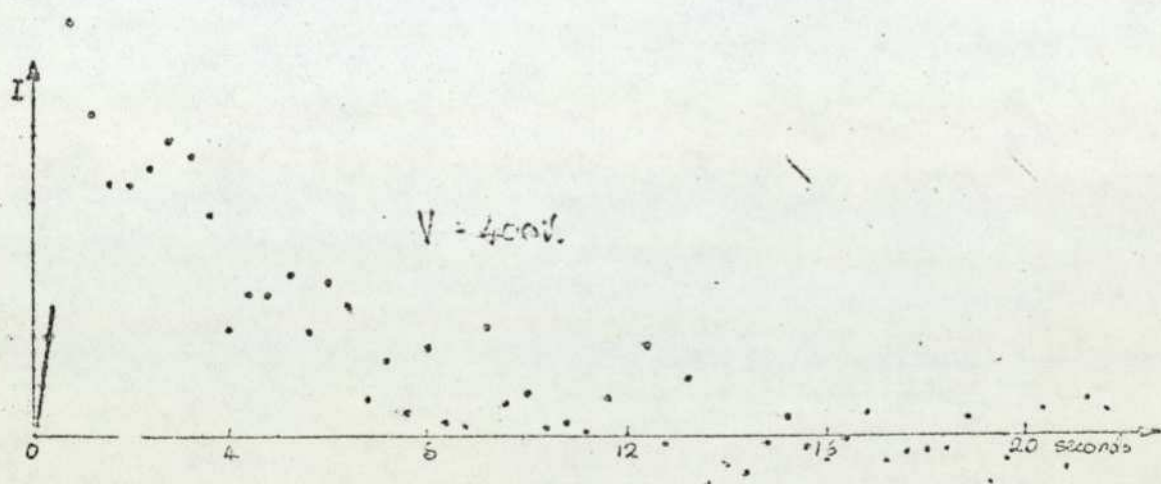
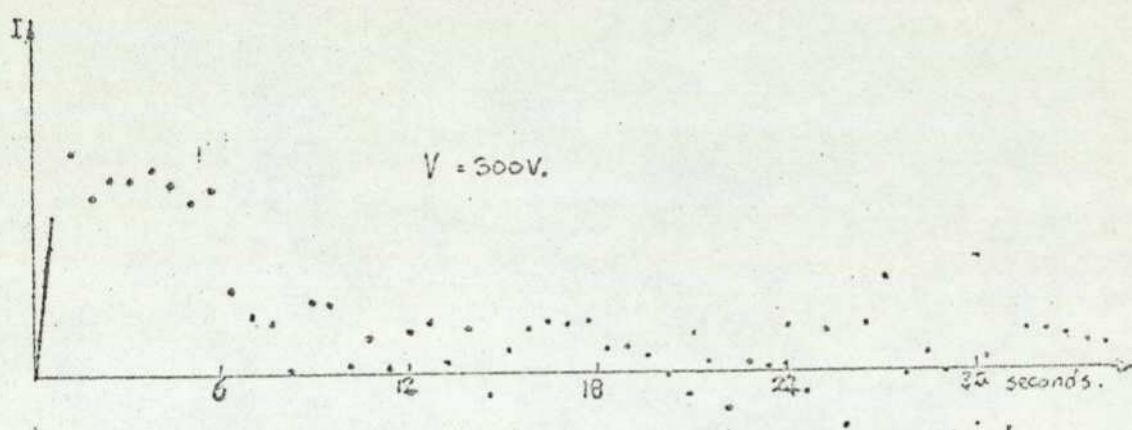


Fig. 3.25 (after Buttle)

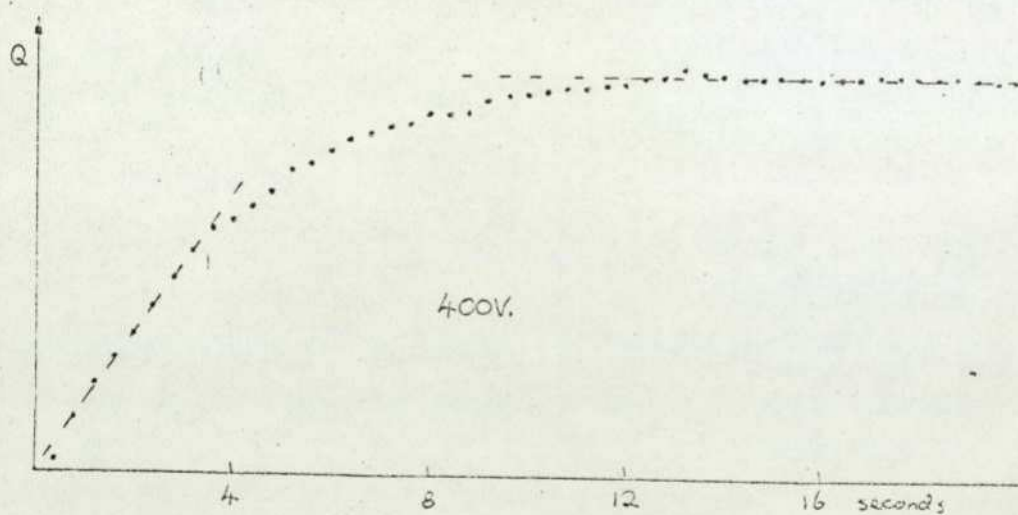
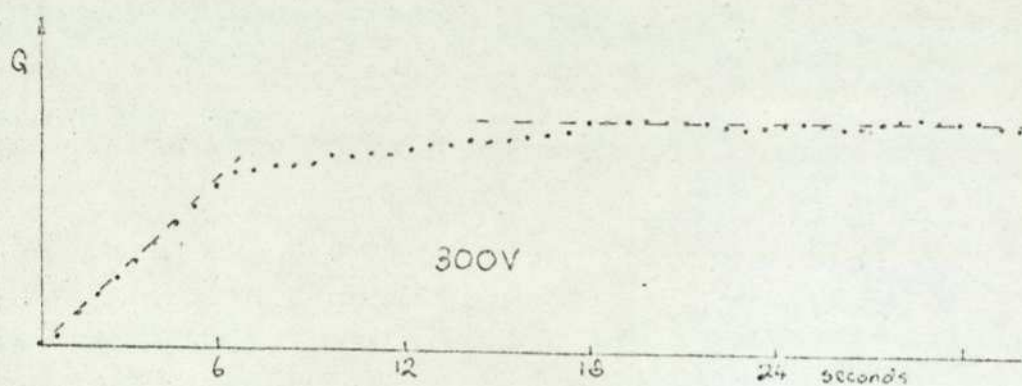
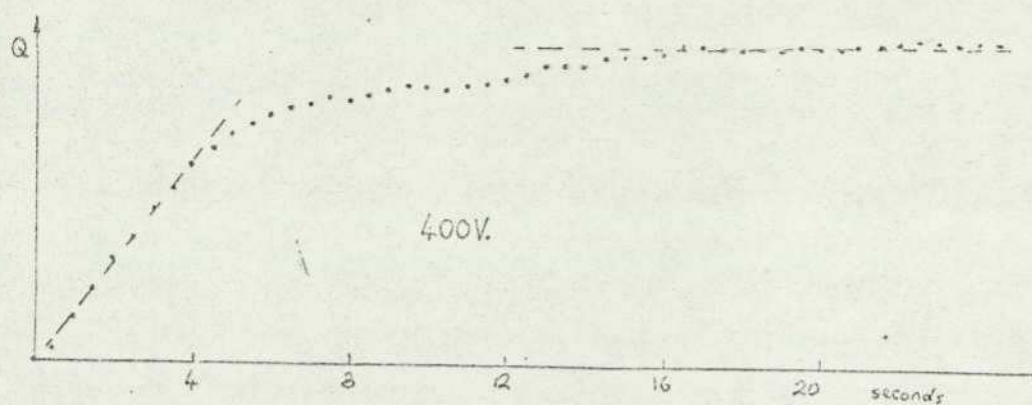


Fig. 3.26 Charge transients obtained by integrating curves of fig. 3.25
(after Bloor)
Buttle

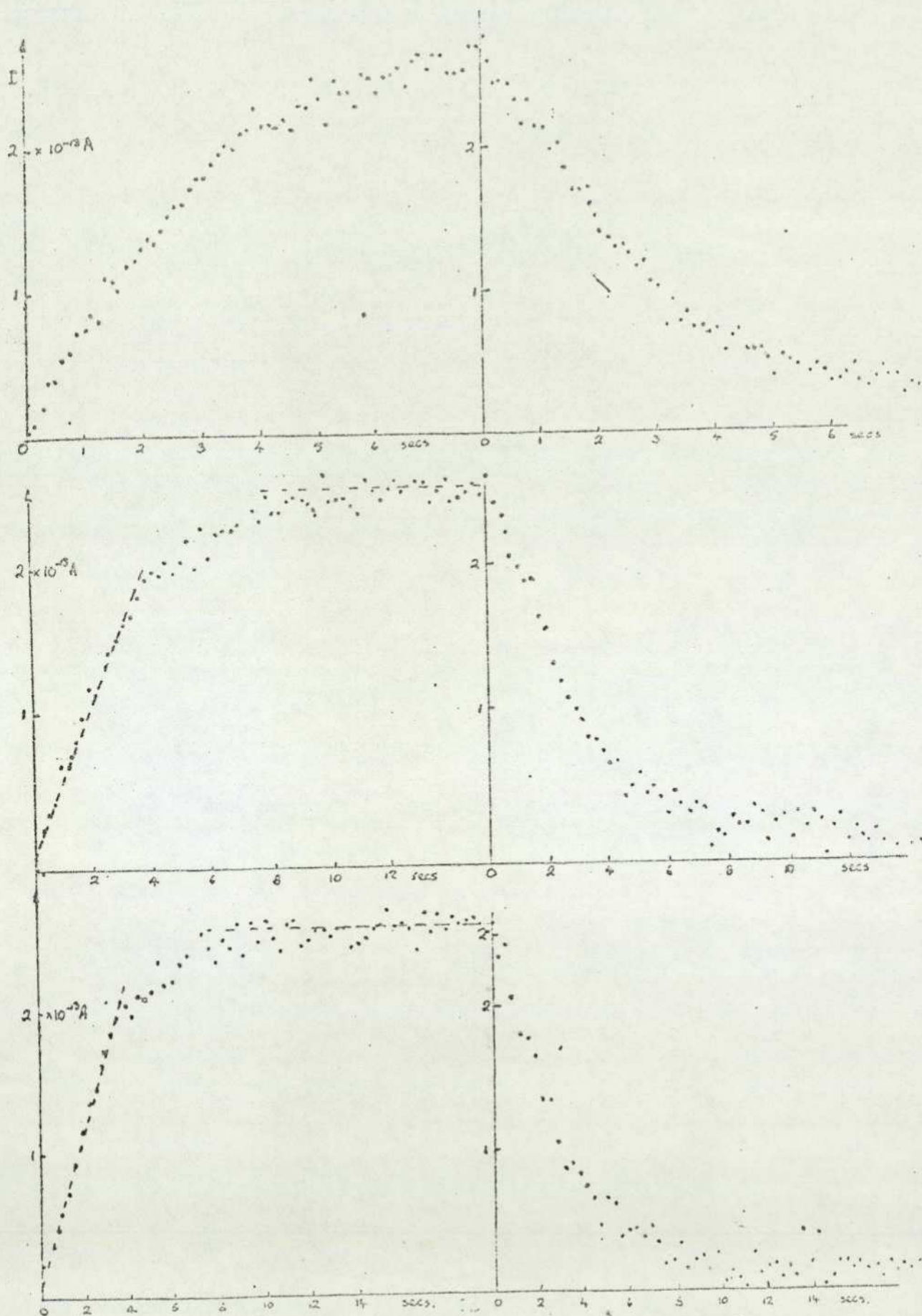


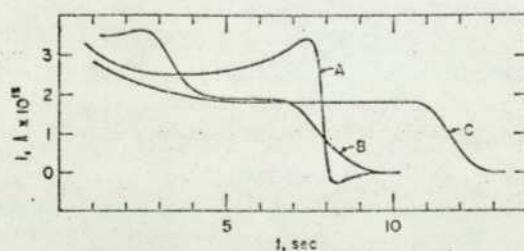
Fig. 3.27 (after Buttle)

that this was because the carriers were then spending a longer time in the liquid and at lower concentrations, thus increasing the chance of trapping occurring to form the slower carrier.

Buttle also measured the response of the system to a simple square wave excitation and the transients are shown in fig. 3.27. The applied voltage was 500V across a 1.2 cm. gap. In the absence of space charge effects, these curves should be the same shape as the charge transients in fig. 3.26.

Buttle performed measurements over a range of currents and voltages to establish if liquid motion was enhancing the mobility results and fig. 3.9 shows he found this not to be so. To obtain the larger input powers he made use of a vapour gap, (Buttle 1969), between the liquid and the photocathode, but this does not seem to have radically altered the transient's shape.

Although the experimental work of this thesis is confined to the generation of carriers by U.V. illumination, and this section has concentrated on current transients produced in this way, it should not be thought that unusual transients are absent with other methods. An illustration is provided by Allen and Hummel (1966) who used X rays to generate a layer of carriers in hexane. Fig. 3.28 shows the resulting transients.



Current-time curves obtained with hexane in the cell
Curve A, negative ions; B and C, positive ions.

Fig. 3.28 (after Allen and Hummel)

Curves A and C are for negative and positive ions formed near the electrodes. The initial surge is due to ions of opposite polarity moving to the nearby electrode. Curve B is for a layer formed near the centre of the gap and the first drop is due to the arrival of the faster negative ions. The curves for the negative ions show an anomalous peak, and curve A exhibits a small negative current. There are no obvious explanations for these effects; the currents used were too small for space charge effects to occur.

3.9 CURRENT TRANSIENTS AND STRESS DISTRIBUTIONS IN LIQUIDS

Brignell (1964) has proved that a non uniform stress distribution will increase the transit time of carriers across the gap, thus giving an apparently smaller mobility. The problem lies in measuring the distribution in a highly insulating liquid such as hexane.

In fact, an original intention of this research was to use the "charge probe" technique to map the internal field distributions of liquids. In section 3.8 it was mentioned that Bloor and Morant (1968) considered interpreting their distorted current transients in terms of non uniform fields. Although Bloor later discounted this concept it is interesting to see what field distribution could cause such a transient.

It has been noted in section 2.6 that Bloor and Morant used an erroneous method so this provides an opportunity to demonstrate the use of a computer programme (S4717) which gives the correct analysis. The sequence of computer graph outputs are shown in figs. 3.29 to 3.32.

The starting point is fig. 3.29, a current transient taken from Bloor's thesis. Fig. 3.30 shows the integral of this graph, which is the charge transient. Fig. 3.31 gives the relationship between the distance moved by the probe against time. Finally fig. 3.32 shows the stress distribution. This method is explained in section 2.6 and appendix C, and is verified in section 4.62.

In fact the final resulting stress distribution is not so different from that obtained by the erroneous method.

Reviews on stress distributions in liquids have been provided by Croituru (1965) and Pollard (1972). These go into the mechanism of the non-uniform stress and the methods of measurement.

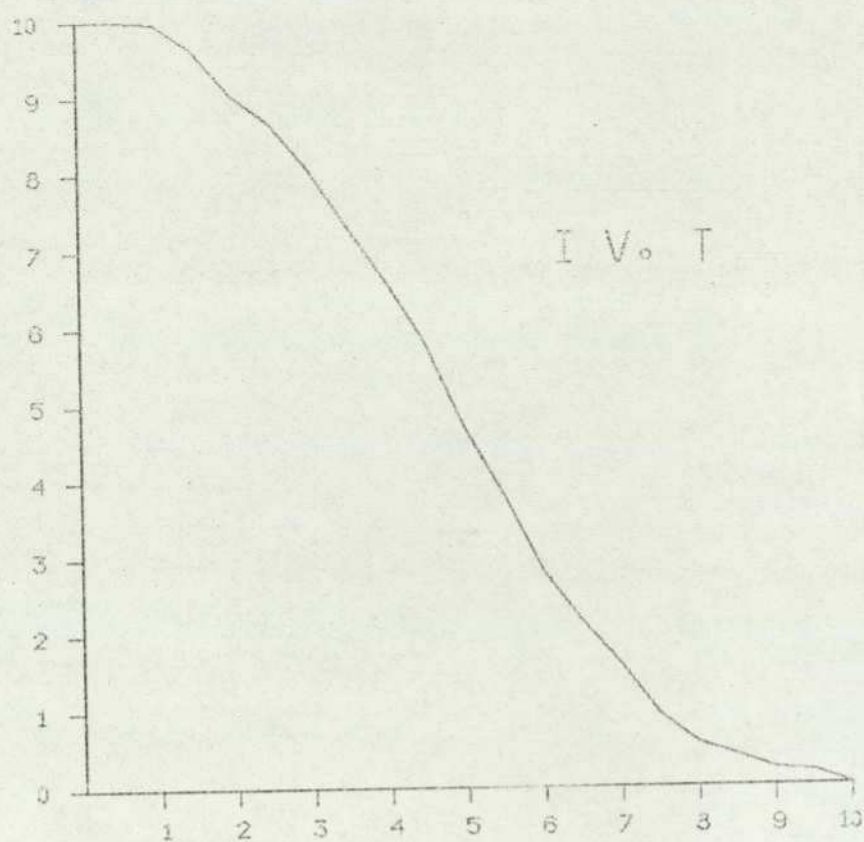


Fig. 3.29 Bloor's current transient

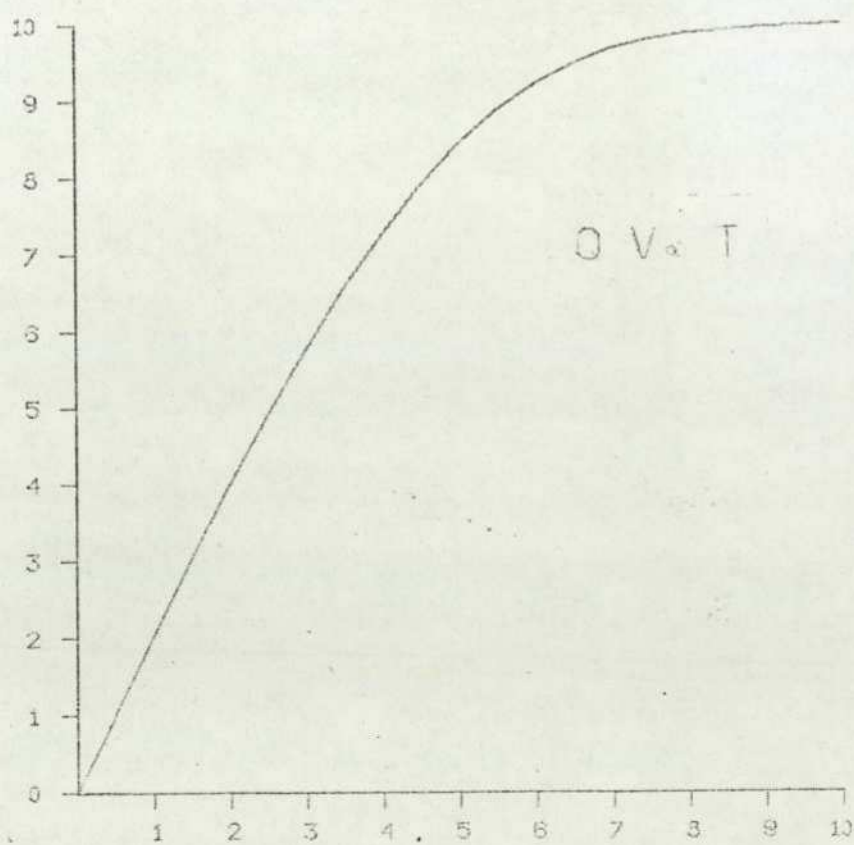


Fig. 3.30 Charge transient (integral of fig. 3.29)

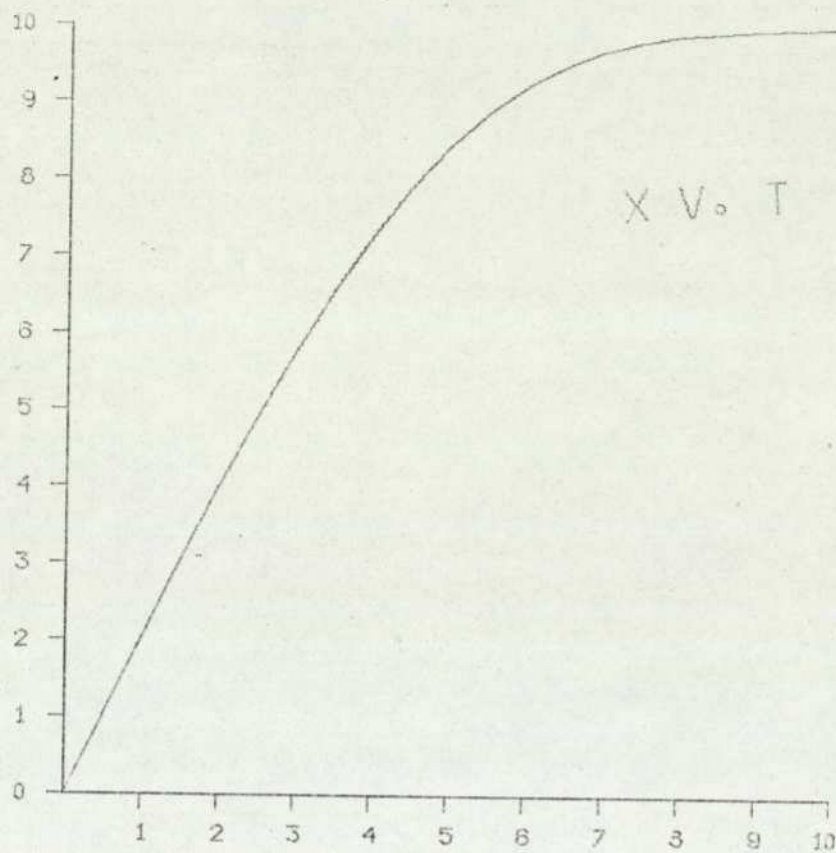


Fig. 3.31 Distance moved by probe against time.

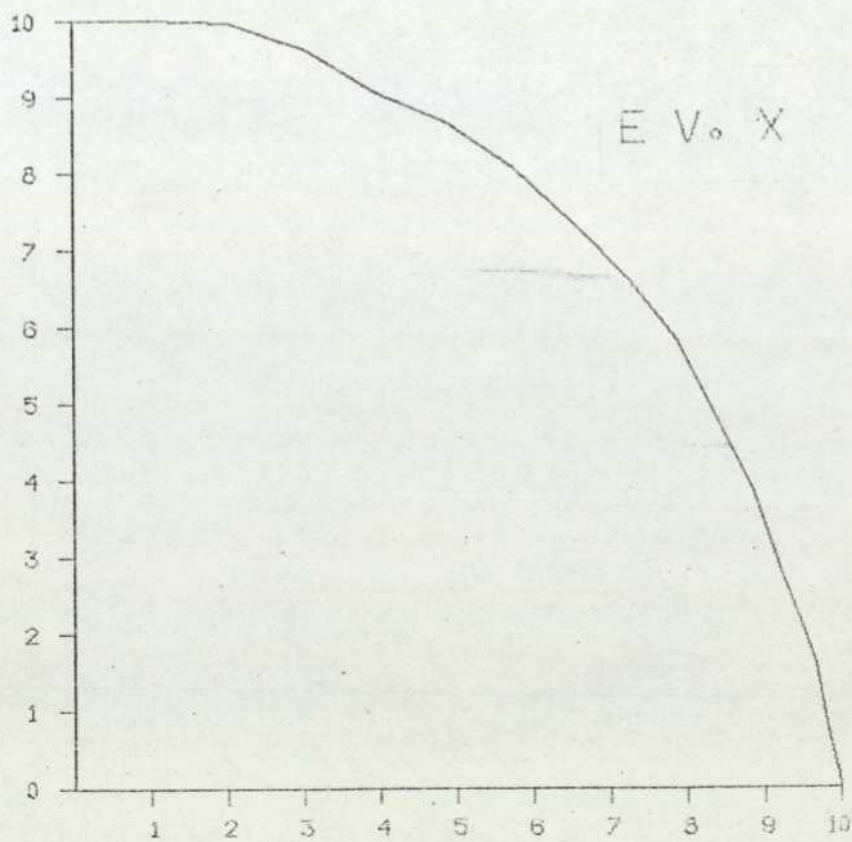
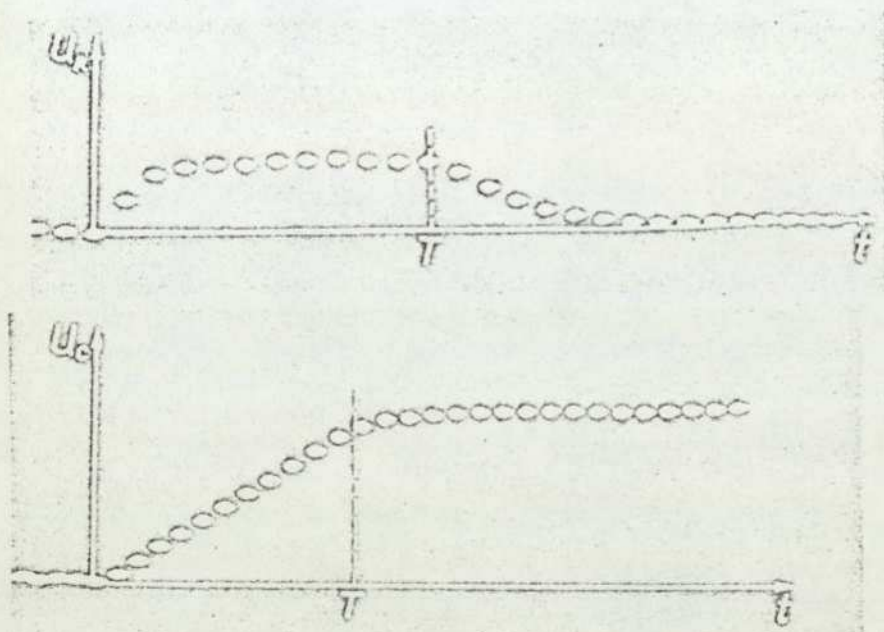


Fig. 3.32 Stress Distribution.

Bloor and Morant (1968) tried to apply the technique of measurement with a metal probe to hexane. They found that the extremely high resistivity ruled out such a method. Pollard (1970) constructed a sensitive ellipsometer to utilise the Kerr effect (which is very small in non polar liquids), and reported results for an applied field of the order of 50kv/cm. He found a certain amount of non-uniformity which seemed to vary markedly with the liquid purity. It is not easy to know what reliance to place on his results, as the method is still in an early stage of development.

Finally the work of Jackym et al (1971) should be considered. They analysed the field distributions in decane, cyclohexane and carbon tetrachloride using the erroneous method of 2.6. They used X rays to generate a thin layer of carriers in liquid, by applying the radiation for a period of time before switching on the field. Then they either measured the resulting current or charge transient. Fig. 3.33 shows such transients for negative carriers in $\text{Si}_3\text{O}_2(\text{CH}_3)_8$ at a field strength of 800v/cm.



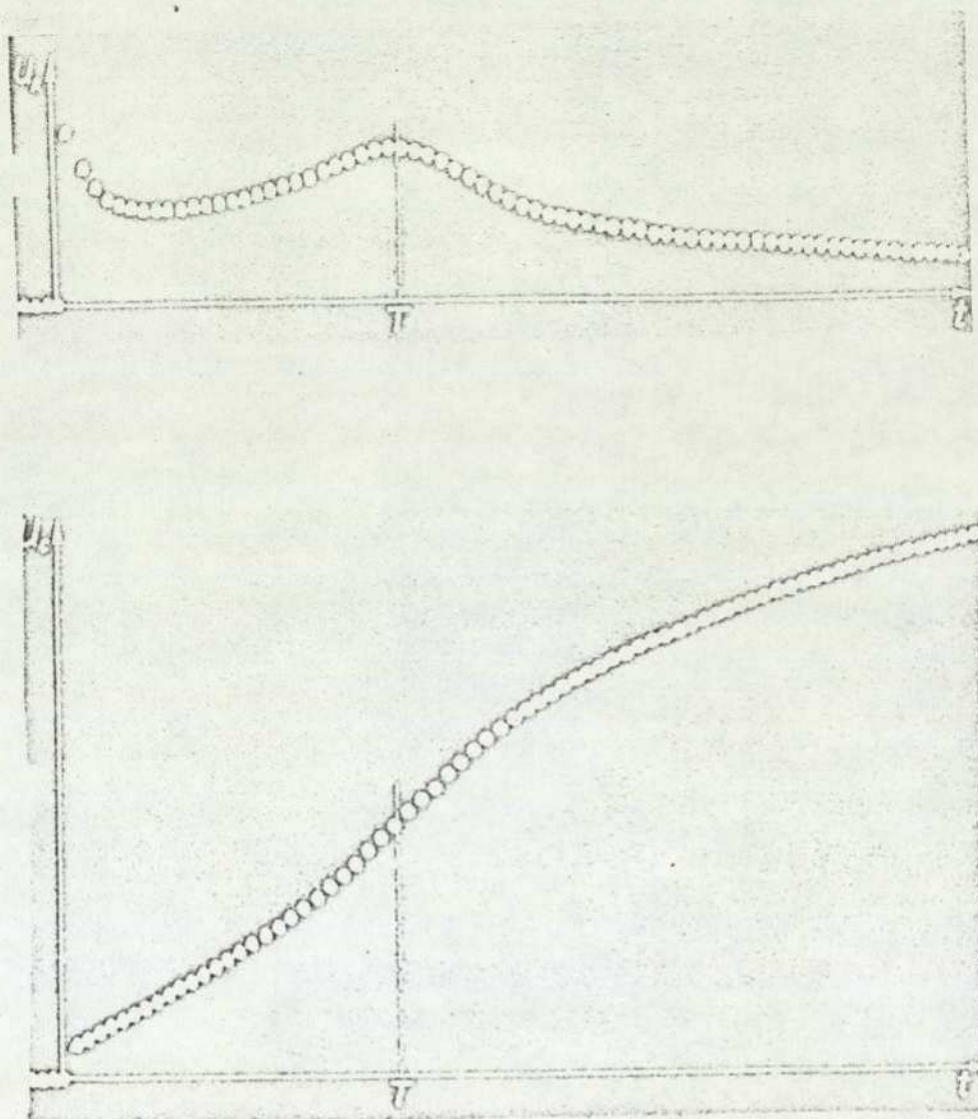
Time dependence of a) potential U_R on resistor, b) potential U_C on capacitor for $\text{Si}_3\text{O}_2(\text{CH}_3)_8$. Time interval between steps is 0.16 sec. Electrode spacing $d = 1$ cm. Applied voltage $V = 790$ V. Negative carriers. Mobilities: $\mu_- = 6.6 \times 10^{-4} \text{ cm}^2 \text{ V}^{-1} \text{ sec}^{-1}$, $\mu_+ = 2.5 \times 10^{-4} \text{ cm}^2 \text{ V}^{-1} \text{ sec}^{-1}$, $\sigma \sim 10^{-12} \Omega^{-1} \text{ cm}^{-1}$

Fig. 3.33 (after Jackym et al)

The mobility of such carriers is about $6.6 \times 10^{-8} \text{ m}^2/\text{v}\cdot\text{sec}$. The flat top to the transient indicates that the field is quite uniform. There is however a slight tail to the current transient which is hidden in the charge transient; showing it is preferable to measure the former.

Fig. 3.34 shows the corresponding curves for decane (conductivity of about $10^{-15} \Omega^{-1} \text{ cm}^{-1}$), at a field strength of about 640 v/cm. The current transient has a very long tail and the current does not fall to zero. A possible explanation is that the natural conduction, due to the low conductivity of the liquid is significant compared to the induced current. There is also the possibility of polarisation currents (c.f. Morant 1968).

These observations suggest that the charge probe technique should not be used when the current transients are obtained without the applied voltage being constant for some time. Croituru has measured the stress distribution in nitrobenzene, by the Kerr effect, and found that it changes rapidly after the application of the field to a liquid. This is to be expected as the non uniform stress is caused by space charge in a dynamic equilibrium; as this charge moves away it is replenished, thus this situation will take time to stabilize.



Time dependence of a) voltage U_R on resistor, b) voltage U_C on capacitor in the case of an inhomogeneous field distribution (decane: $\sigma \sim 10^{-15} \Omega^{-1} \text{ cm}^{-1}$). Electrode spacing $d = 1.4 \text{ cm}$. Applied voltage $V = 902 \text{ V}$. Positive carriers. Interval between steps is 0.16 sec

Fig. 3.34 (after Jackym et al)

3.10 CONCLUSIONS

The transients examined are not consistent with a model of a simple carrier moving in a uniform field. It is not possible to draw many conclusions from these results, but they suggest various lines for further study. Experimenters, apart from Le Blanc, have had difficulty in obtaining noise-free transients from pulse injection and have resorted to integrating the transients, which although reducing the noise, hides possibly valuable information. They have also tended to approach the subject with a fixed concept of a measurable discrete carrier mobility, and ignore any irregularities.

The outstanding question is - what is causing these unusually shaped transients? There are several factors which could contribute, such as the effects of bulk ionisation, liquid motion, measuring technique, cell design, amount of degassing, non uniform stress distribution, multiplicity of carriers, etc.

Some clues have been provided by the research cited in the review. But there is clearly a need for experiments to determine the shape of the current transients for various conditions and to eliminate some of the explanations given in the past.

CHAPTER 4

REAL TIME COMPUTING

4.1 INTRODUCTION

The review of experimental research in liquids has shown that there is considerable uncertainty over the shape of the current transients due to slow carriers and their consequential mobility. This situation has lead to several hypotheses being put forward to explain the transport mechanism.

At the start of this research, it became apparent that the experimental measurements to be undertaken would be likely to produce results, which would be very hard to interpret in terms of the more simple hypotheses. Thus recourse to more complicated theories would be likely. Possibilities could include the effects of two carriers (as suggested by Buttle 1969 and Taylor 1972) surface trapping, non uniform fields and bulk ionisation etc. These factors could vary with field strength and also be non linear.

To anticipate these complications, a computer model of the experiment was developed, sufficiently capable and flexible to be able to cope with all these possible exigencies. The concept was not only that of aiding transient analysis, but also of running the model concurrently with an experiment. This would act as an aid to the performance of the experiment. The background, development, testing and application of this model is described in the rest of this chapter.

4.2 THE APPLICATION OF REAL TIME COMPUTING

The computer used for this work was a Ferranti 1600B minicomputer. This is a 24 bit machine, unlike most minis which are 16 bit. This has three addresses per instruction, making it effectively twice as powerful as a one address machine. So its processor is roughly comparable in power to an IBM 360/50 or an I.C.L. 1904A.

The configuration initially included 16K of store, 3 teletypes, a paper tape reader and punch, a D/A convertor and a graph plotter. This meant that paper tape was used as the storage medium and due to the lack of a line printer, output was obtained from one of the teletypes. The configuration is shown in fig. 4.1.

Towards the end of the duration of this research, the computer was lost for a year to be modified to have 32K of store (incorporating hardware store protection) and a disc store. These extra items are shown in dotted lines in fig. 4.1.

The available software was very limited and was programmed in "FIXPAC", an assembly level language having almost a one to one correspondance between written instructions and the words in store. For example, there was no software for plotting graphs and virtually no operating system.

One developed feature was an alpha-numeric visual display unit (V.D.U.) made up from a Tekronix storage tube. The hardware, including the interface, was produced by a series of undergraduate projects. This V.D.U. provided a fast noise free output facility when a permanent copy was not required.

The author also developed a suite of graph plotting subroutines incorporating several facilities, which would ease the process of analysing the current transients and assist the preparation of graphs for publication.

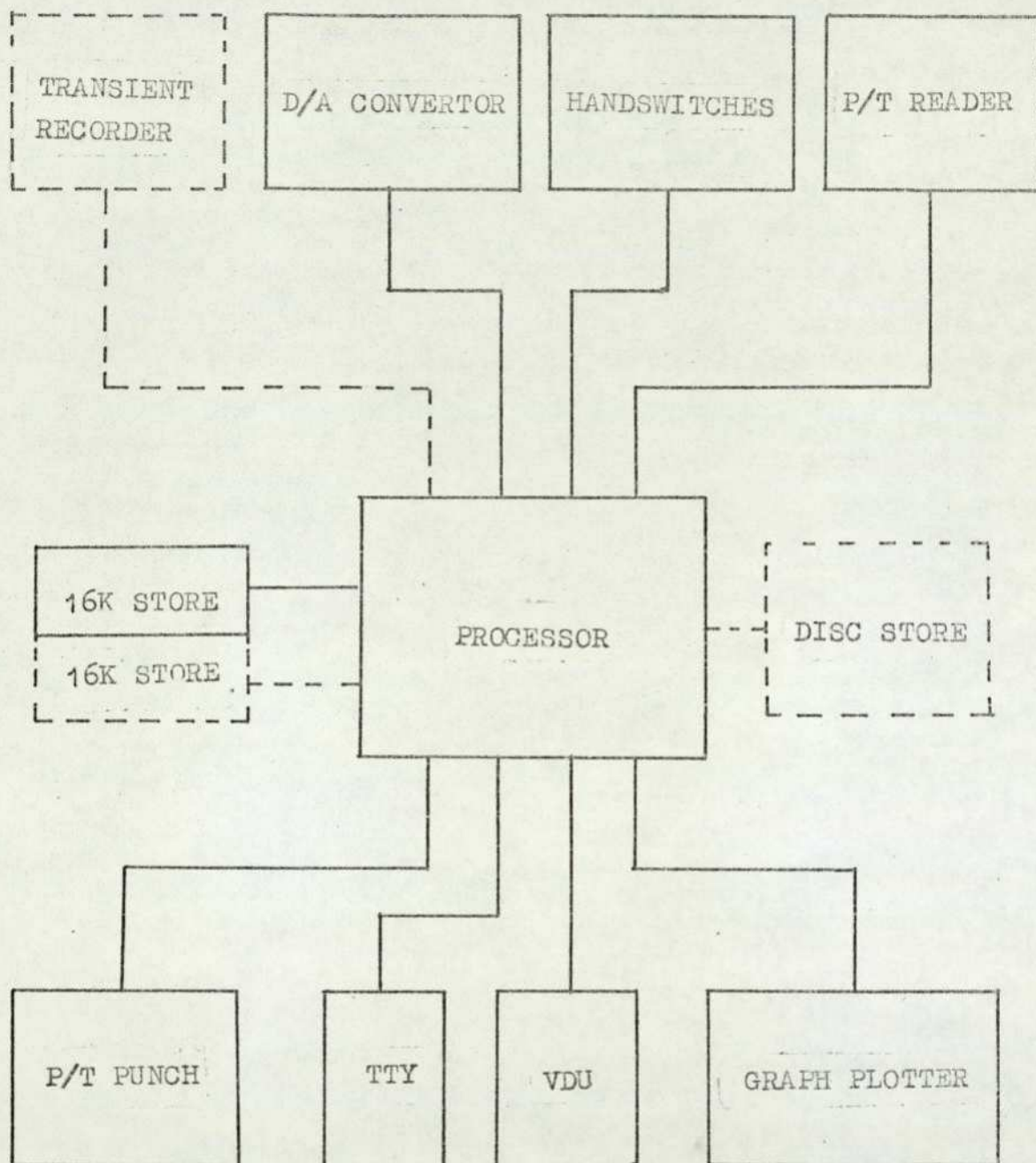


Fig. 4.1 The computer configuration (dotted lines indicate later additions).

The facilities of this included:

- (a) variable sized axes
- (b) preset or auto scaling of the X and Y axes
- (c) the ability to easily title the axes etc.
- (d) the possibility of having several plots on the same axes
(with different symbols)
- (e) plots on either the V.D.U. or plotter
- (f) numerical integration of curves (using Simpson's rule)

The author also supervised an undergraduate project which aimed at further enlarging the scope of these programmes to incorporate such facilities as log scales. An example of the use of these facilities is the graphs of the analysis of current transients in terms of a non uniform stress distribution, given in section 3.9.

Thus, as the computing facilities were developed throughout the time of this study, a flexible approach was adopted in their implementation. However, there was an overall conception which acted as an objective. This was to feed the output of the current measuring apparatus (section 5.6) into the computer via the D/A convertor and to store all the current transients on disc. This would provide a permanent storage of the test results enabling the use of the computing power to analyse them; and the output facilities to produce the results in a convenient format (either graphically or numerically) on any of the output peripherals.

It was also hoped to use the computer in a real time mode while performing measurements. Possibilities included the prediction of the onset of space charge, and liquid motion effects. In the event of the experimental results being non linear, the model was planned to be used concurrently with the experiments to guide the direction of the experiment and to test various hypotheses.

The latter adaptive application is one of the main uses of a real time computer in a laboratory. The development of a computer model suitable for such purposes is described in the following sections.

4.3 THE BACKGROUND TO THE DEVELOPMENT OF THE COMPUTER MODEL

Before any experimental work was undertaken, a computer model was to be developed which would simulate the motion of charge in a dielectric. Such a model would have to be capable of adaptation to cope with many different situations.

Now the analytical solutions of charge transport, discussed in section 2.2, are of very limited application. One reason is that these solutions are for only certain charge injection rates, (such as step, pulse or S.C.L) and cannot be adapted for any other function of time.

It is possible to use numerical techniques to obtain a wider set of solutions. Hartmann and Lipari (1973) gave the variation in field distribution and the resulting current transient for various time dependent rates of injection in the S.C.P. regime.

The general expression for the charge injection was

$$Q = A \left[1 - \exp\left(-\frac{t}{\tau}\right) \right] \quad 4.1$$

where A is the total charge injected

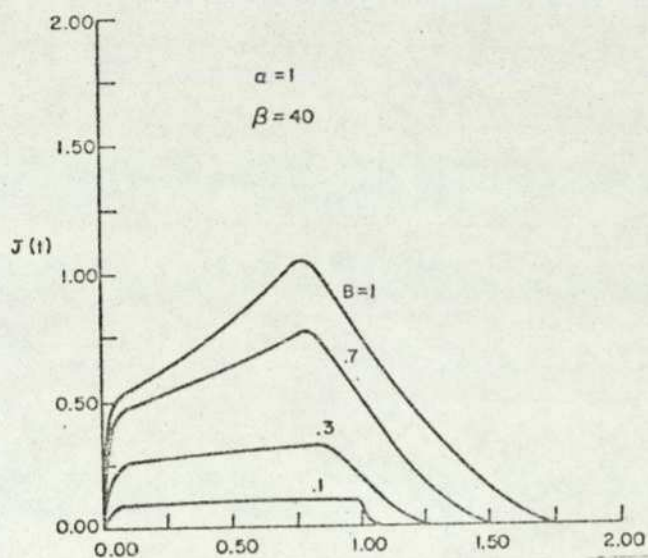
and $\frac{1}{\tau}$ is the rate of injection.

The results are expressed in terms of two parameters, β and B . B is a measure of the quantity of charge and β of the rate of injection as a fraction of the transit time τ .

Fig. 4.2 shows the current transient for $B=40$; a very high rate of injection (i.e. approaching a pulse). It reveals that as B approaches 1, (the S.C.L. case), the transients show more distortion from the rectangular shape in the absence of space charge. Figures 4.3 to 4.6 show how the transients change with a decreasing rate of charge injection. In the absence of space charge, the current has the form of

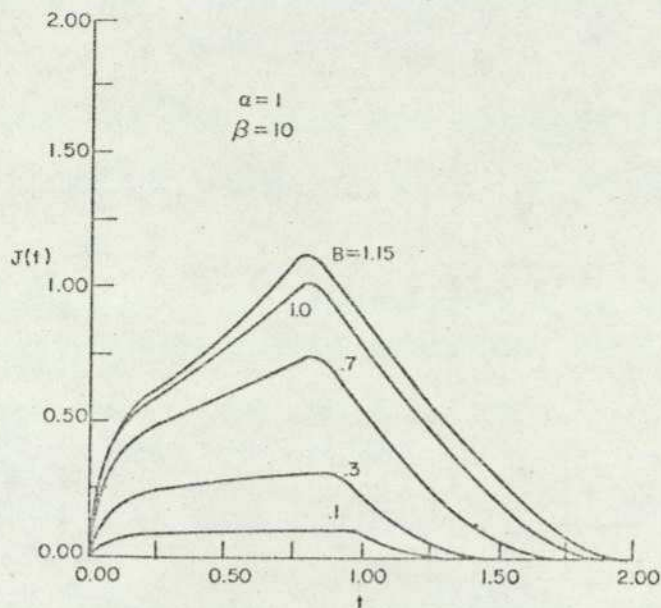
$$i \propto 1 - \exp(-\beta t) \quad \text{for } t < \tau$$

$$\text{and } i \propto (-\beta t) \quad \dots \quad t > \tau$$



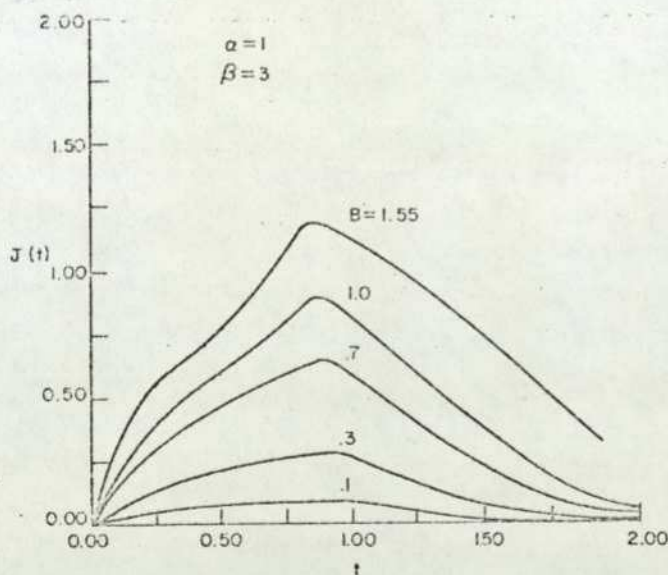
(a) Time dependence of the current for different values of the injected charge. There is no blocking layer ($\alpha = 1$), and the rate of injection is very high ($\beta = 40$).

Fig. 4.2 S.C.P. pulse current transients (after Hartmann and Lipari)



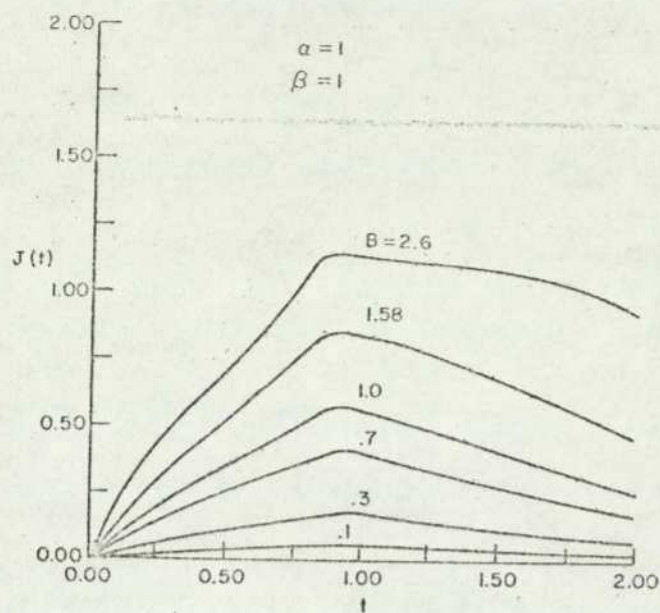
Time dependence of the current for different values of the injected charge. The value $B = 1.15$ corresponds to a space-charge-limited condition. There is no blocking layer ($\alpha = 1$), and the rate of injection is high ($\beta = 10$).

Fig. 4.3 S.C.P. pulse current transients (after Hartmann and Lipari)



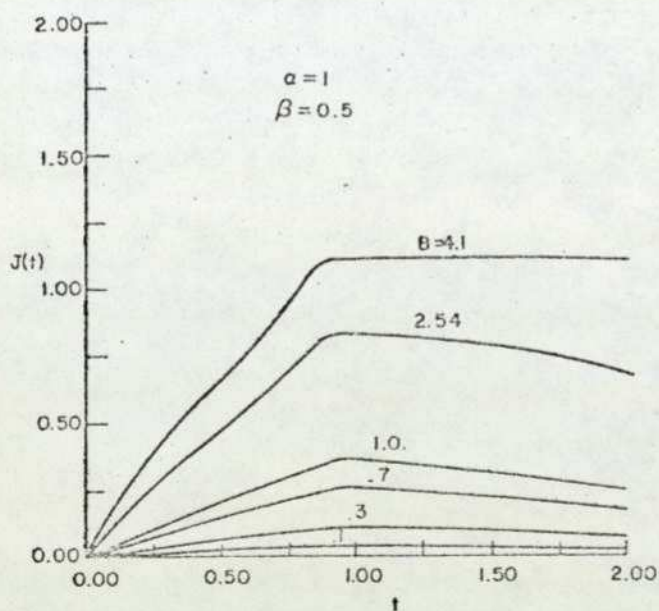
Time dependence of the current. The value $B = 1.55$ corresponds to a space-charge-limited condition. There is no blocking layer ($\alpha = 1$), and the rate of injection is intermediate ($\beta = 3.0$).

Fig. 4.4 S.C.P. pulse current transients (after Hartmann and Lipari)



Time dependence of the current. The value $B = 2.6$ corresponds to a space-charge-limited condition. There is no blocking layer ($\alpha = 1$), and the rate of injection is low ($\beta = 1$).

Fig. 4.5 S.C.P. pulse current transients (after Hartmann and Lipari)



Time dependence of the current. The value $B = 4.1$ corresponds to a space-charge-limited condition. There is no blocking layer ($\alpha = 1$), and rate of injection is very low ($\beta = 0.5$).

Fig. 4.6 S.C.P. pulse current transients (after Hartmann and Lipari)

Zahn et al (1974) used numerical techniques to calculate the charge and field distributions as well as the current transient for various non-zero emitter fields.

To summarise, the above numerical methods can cope with unusual conditions, but they are still rather unadaptable because they start from analytical expressions.

An alternative approach has been provided by Buttle (1969). The basis of his method is to divide, or quantise the dielectric into a number of cells in series. The method starts with a known distribution of charge, i.e. the quantity in each cell. This known distribution can, in fact, be the redistributed output provided by the previous cycle of the simulation. From the use of Poisson's law, it is then possible to calculate the stress distribution across the dielectric, and hence in each cell.

From then on, each cell is individually dealt with in turn. The distance the charge would move in the simulation time is calculated, and the charge is moved (or rather redistributed) accordingly. This is effectively one complete cycle of the simulation and provides as an output, the charge and stress distribution and the instantaneous current.

This process is illustrated in fig. 4.7 for the charge in the nth cell.

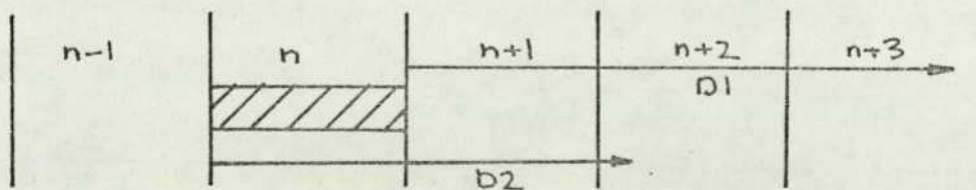


Fig. 4.7 Illustration of Buttle's computer model for the nth cell

The distance D1 is calculated from the average of the field strength in the nth and n+1 cells, and correspondingly from n-1 and n for D2. The charge is then replaced into cells n+2 and n+3 in the correct proportions. Special provision is incorporated for when the charge reaches the collector.

This programme was written in the FORTRAN language, and as this approach seemed promising, the author reprogrammed it in FIXPAC language, to run on the FM1600B computer in order to assess its potential. The advantages of doing this were several. The computer was entirely at the user's disposal, thus enabling him to drive the model on line. The V.P.U. was used to monitor its progress; either with graphs of the various distributions or with numerical data from the number of trace facilities built into the programme. A total of four levels were provided and examples will be provided.

When completed, the programme was tested and was found to give a reasonable S.C.L. transient. To obtain this, the model initially contained no charge. Then sufficient was injected into the first cell to bring the cathode stress to zero. After a cycle more charge was added to the first cell to again bring the cathode stress to zero.

This provides an opportunity to illustrate and enlarge on the explanation of S.C.L. process described in section 2.2.

The model was run with a 100 cells and a normalised time per simulation of .05.

The following is part of a trace output.

```

CYCLE NO.      1
TOTAL Q1=  +0.000000 E + 0
TOTAL Q2=  +0.000000 E + 0
TOTAL Q3=  +0.000000 E + 0
PREVIOUS JT=  +0.505173 E + 0
CHARGE ADDED=  +0.100497 E + 1
NEW Q1 1  +0.100497 E + 1
EO=  +0.223993 E - 3
EBAR=  +0.502606 E + 0
1

```


D1= +0.112224 E - 2
 D2= +0.603589 E + 1
 Q DASH= +0.166523 E + 0
 A(1)= +0.166340 E + 0
 JT= +0.505173 E + 0
 A(7)= +0.597620 E - 2
 2

EBAR= +0.100516 E + 1
 D1= +0.603589 E + 1
 D2= +0.703589 E + 1
 Q DASH= +0.000000 E + 0
 A(7)= +0.597620 E - 2
 JT= +0.505173 E + 0
 A(8)= +0.000000 E + 0
 3

1 +0.166340 E + 0
 2 +0.166523 E + 0
 3 +0.166523 E + 0
 4 +0.166523 E + 0
 5 +0.166523 E + 0
 6 +0.166523 E + 0
 7 +0.597620 E - 2
 JT= +0.505173 E + 0
 EO= +0.255671 E - 1

CYCLE NO. 2
 TOTAL Q1= +0.100488 E + 1
 TOTAL Q2= +0.000000 E + 0
 TOTAL Q= +0.100488 E + 1
 PREVIOUS JT= +0.505173 E + 0
 CHARGE ADDED= +0.256348 E - 1
 NEW Q1 1 +0.192032 E + 0
 EO= +0.799967 E - 4
 EBAR= +0.960922 E - 1
 1

D1= +0.400804 E - 3
 D2= +0.196243 E + 1
 Q DASH= +0.978599 E - 1
 A(1)= +0.978298 E - 1
 JT= +0.184517 E - 1
 A(2)= +0.941906 E - 1
 2

EBAR= +0.275952 E + 0
 D1= +0.196243 E + 1
 D2= +0.379669 E + 1
 Q DASH= +0.907793 E - 1
 A(2)= +0.976009 E - 1
 JT= +0.643024 E - 1
 A(4)= +0.723209 E - 1
 3

An explanation is as follows. The "TOTAL Q1 = 0" statement shows that the medium is initially free of charge. (Although Q2 is printed, it has no relevance for the same driving programme 4718

is used for both this and the later model).

The programme then adds a unit of charge into the first cell which brings the cathode stress (E_0) to zero. The model calculates D_1 and D_2 , the distances the front and rear edge of the charge moves, from the mean stress level at the cell boundaries ($\bar{E}$). It then calculates the new level of charge (Q_{DASH}) and redistributes it accordingly. This is shown at the end of the first cycle.

The current due to this motion is given as .505173 and the cathode stress has risen to .025; this will be used to calculate the charge injection for the next cycle.

The process continues like this and the resulting current transient is shown in fig. 4.8 (c.f. fig. 2.3) for the model run with 200 cells.

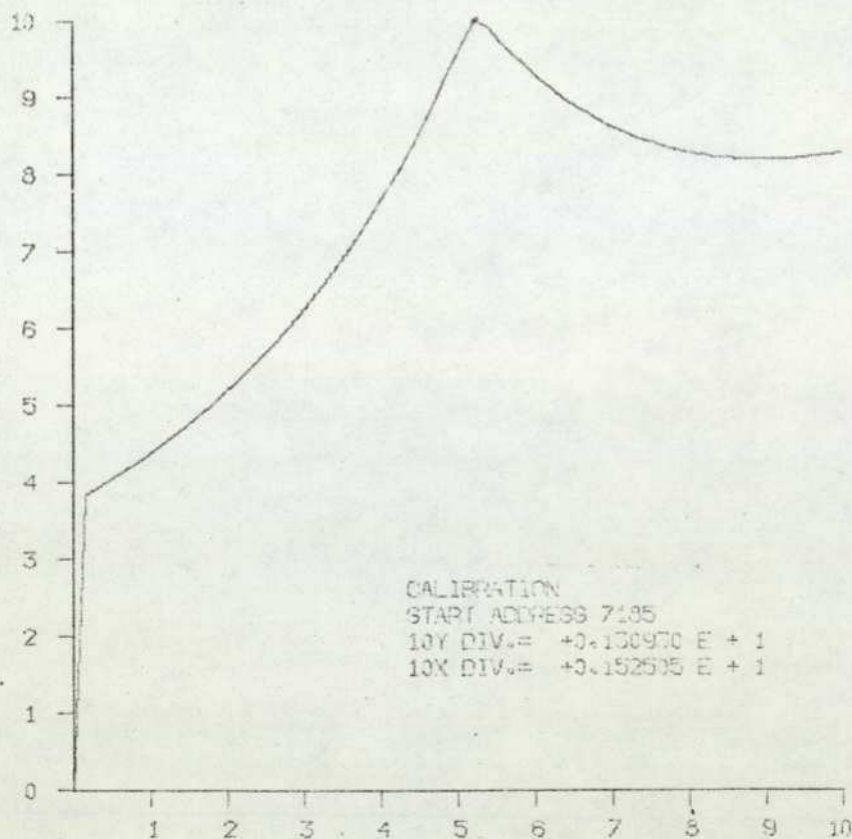


Fig. 4.8 S.C.L. current transients from the model

Curves of the stress and charge distributions at various times are given in figs. 4.9 to 4.12. The calibration on the graphs gives the cathode stress and the current level. These are summarised below together with the correct values calculated from the analytical expressions (shown in brackets) and the % error.

<u>TIME</u>	<u>COLLECTOR STRESS</u>	<u>CURRENT</u>
.425	1.27106 (1.26984) +.01%	.783356 (.806248) -2.84%
.65	1.47995 (1.26984) - .1%	1.05649 (1.09739) -3.73%
.80	1.63394 (1.63930) - .33%	1.3093 (1.3437) -2.56%

These figures reveal the model to be reasonable accurate. This will be considered further in section 4.5.

It can be appreciated that such a model can accommodate any injection rate. However the purpose of the model was to aid the interpretation of current transients. The review of liquids in chapter 3 indicated that the causes of distorted transients could include the effects of static space charge and multiple carriers. This model is unable to cope with such things; for, from the above description it is clear that the model assumes the stress distribution to be either constant, or that it moves with the charge (i.e. under S.C.P. conditions). Hence the model would not be able to cope with static space charge.

So a very general adaptable model was constructed, capable of coping with most of the possible phenomena which could occur in both solid and liquid dielectrics. For example, it seems possible that slow carrier transients in liquids could be the consequence of injected carriers being trapped, while traversing across the gap, to form larger

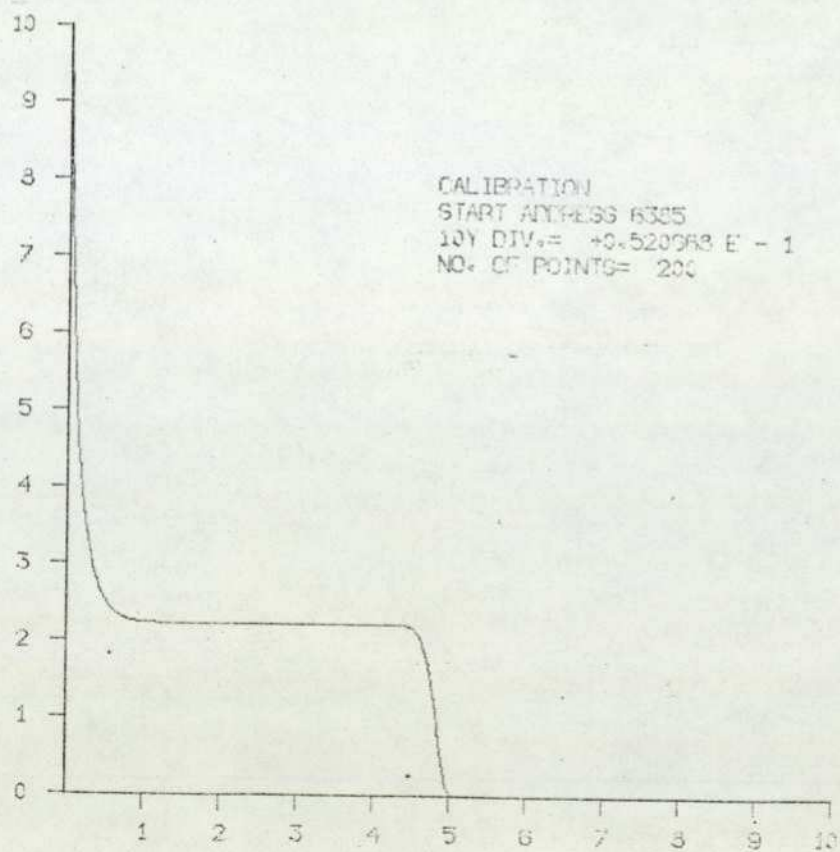
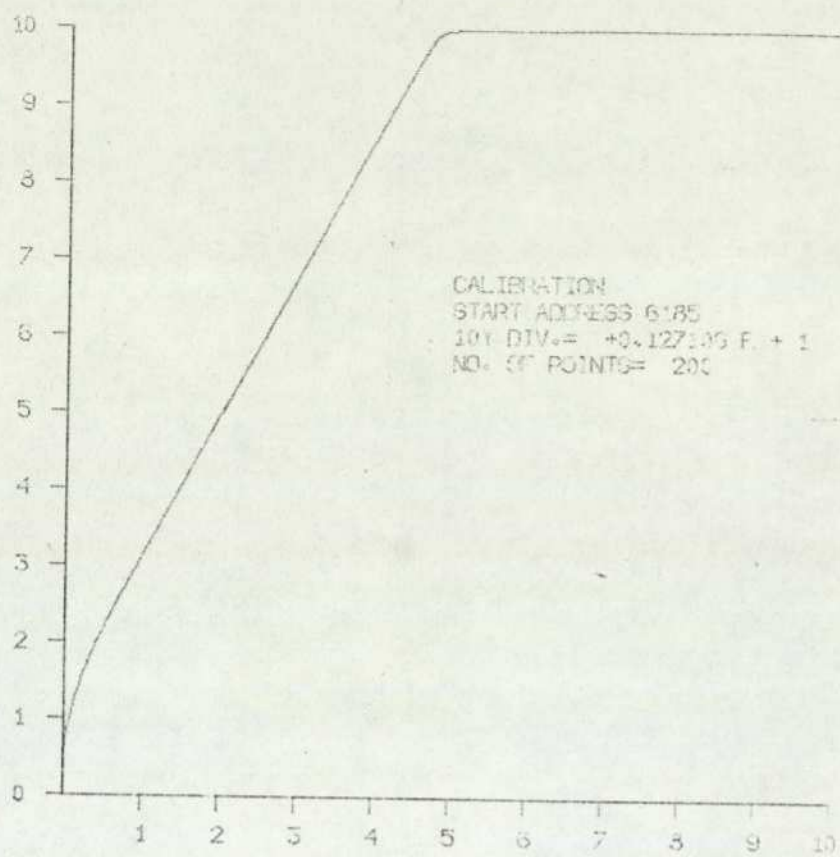


Fig. 4.9 Stress and charge distributions at $t = .425$

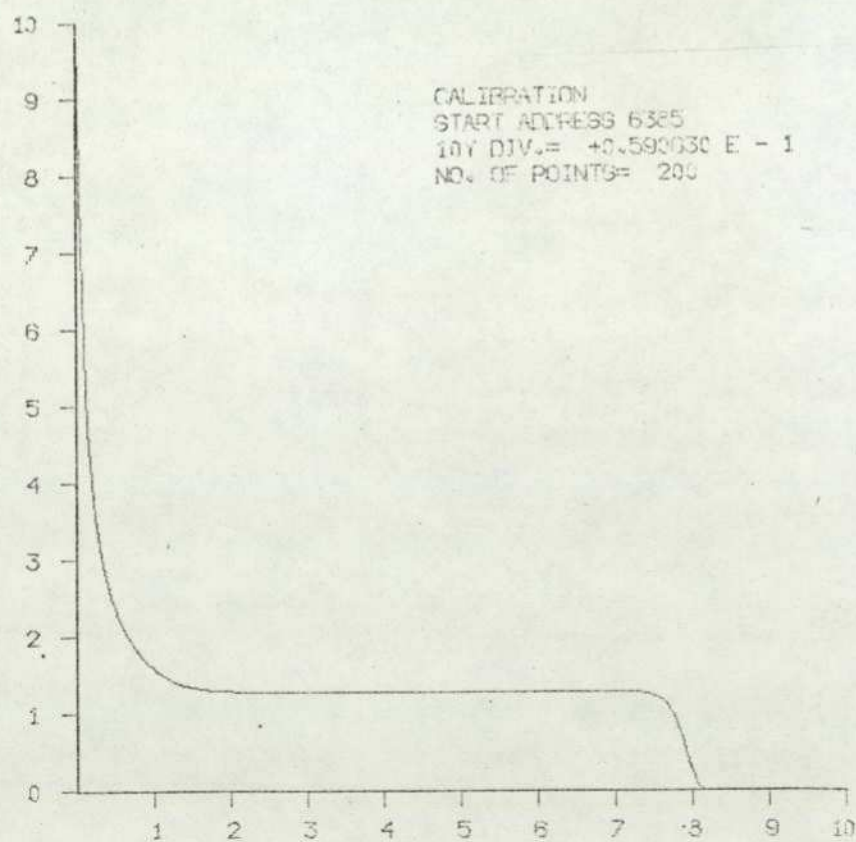
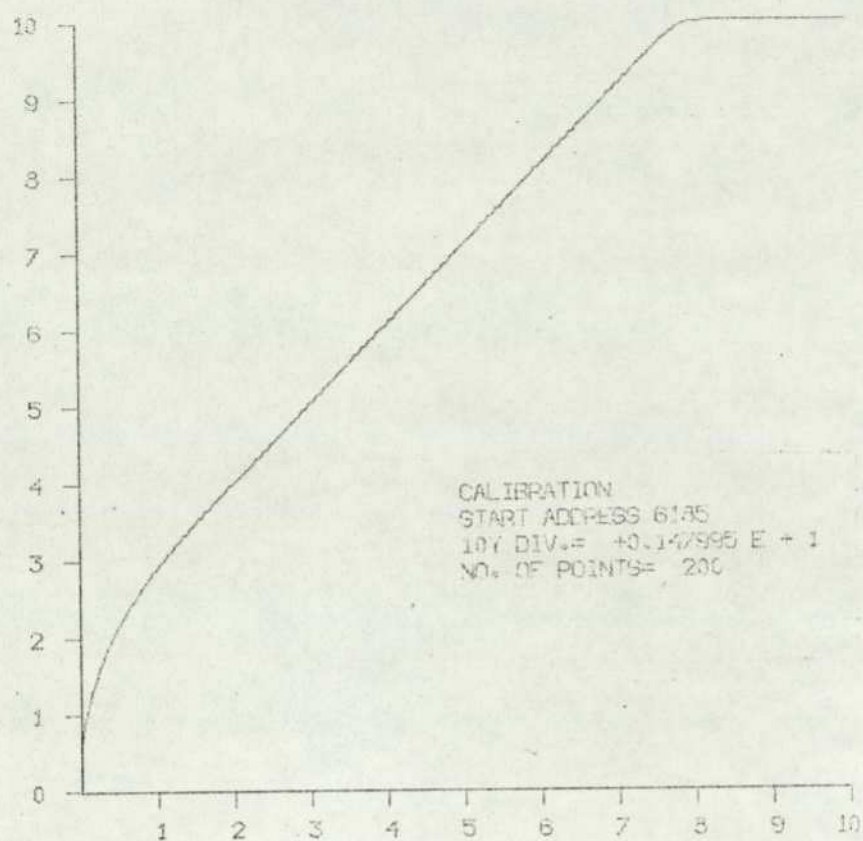


Fig. 4.10 Stress and charge distributions at $t = .65$

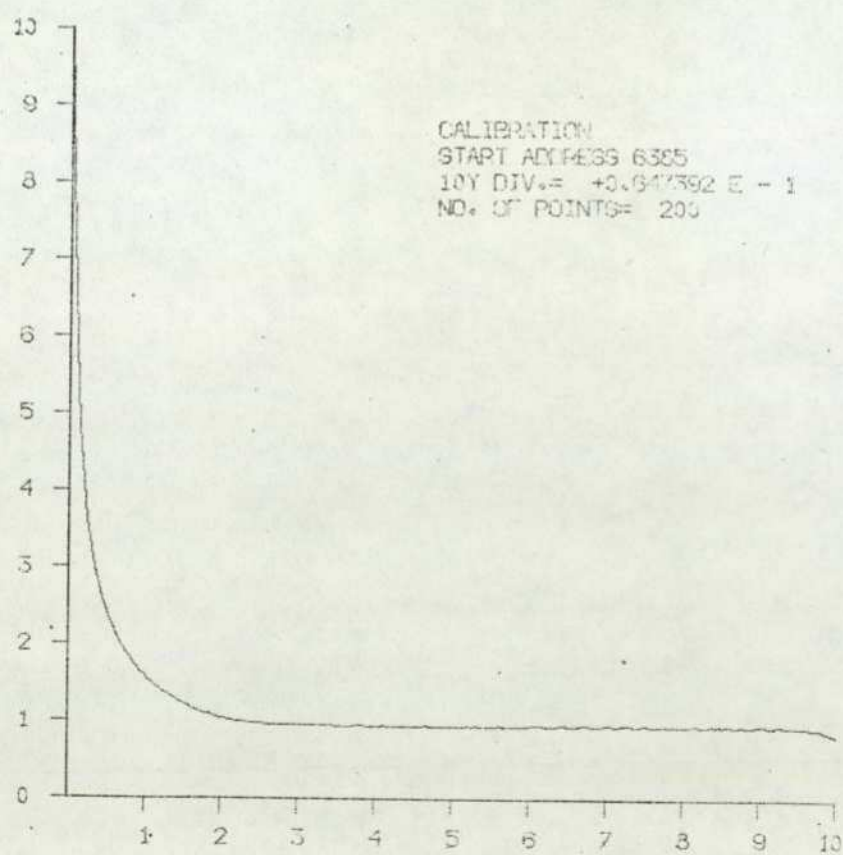
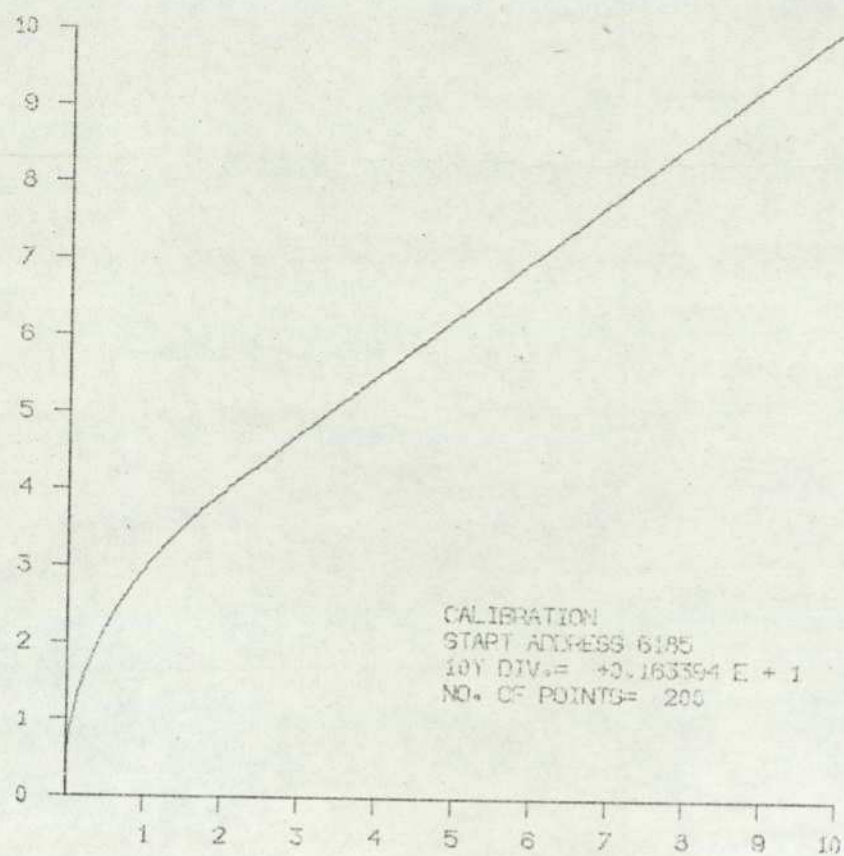


Fig. 4.11 Stress and charge distributions at $t = .8$

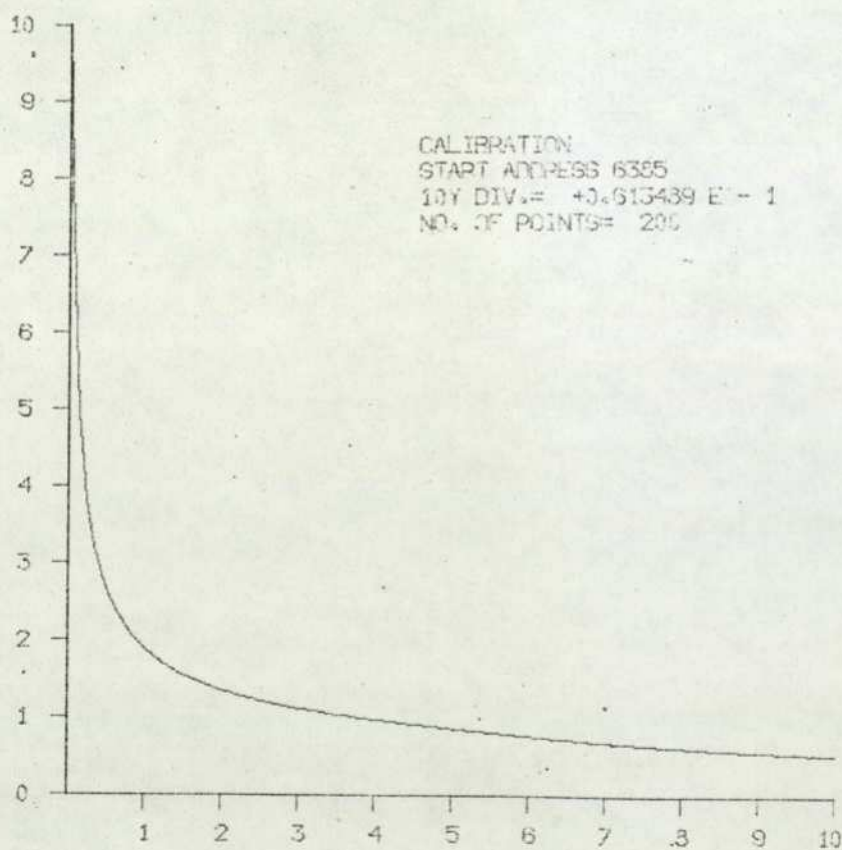
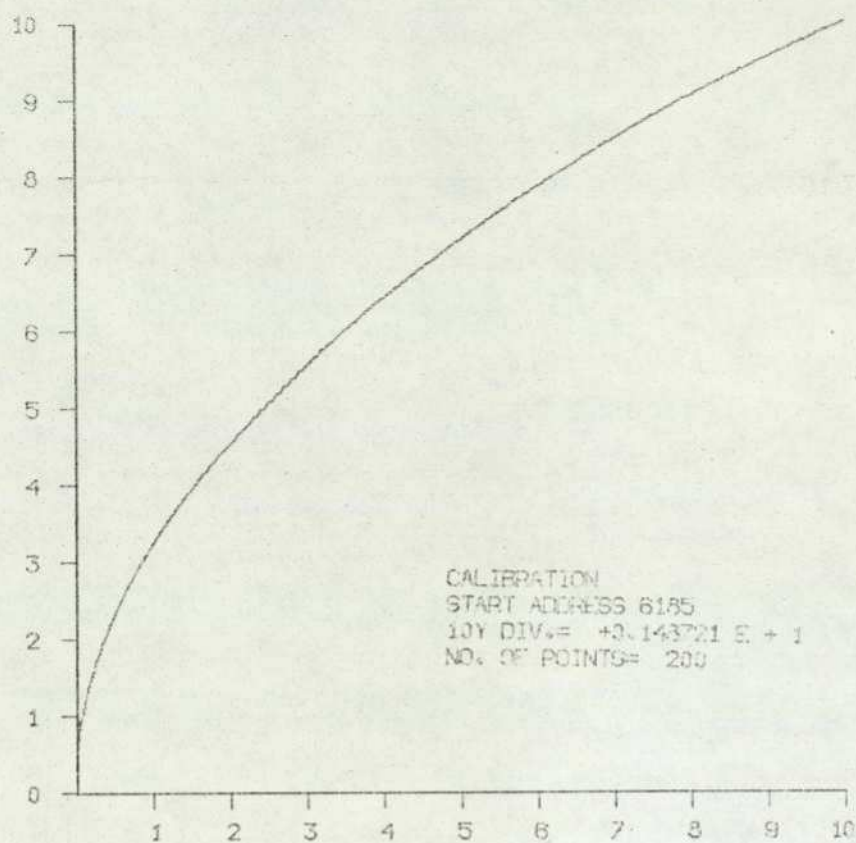


Fig. 4.12 Stress and charge distributions at $t = 1.525$

slower carriers. This could fit in with Buttle's contention that his results indicated two slow carriers mobilities, with a ratio of 3. Such a phenomenon had not been considered before and its effects on the transients were unknown.

The model would also cope with the trapping of fast carriers in ultra pure liquids into the slower ionic carriers. Another use would be to emulate the process of ionic carriers breaking up at high fields, to form fast carriers.

Such uses of a model are comparable to the trapping phenomena in solids, if the second carrier has zero mobility. Thus the analysis of trapping in solids will provide useful tests of this proposed model.

4.4 DESCRIPTION OF THE MODEL AND ASSOCIATED SOFTWARE

This section is devoted to a description of the software developed to aid the interpretation of current transients. The major part of this software is the programme which simulates charge motion in a dielectric (4720). The programmes are listed below with the number of words of store taken up.

<u>No.</u>	<u>Storage</u>	<u>Title</u>
4718	224	CHARGE INJECTION SUBROUTINE
4719	463	DATA INPUT PROGRAMME
4720	1360	MODEL
4725	90	CALCULATES STRESS FROM CHARGE
4722	16	KEY TO DATA AREAS
4741	21	CALCULATES S.C.L. INJECTION
4825	81	OUTPUT SUBROUTINE FOR MODEL
4807	59	DICTIONARY FOR OUTPUT SUB.
4805	463	GENERAL GRAPH PLOT AND DATA O/P.
4714	88	FOR USE OF PLOTTER SUBS ON V.D.U.
4715	65	GRAPH PLOT ON V.D.U.
4850	187	INTEGRATION SUB.

The total storage required for these programmes is 3,117 words. This total does not include the data areas for the model (which are variable in size) or the general Ferranti subroutines.

The information which can be output from the model includes:-

- (a) stress distribution (E v.x)
- (b) major carrier distribution (Q1 v.x)
- (c) minor carrier distribution (Q2 v.x)
- (d) current against time (j v.t)

This data can be onput numerically or graphically. There are also 4 trace levels incorporated into the programme, and a few optional stops to aid programme development.

All these options are controlled by the use of 24 handswitches, which are continually assessed by the software. This provides a very powerful means of driving the simulation. At the flick of a switch, at any point in the simulation, numerical data can be onput on either the tape punch, the V.D.U. or a teletype. The dictionary subroutine can be used at the end of a cycle to onput any of the above distributions.

At the heart of this software is the simulation programme. This is somewhat analogous to the model of Buttle, except that there is provision for two species of charge carrier, with the options of either carrier being converted into the other while in transit. This requires specifying four parameters; the mobilities of each of the two carriers, and the mean lifetime of each carrier before being converted to be the other carrier.

By a suitable choice of these parameters, the model can be made to represent a large number of different transport phenomena. For example if one mobility is put to zero, the model represents a solid dielectric with different trapping and release lifetimes. Or if one lifetime is put to zero, the model represents an ultra pure liquid with the first carriers being trapped to form slow carriers.

The following is a simple description of the model's basic operation. Again the medium is divided into a variable number (I) of serial cells. The stress distribution is claculated from the given charge distributions, Q_1 and Q_2 . Then each cell is dealt with in turn.

Consider the n th cell shown in fig. 4.13.

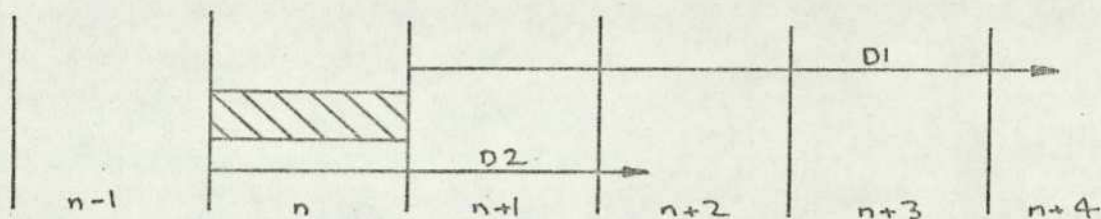


Fig. 4.13 Illustration of the operation of the 2 carrier model

The programme calculates the distances $D1$ and $D2$ moved by the front and back edges of the charge in the simulation time. A fundamental difference between this and Buttle's model is that these distances are calculated from the fields through which the charge passes (i.e. $D1$ from the stress in the cells $n+1$ to $n+4$). It does this by calculating the time to pass through each cell. From these distances, the programme considers each cell which is passed in turn (i.e. $n+1$ to $n+4$). So effectively, as the charge passes through these cells, the programme works out what proportion is transferred to form the other carrier, and what proportion is left which is placed in cells $n+2$ to $n+4$.

Although simple in principle, it is rather complicated to put into effect. One problem is calculating the net current resulting from such motion. For the charge transferred to form the alternate carrier also moves, it will do so at the other mobility and for only a proportion of the cycle time.

It would be a very difficult task to communicate, and explain in detail, the operation of such a complex model. It would also perhaps be undesirable to attempt to do so. Firstly, it would be very hard to

follow, and secondly it would constrain anyone who wished to continue development to follow exactly the same method. It is felt better to communicate the general principles, to enable progress to be made towards a superior version. To this end, several detailed trace outputs of the model will be given as well as suggestions for improvements. The author writes in the light of his experience in understanding and implementing the basic FORTRAN programme given by Buttle.

It is, however, important that any shortcomings in the model should be appreciated and that it should be exhaustively tested for correct operation.

The accuracy is a function of the number of cells the charge passes through per cycle. This is related to the number of cells I , and the time per cycle. Obviously, if this discrete model is too coarse, then poor results will occur as charge can only be injected between each cycle. An extreme example would be to have the injected charge totally traversing the gap before more charge is added.

Alternatively, if too many simulation cycles occur per transit time and the number of cells is too few, then the distances D_1 and D_2 will be less than the cell width causing quantisation errors. These problems can be eliminated by the correct setting of the parameters.

The other major problem occurs when the mobilities of the two carriers are similar. This is related to the way the programme copes with the two mobilities. The faster carrier (say n times more than slower) is deemed to be the major carrier and the other, the minor. Every simulation cycle, the major carrier movement is calculated, allowing charge to be transferred into the minor carrier. Also charge is transferred from the minor into the major carrier. But only once in every n cycles is the minor charge carrier motion accounted for. This is to avoid the

previous shortcoming concerning charge having to move at least one cell width.

Thus a more severe test of the charge transfer mechanism occurs as the mobilities become closer together. These problems are better illustrated by actual test results and will be described in the next section.

4.5 SIMULATION TESTS

Perhaps the easiest way of gaining an understanding of the use and working of the model is by the use of examples of simple tests. These tests were undertaken to check for correct operation. It is important to realise that care must be employed in the selection of such tests and the weight attached to their results.

For example, the test used by Buttle was to simulate the S.C.L. transient. This is characterised by a peak current of about $4/3$ at a time of .787, and an initial current of .5. The FIXPAC version of Buttle's programme was used for this, and the results have been given in section 4.4.

It is possible to test the two carrier model in this way by using only one of the carriers. The results were obtained with a 100 cells and a time per run of .05. They are given here together with the results of the earlier programme run under identical conditions.

<u>Time</u>	<u>1st Model</u>	<u>2nd Model</u>
.75	1.17596	1.17303
.8	1.25815	1.25595
.85	1.25467	1.25561
.90	1.21225	1.21335
.95	1.17538	1.17627

This shows that the two models give almost identical results in spite of functioning by different methods. The results also illustrate quantisation errors.

To get more information on these errors the model was run with varying parameters for S.C.L. injection and the test of the accuracy is to determine how close the current is to the correct value of 1.359 at a time of .787.

These results give the current, the % error, the number of simulation cycles (N) to reach that time, the number of cells (I) and the approximate number of cells passed per cycle (D).

<u>N</u>	<u>I</u>	<u>D</u>	<u>i at .787</u>	<u>% error</u>
	1000	8	1.34595	.98
	500	4	1.33820	1.55
100	250	2	1.33170	2.03
	100	1	1.31445	3.30
	50	.5	1.25720	7.5
500		.5	1.29053	4.70
200		1	1.33002	2.15
100	250	2	1.33170	2.02
50		4	1.31879	2.97
25		8	1.27640	6.09
200	1000		1.34903	.75
100	500	4	1.33820	1.55
50	250		1.31879	2.98
25	125		1.29477	4.74

The first group of results show the effect of keeping the time per simulation, N, constant. The second set is for a constant number of cells, and the last is keeping D constant.

The results show that if N is less than 50 or I less than 100, then the simulation is too crude. Also if D is less than 1, even if N and I are very large, then quantisation errors give poor results.

A test capable of checking the charge transfer mechanism is the simulation of deep trapping with just one carrier with a lifetime before trapping of .5 (i.e. the carriers on average get halfway across the gap before being trapped). The distributions of the free and

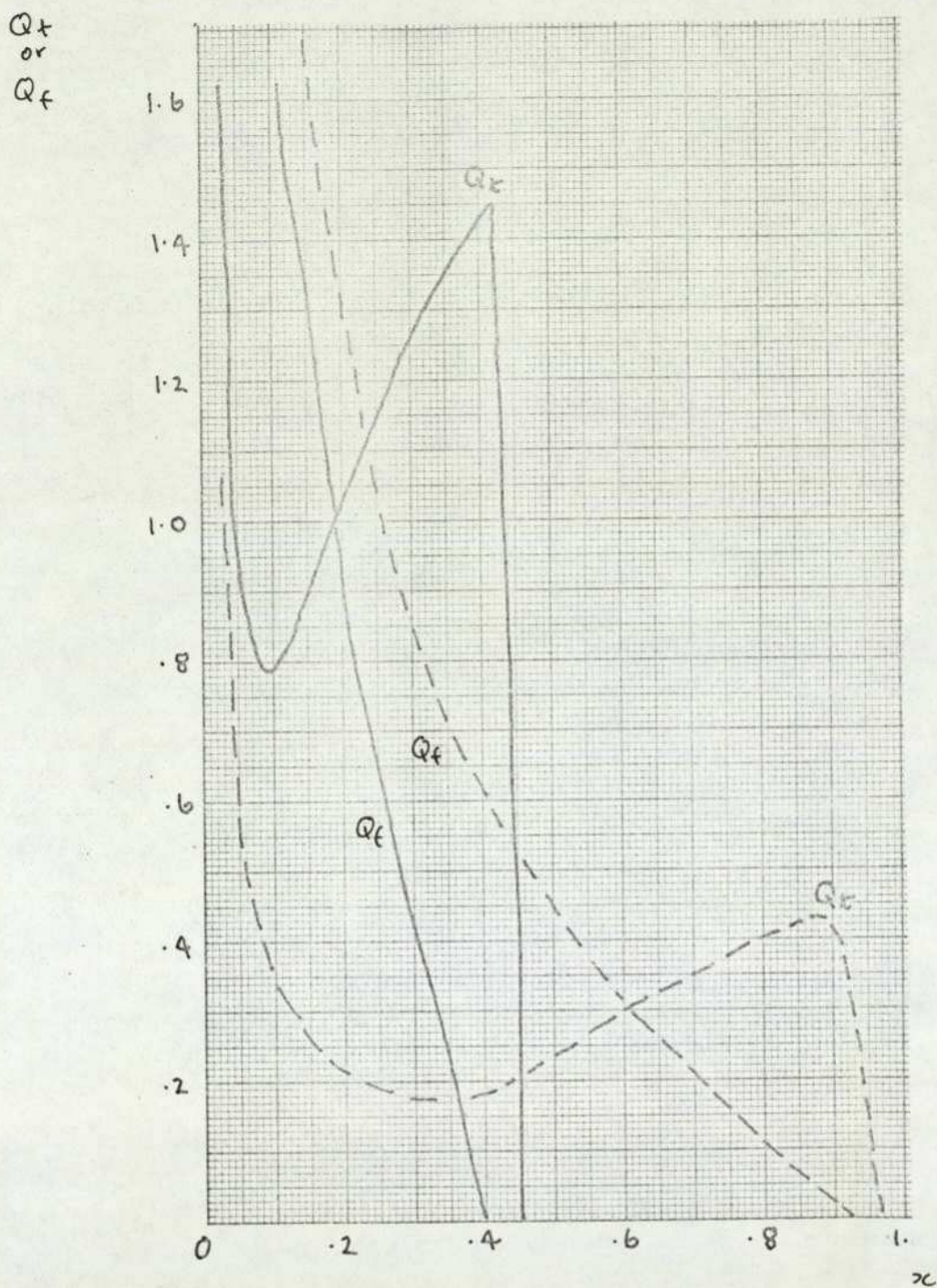
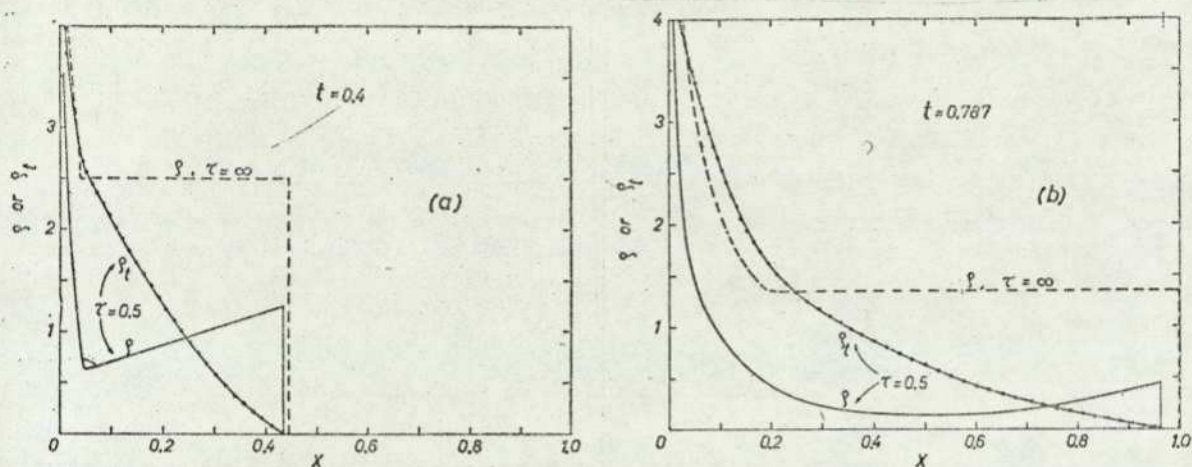


Fig. 4.14 Trapped and free space charge distributions from the model

——— $t = 0.4$
 - - - $t = 0.787$

trapped carriers are shown in fig. 4.14 at times of .4 and .787. These should be compared with the results in fig. 4.15 from Many and Rakavy's paper.



Space-charge distribution along an insulating crystal at two instants following the onset of injection; (a) $t=0.4$, (b) $t=0.787$. The dashed curves are for a trap-free insulator. The solid curves correspond to the free and trapped charge-densities for an insulator with traps ($\tau=0.5$, $\theta_0=0$).

Fig. 4.15 Trapped and free space charge distributions (after Many and Rakavy)

However, neither of these results have taken account of diffusion which can be significant in the S.C.P. regime. This has been considered by Rosental (1973).

During the development of these models it was found possible, on using this S.C.L. test, to get quite reasonable results with a basic error in the model's operation. Such faults could be detected using a detailed trace output. Appendix E gives an example of a trace output for the first two cycles of a simulation of low level pulse injection. In this case the two carrier mobilities are equal and

charge transfer occurs in both directions. The rate of charge transfer used here and in all subsequent tests is the very severe test of having half the charge transferred each cycle. This happens in both directions.

The following is a summary of several cycles indicating the amount of charge "lost" in transit, if any, and the current against time.

<u>Time</u>	<u>Charge remaining</u>	<u>Current</u>
.05	.100001 E-6	.100490 E-6
.10	.999935 E-7	.115231 E-6
.95	.999335 E-7	.115162 E-6
.10	0	.107664 E-6
.15	0	0

This shows that the model has problems coping with two carriers having equal mobility, with charge transfer occurring in both directions. Under low level conditions this produces an error of about 15% in the current. The results for single carrier operation give a corresponding error of .46%. However, virtually no charge is lost and it is moved correctly, this is indicated by the lack of charge in the dielectric after the transit time.

The corresponding results for a ratio of carrier mobilities of 2 is 3% and is 1% for a ratio of 3. So these results demonstrate that the model works adequately under low level conditions provided the ratio of mobilities is not less than 2.

The accuracy under S.C.P. conditions can be gauged by comparing the results with those for a single carrier. Appendix F gives part of a trace output for S.C.P. pulse injection in the two carrier model and the results are summarised below together with those for just one carrier.

<u>time</u>	<u>1 carrier</u>		<u>2 carriers</u>	
	<u>charge</u>	<u>current</u>	<u>charge</u>	<u>current</u>
.05	1.00488	.50518	1.00488	.53695
.10	1.00476	.53048	1.00479	.62016
.15	1.00464	.55798	1.00470	.63911
.80	.98511	1.04715	1.00360	1.12796
.85	.90103	1.01221	.97566	1.17026
.9	.81587	.92551	.90028	1.12848
.95	.73707	.84129	.82341	1.04523
.1	.66406	.76214	.75514	.96425

These results reveal that the 2 carrier case gives too large a current and that the distance moved by the charge is too small.

The final test is S.C.L. injection with two carriers. The results for a 100 cells and a cycle time of .05 are summarised as follows with the results of the model with only one carrier.

<u>time</u>	<u>one carrier current</u>	<u>two carrier current</u>
.1	.505	.537
.15	.530	.620
.75	1.09	1.15
.80	1.17	1.23
.85	1.25	1.31
.90	1.25	1.39
.95	1.21	1.38

These results are consistent with the above conclusions. To summarise it may be said that these tests reveal the model is accurate, providing it is not operated with the carrier mobilities having a ratio of less than 2.

4.6 APPLICATION OF COMPUTER MODEL

4.6.1 Introduction

The computer model is sufficiently adaptable that its possible uses are virtually endless. The adjustable parameters and input data are listed below.

μ_1	1st charge carrier mobility
Q1	1st charge carrier distribution (I values)
μ_2	2nd charge carrier mobility
Q2	2nd charge carrier distribution (I values)
$\tau_{1 \rightarrow 2}$	charge transfer rate from Q1 to Q2
$\tau_{2 \rightarrow 1}$	charge transfer rate from Q2 to Q1
V	the applied voltage
L	gap width
T	simulation time
I	the number of cells.

After one cycle of simulation, the model provides the following:

Q1	the new 1st charge carrier distribution
Q2	the new 2nd charge carrier distribution
E	the stress distribution
i	the resulting current

Of course, between each simulation run, it is possible to adjust Q1 and Q2 to represent charge injection, bulk ionisation and trapping etc.

Some conception of its possible uses can be gained from the following examples.

4.6.2 Application of model to stress analysis

It is possible to provide a verification of the correct operation of both the computer model (4720) and the computer program (4740) which was used in section 3.9 to analyse the current transients in terms of a non uniform stress distribution.

This verification involves putting a fixed charge distribution (i.e. $\rho_2 = 0$) in the model to set up a non-linear field. In this example, a layer of positive and negative charge was placed either side of the centre of the dielectric as shown in fig. 4.16. For this, the graph plotting subroutine have been programmed to plot only the magnitude (which is .05 units) and not the sign of the charge.

The model was then run without any mobile charge in Q1, to give the stress distribution (fig. 4.17). Then a small quantity of charge was injected (.0001 units) to probe this stress distribution. The resultant current is shown in fig. 4.18.

The model was run for a 117 cycle, a few past the transit time which is equivalent in this case to 100 cycles. The correct shape of this current transient would have a square tail. The slight tail given by the model, indicates that the use of 250 cells has caused the charge to spread out slightly. This failing in the model should not be confused with the effects of diffusion, of which the model takes no account.

This current transient was used as input data for the stress analysis programme to see if when the circle is completed if its output of the stress distribution agrees with the original of fig. 4.17.

It is however, necessary to make a judgement of the transit time when deciding how much of the current transient to use in this method of stress analysis. In order to accommodate the slight tail on the transient, and to illustrate the effect of an incorrect estimation; all 117 points will be used instead of the correct value of 100.

The charge transient obtained by integrating fig. 4.18 is shown in fig. 4.19. The results of the stress analysis programme gives the distance moved by the probe against time shown in fig. 4.20 and the

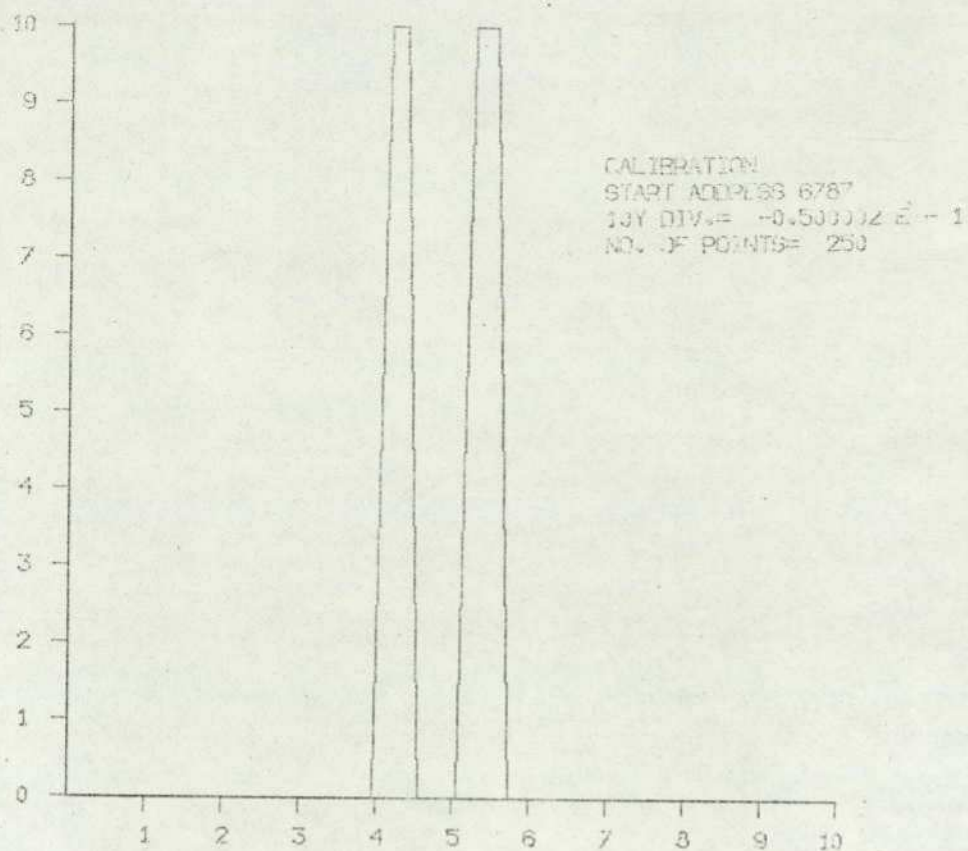


Fig. 4.16 Input charge distribution

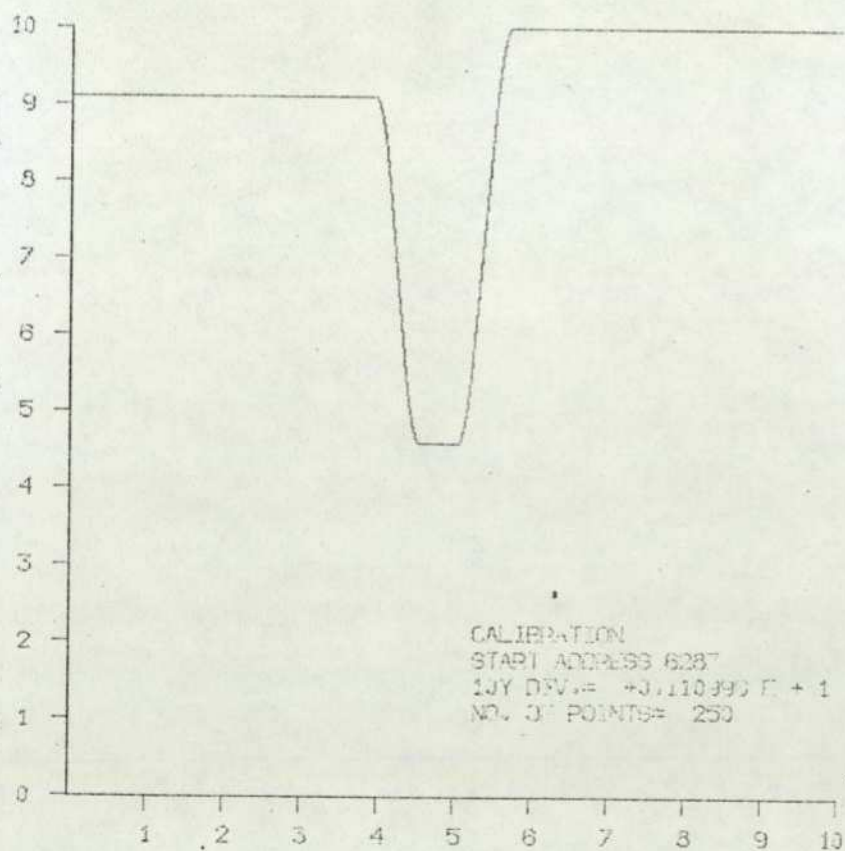


Fig. 4.17 Stress distribution due to the charge of fig. 4.16

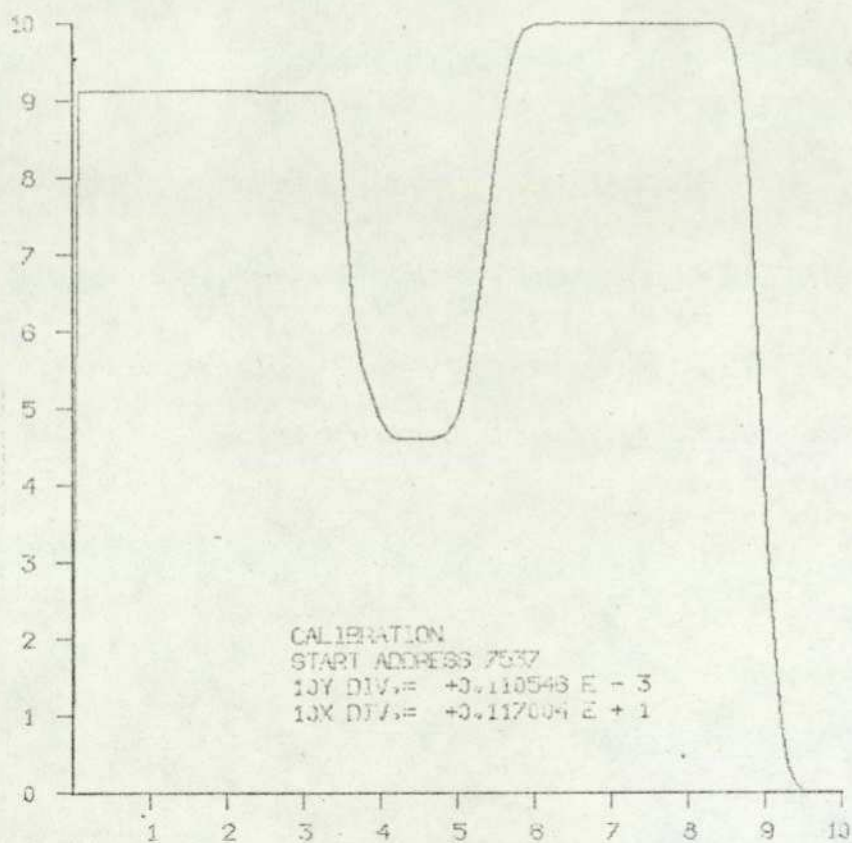


Fig. 4.18 Current transient

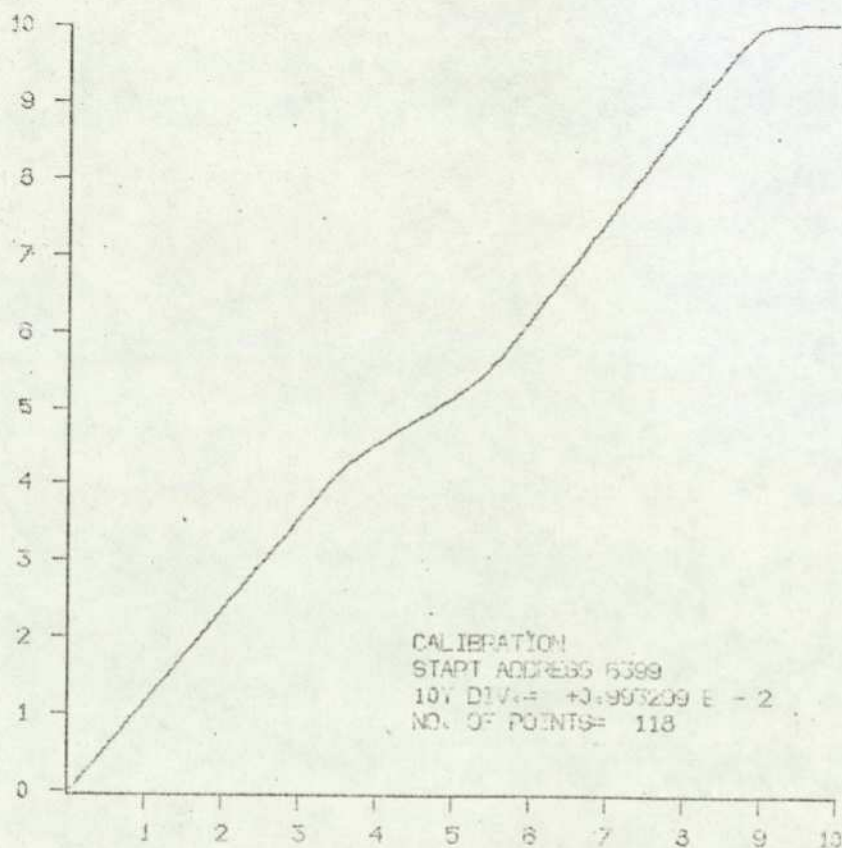


Fig. 4.19 Charge transient (integral of fig. 4.18)

stress distribution shown in fig. 4.21.

A comparison between figs. 4.21 and 4.17 reveals the success of these methods. The reason for the stress falling to zero at the collector is due to putting in too much data, as discussed above.

4.6.3 The production of S.C.P. transients

It is possible to use the model to produce the current transients due to any time dependent rate of charge injection, whether from one electrode or throughout the bulk of the material.

Fig. 4.22 show the S.C.P. transients due to step and pulse injection of charge for injection levels of .1 and .5. The transients have been normalised to aid comparison. These transients will be useful in the detection of S.C.P. effects in the experimental results. Apparently, the transients for S.C.P. step injection have not previously been given in the literature.

4.6.4. Adaptive use of model

Up to now, the model has been used to provide the current transient from a known input stimulus, given certain constraints. But there is no reason why, for the same constraints, that the model could not be used to calculate the necessary charge input for a given instantaneous current. These constraints include assumptions about the charge transport mechanism, such as the mobility and so on.

A special programme was written to use the model in this mode. In simple terms, this programme makes a guess at the charge injection and then runs the model to find just what current this produces. If this trial current is more than .5% out, the programme restores the original charge distribution, re-adjusts the charge injection pro rata and has another trial attempt. This process continues until the target is met. Thus this method can cope with S.C.P. current transients

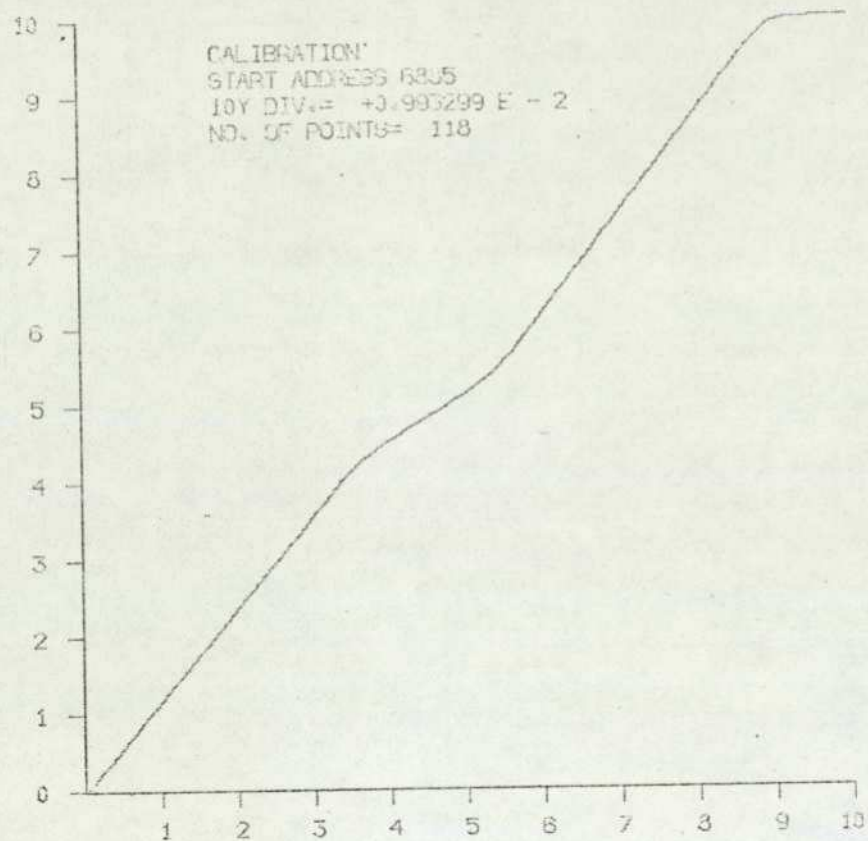


Fig. 4.20 Distance against time for charge probe

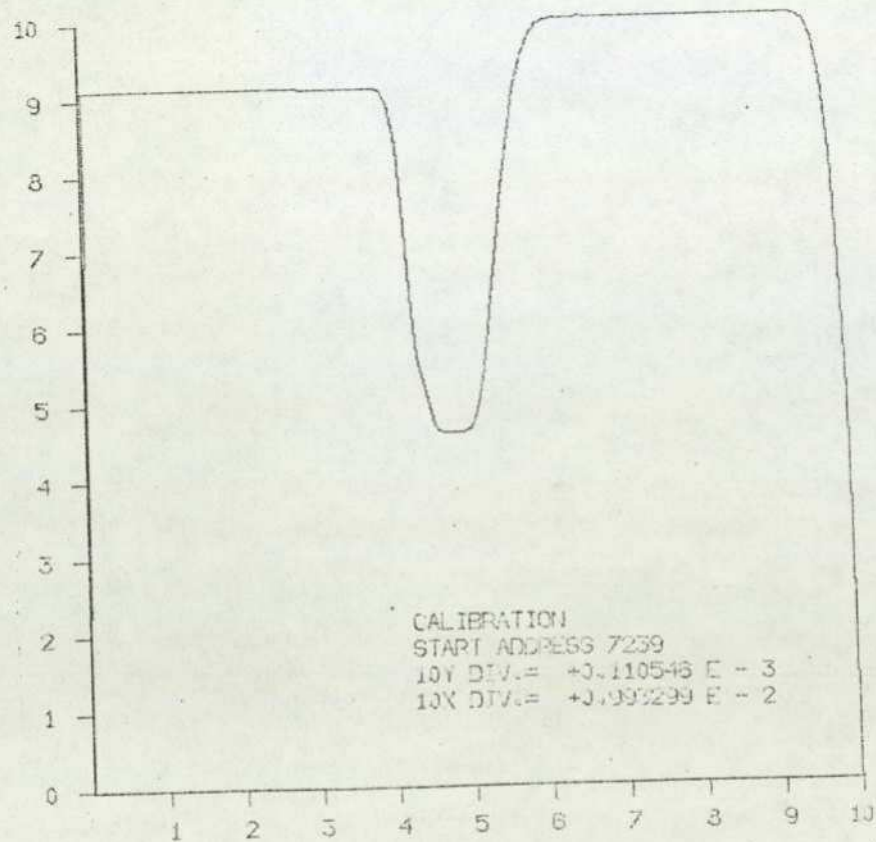


Fig. 4.21 Analysed stress distribution

which do not have a linear relationship between charge injected and current. It is, of course, necessary to take precautions for extreme conditions and this is done by preventing charge injection greater than the S.C.L. value and excluding negative charge injection.

This process continues for all the current input data and the programme provides an output of the charge injection against time as well as the resulting current transient. The success of the method can be gauged by comparing this with the original input transient.

Part of a trace output is given here to illustrate the method.

```
CATHODE STRESS= +0.933643 E + 0
JT TARGET= +0.233273 E - 1

TRIAL Q(1)= +0.532656 E - 2
JT DUE TO 1ST LAYER= +0.233390 E - 2
TRIAL JT= +0.231739 E - 1
FRAC. ERROR= +0.231444 E - 1

TRIAL Q(1)= +0.733453 E - 2
JT DUE TO 1ST LAYER= +0.53331 E - 2
TRIAL JT= +0.233253 E - 1
FRAC. ERROR= +0.424514 E - 4
CHARGE ADDED= +0.733534 E - 2
TOTAL Q(1)= +0.733534 E - 2
JT 13
TRIAL JT= +0.233253 E - 1
```

The term "JT due to 1st layer" is the current contributed by the first layer only. This used to re-adjust the charge injection in the next trial.

A good candidate for this method is the sort of current transient obtained by Hewish and Brignell (1972) shown in fig. 4.23.

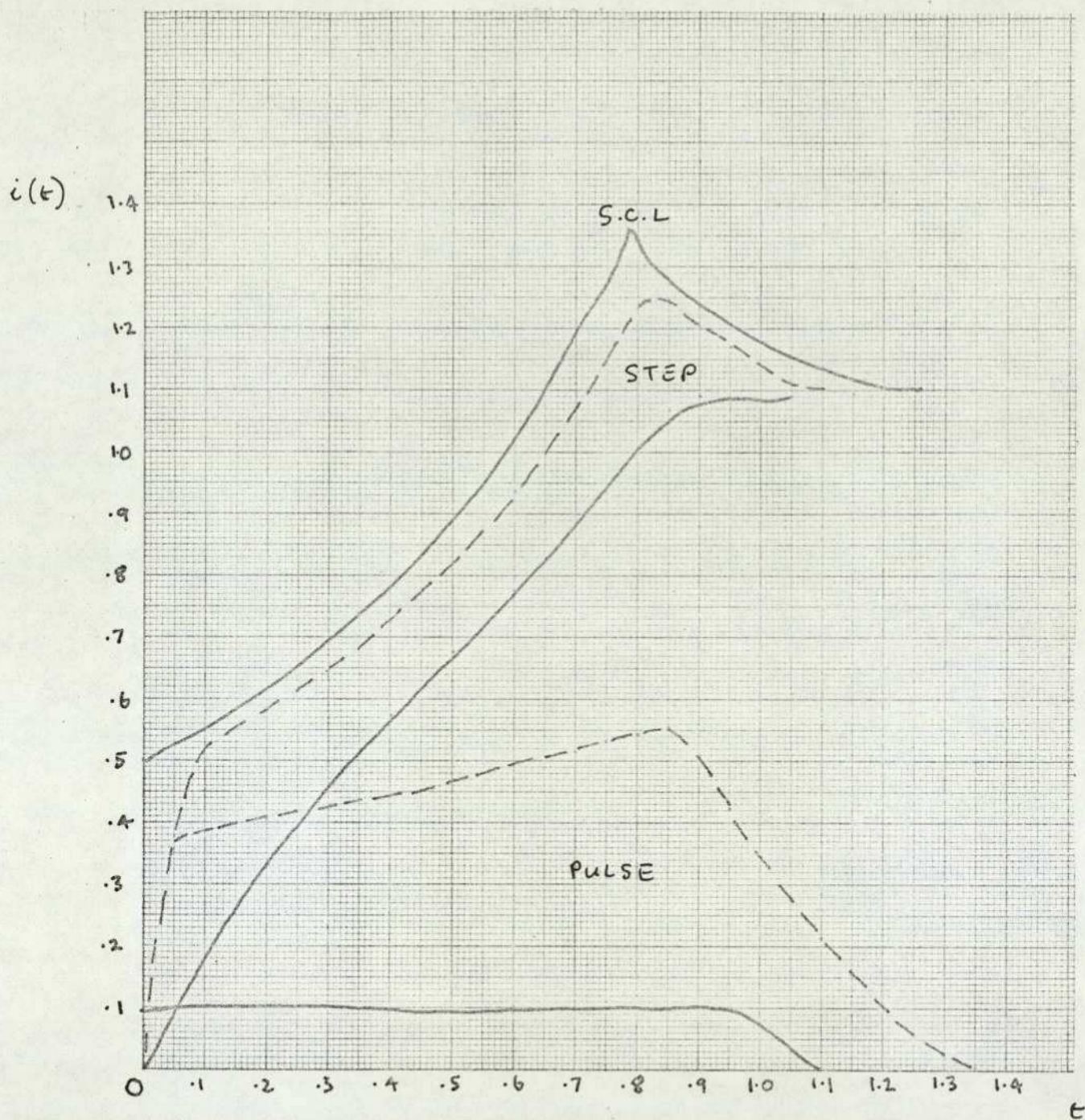


Fig. 4.22 S.C.P. ~~step~~ transients from the model

— .1
 --- .5

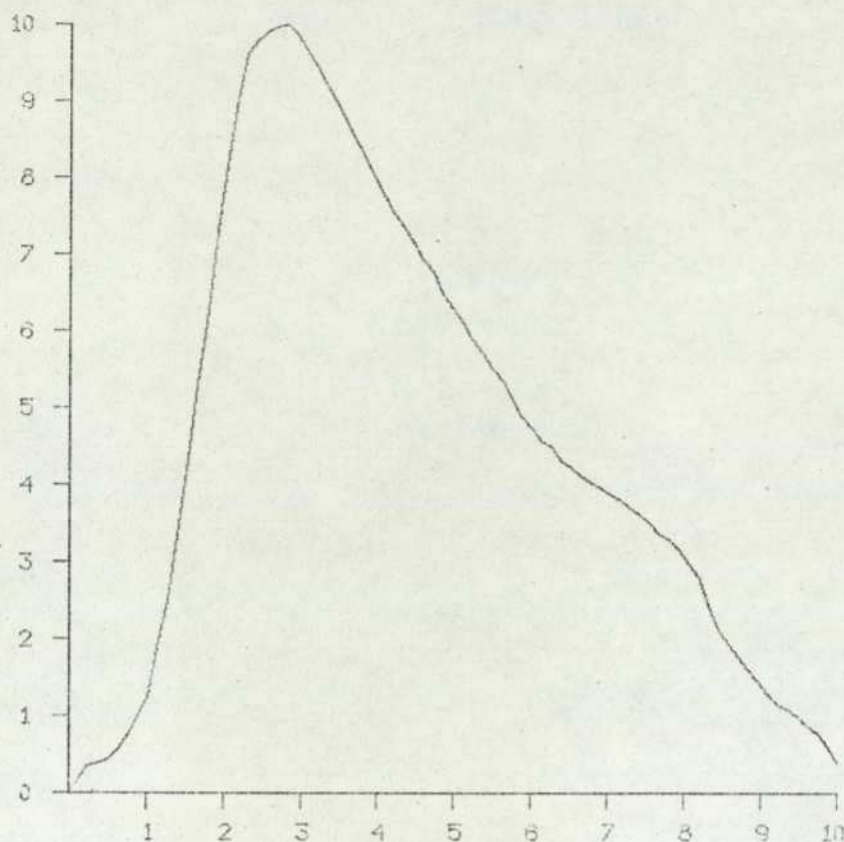


Fig. 4.23 Input current transient

This transient should be the rectangular shape which results from a pulse injection of charge. The actual shape of the transient is attributed by the authors to the fact that the charge carriers have to pass through a very fine metal grid. It is argued that the low interstitial field causes the charge layer to spread out.

This adaptive method was used on this transient (kindly supplied by Hewish) to predict the rate at which charge emerged from the grid.

Fig. 4.24 shows this charge injection curve and the resulting current transient. The charge injection curve exhibits a form of instability. A certain amount of effort was devoted to tampering with the injection rate in the programme, to attempt to damp this oscillation at the expense of the resulting current transient. However all attempts

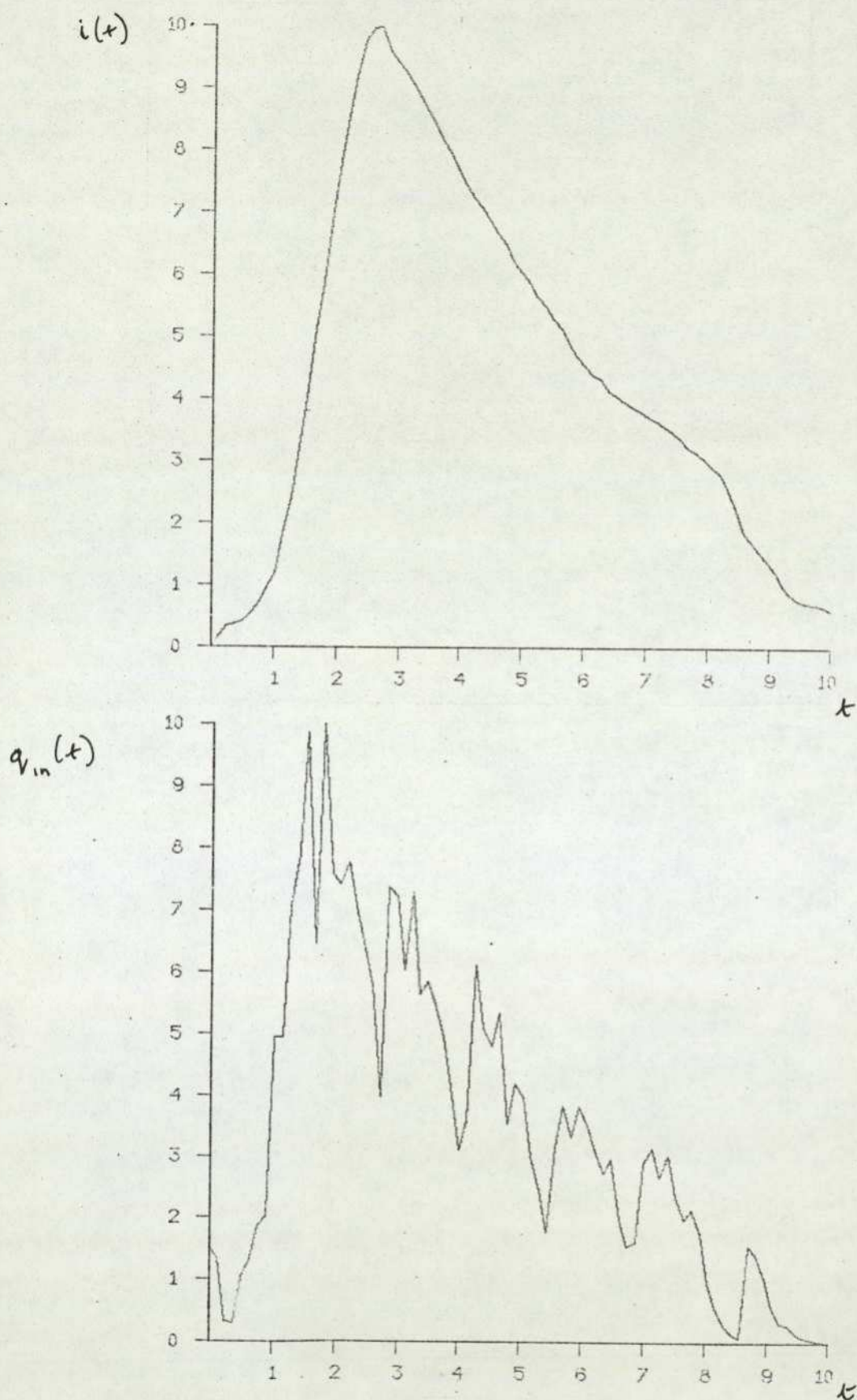


Fig. 4.24 Analysed charge injection rate (for $\mu = 7.5 \times 10^{-8} \text{ m}^2/\text{V}\cdot\text{sec.}$)

failed, so as an alternative the method was run with values of mobility different from the value of $7.5 \times 10^{-8} \text{ m}^2/\text{v.sec.}$, given by Hewish and Brignell.

Fig. 4.25 shows the result for a mobility of $6.75 \times 10^{-8} \text{ m}^2/\text{v.sec.}$. The effect on the instability is very marked. After experimenting with several values the optimum was found to be $9 \times 10^{-8} \text{ m}^2/\text{v.sec.}$, this is shown in fig. 4.26.

Whether these results mean anything in physical terms is hard to say, but they do illustrate a powerful application of the computer model.

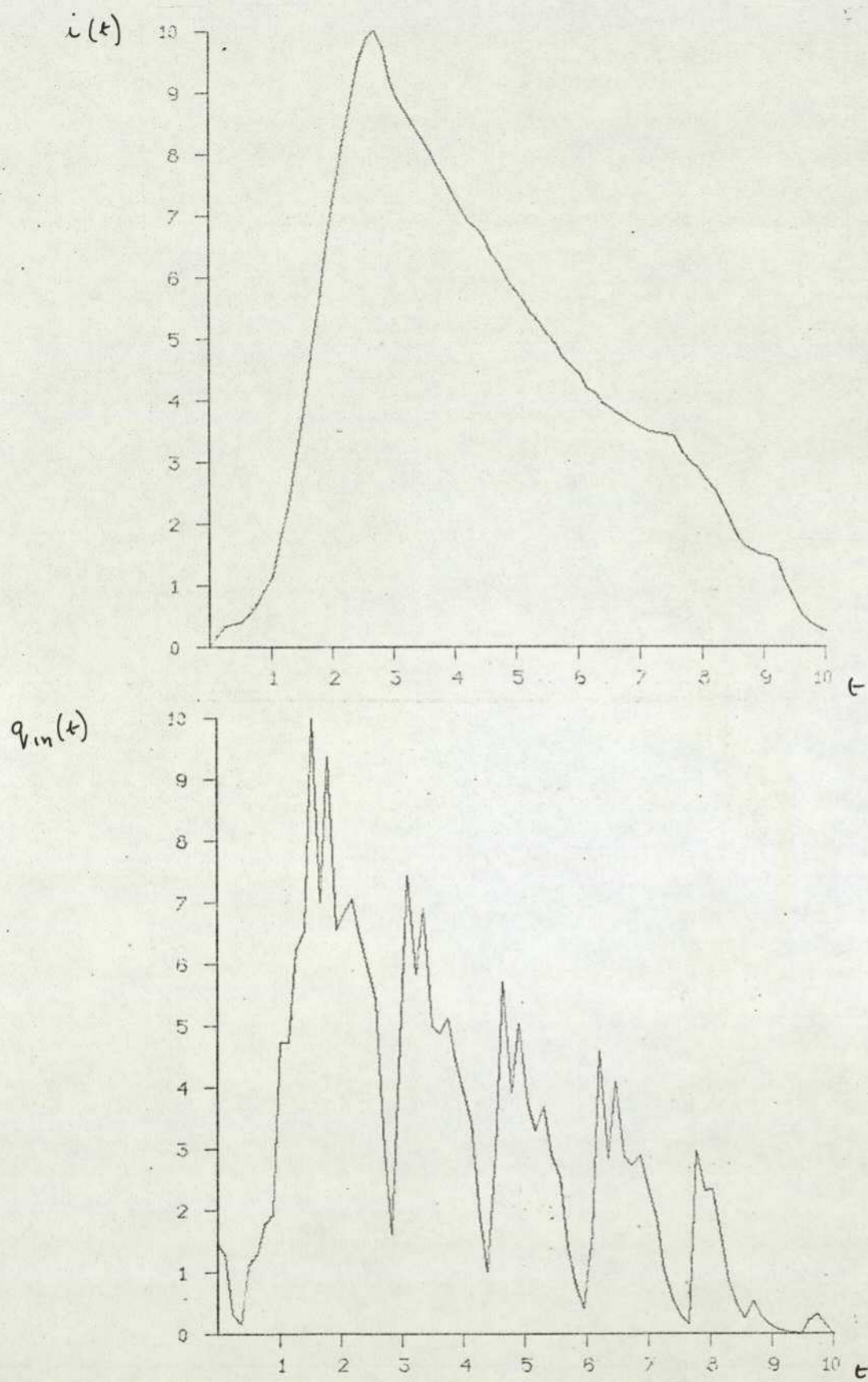


Fig. 4.25 Analysed charge injection rate (for $\mu = 6.75 \times 10^{-8} \text{ m}^2/\text{V.Sec.}$)

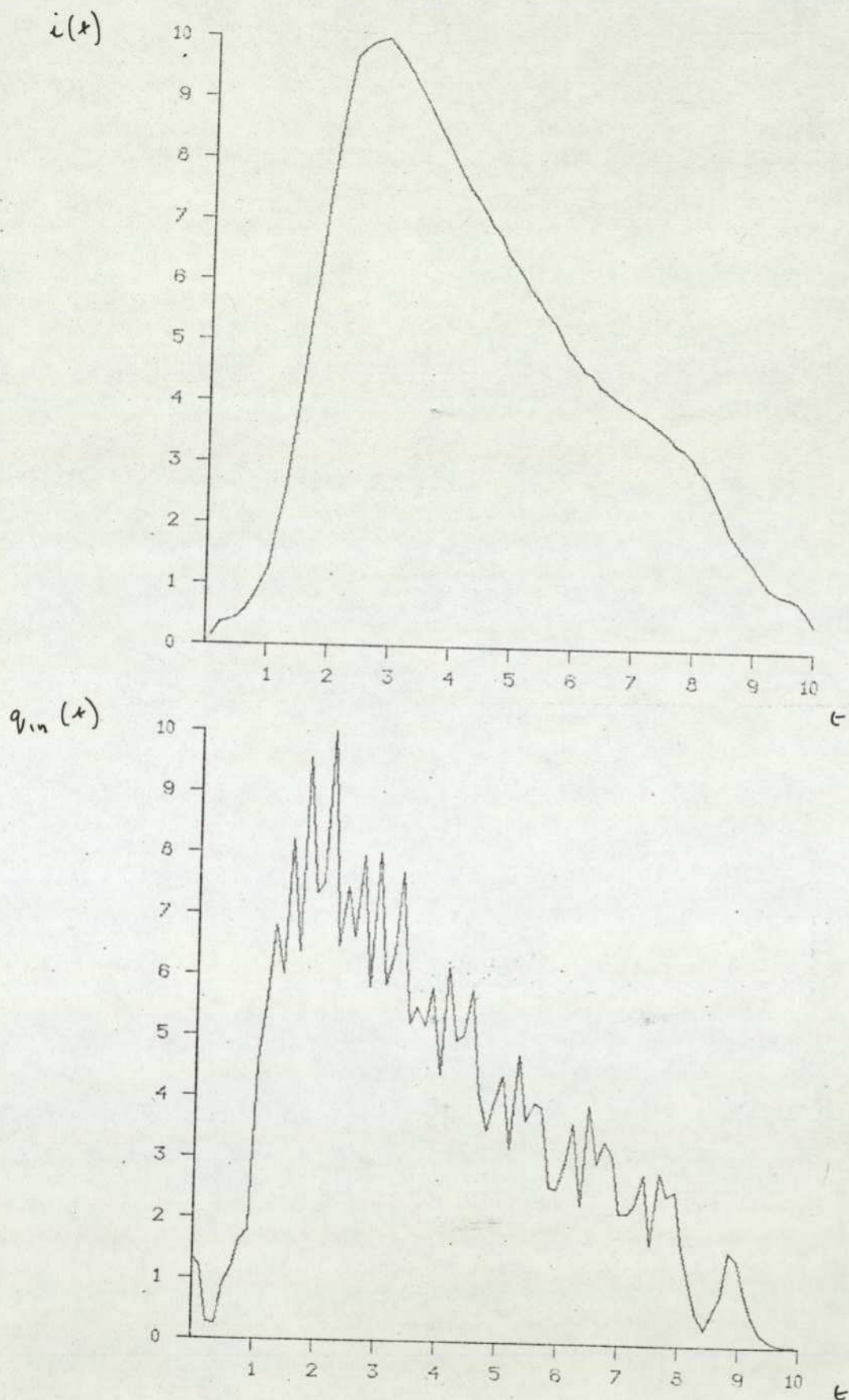


Fig. 4.26 Analysed charge injection rate (for $\mu = 9 \times 10^{-8} \text{ m}^2/\text{V}\cdot\text{sec.}$)

CHAPTER 5

EXPERIMENTAL METHOD

5.1 INTRODUCTION

The object of the experimental work is to measure stimulated current transients in a hexane liquid under a wide range of conditions. This liquid was chosen because it has been commonly used in the past and thus these new results will have continuity with the past measurements.

The "tail" effect in current transients in liquids have been attributed to such possibilities as liquid motion, space charge effects, liquid purity and the generation of carriers at the electrode edges and so on. It is thus important that these new measurements should minimise or eliminate these possibilities.

The major components of the experimental work are U.V. sources, test cell, current measuring apparatus and liquid preparation. In this chapter, these are considered individually, with a lot of attention being paid to design optimisation.

5.2 U.V. SOURCES

The U.V. source was required to generate all possible waveforms from a pulse to a step. It would also be desirable to be able to pulse the light source off, from a continuous on state. It is important for it to have a fast rise and fall time, compared to the transit time of the carriers.

These requirements were met by using a continuous U.V. source with a Compur photographic shutter. This source was a Haneau tube with the optional spectrosil window powered by a 60W Ealing Optics supply. This tube, however, generates a certain amount of electromagnetic mains interference. The effect of this on the more sensitive measurements was minimised by placing the test cell inside a screened room and shining the U.V. through the grilled window.

The shutter had a fastest speed of 1/200 sec., B and T settings and was attached to the front of the U.V. source. The current measurements were so sensitive that any attempt to operate this shutter near the cell by hand, would have caused interference, so use was made of a 20 ft. pneumatic cable lead.

As the amount of light on the fastest speed was rather small, use was made of a xenon flash tube built in a similar fashion to that of Bloor (1970).

Some method was needed for measuring the risetimes and time duration of these sources. This was met by the use of a IPL 16B photodetector which is a small 8 pin I.C. can, containing a light sensitive diode and an integral amplifier. As this has a high output it was coupled by a short level to a μ A7800 op.amp. acting as a $\times 10$ amplifier. This was coupled to a remote control box in a similar way as the picoameter (section 5.8).

This detector only responds to visible light which was fine when used with the flash tube (which emits a wide spectrum of radiation) but gave low output when used with the continuous source. Measurements on the shutter are given below together with the nominal times.

<u>nominal time</u> (m sec)	<u>measured duration</u> (m sec)
5	9
10	13
20	36
40	49
100	102
200	146
500	404
1000	755

The time to open or close the shutter was $200\mu\text{S}$. The time duration of the flash was about $160\mu\text{S}$ and had a similar envelope to Bloor's. This light measuring system was sometimes used to trigger the current measuring apparatus.

At times, the U.V. was filtered by using Chance-Pilkington filters, type OX7 and OX1. The transmission of these filters, both for U.V. and visible light is shown in fig. 5.1 (a) and (b). To give perspective, fig. 5.2 shows the transmission of Koch-light n-hexane before and after purification. It should be noted that $.2\mu$ is equivalent to $2000\text{\AA}$.

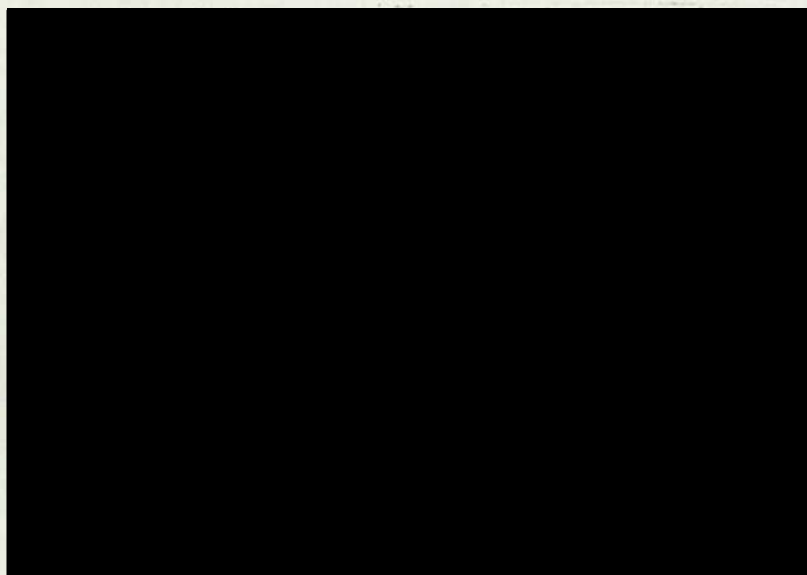


Fig. 5.1 Transmission of filters

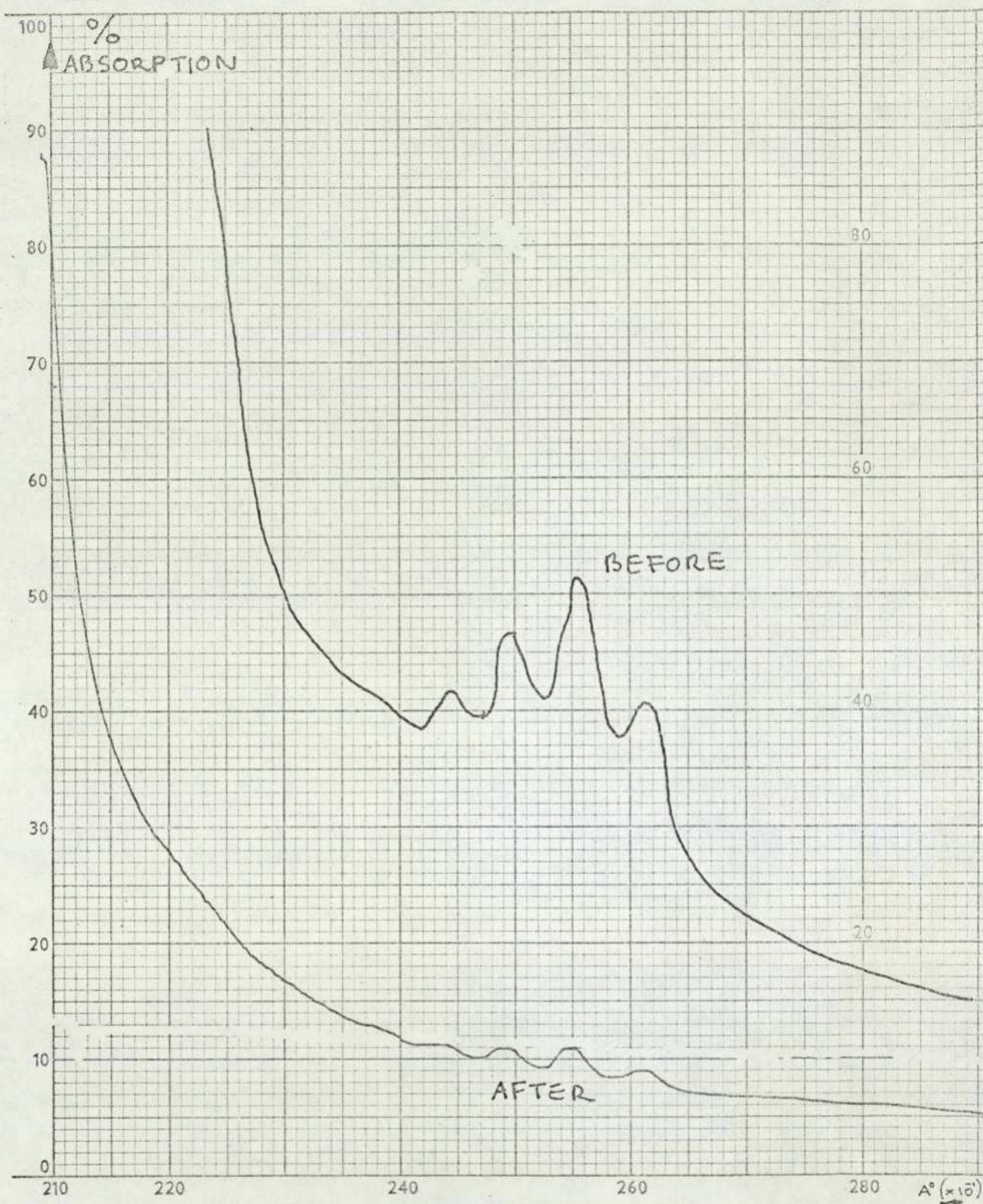


Fig. 5.2 Transmission of n-hexane (before and after treatment).

5.3 TEST CELL DESIGN FACTORS

A lot of thought was given to the design of the test cell because this can have a crucial bearing on the success of any experimental work undertaken in this field. Some considerations are discussed here.

Hexane, the proposed experimental liquid, reacts with most constructional materials thus limiting the choice to glass, PTFE and stainless steel. Glass has the advantage that it is easy to connect to a glass vacuum system, transparent and non conducting. This means that it provides no electrical screening and allows the possibility of charge storage effects (Morant 1968).

PTFE has similar disadvantages and is difficult to machine because of its pliability. This latter feature means that it can be incorporated in a design to act as a vacuum seal, as well as an insulator.

Stainless steel is conductive, provides very good electrical shielding and is comparatively easy to machine. The main setback is the difficulty of providing vacuum seals.

The art of test cell design is to utilise the respective virtues of these materials without incurring their disadvantages. The requirements of the test cell include the following. It should be vacuum tight to 10^{-6} torr (a better vacuum would require a bakable apparatus). It should have a very easily replaceable photocathode with as large a surface area as possible, to improve the overall signal to noise ratio. It would further be desirable to be able to replace this without disturbing more than one vacuum seal, or (more importantly) the electrode geometry. A guard ring is needed to eliminate the possibility of surface leakage currents. Finally, the design should be such that the effects of electrostatic and electromagnetic interference are minimised.

Some of the variation in earlier results has been attributed to the effect of the cell electrode configuration on liquid motion, and that carriers move in non linear regions of field. It thus seemed desirable to aim for an electrode design which approximated to a pair of infinite parallel planes. If subsequent workers were also to aim at this ideal, then this would eliminate one cause of uncertainty, for this should result in a defined hydrodynamic inpedance. It should also be remembered that equ. 2.1 assumes two infinite parallel planes.

Now as the electrode area is reduced, then a point will be reached when the current transient's shape will be affected. An estimate could be made if an analytical expression could be derived which related the flux density D , at the surface of the electrodes, as a function of the distance d from the axis due to a charge q placed on the axis at a distance x . An attempt was made to derive such an expression by the method of images, but without success.

In the absence of this expression an estimate can be made using the relationship for just a simple infinite plane:-

$$D = \frac{q}{2\pi (d^2 + x^2)^{\frac{3}{2}}} \quad 5.1$$

This shows that the flux density falls off rapidly with d .

5.4 DESCRIPTION OF TEST CELL

The test cell is shown in cross section in fig. 5.3. Plate 1 shows an exploded view of it and Plate 2 shows it partially assembled giving a view of the electrodes. Incidentally, there is a feature in Plate 1 which is incorrect and occurred because the author had just received the parts from the workshop, a long time after he had designed it, and had forgotten how it went together. This concerns the P.T.F.E. collar, which in the photograph is on the stainless steel anode and the P.T.F.E. washer on the other side of the main body. These two should be swapped around.

The maximum diameter of the cell was limited to 4" by the available lathes. In spite of this, the ratio of gap to internal cell diameter is about 1 to 8.

The cells consist basically of three parts: the anode and the high voltage cathode, both contained in the heavy earthed body of the cell. The photocathode holder allows the photocathode to be replaced in minutes, without disturbing the electrode geometry.

A 45 mm. diameter, 2.5 mm. thick quartz disc coated with aluminium thin film acts as the photocathode. This is held in place by a 5 mm. lip which provides electrical contact and acts as a liquid seal. The vacuum seal is provided by an O ring on the other side of the disc.

The electrode diameter is 54 mm. and the maximum diameter of electrode illumination is 35 mm., an area of 11.8 cm^2 . The photocathode retaining lip causes the field to be slightly greater at the edges of the electrode system but this is well away from the volume containing charge carriers.

A region of high stress has been incorporated into the design, between the photocathode holder and the main body of the cell. The concept is that liquid particles will preferentially accumulate in this higher stress region, away from the electrode gap.

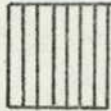
● P.T.F.E O RING



STAINLESS STEEL



P.T.F.E



QUARTZ

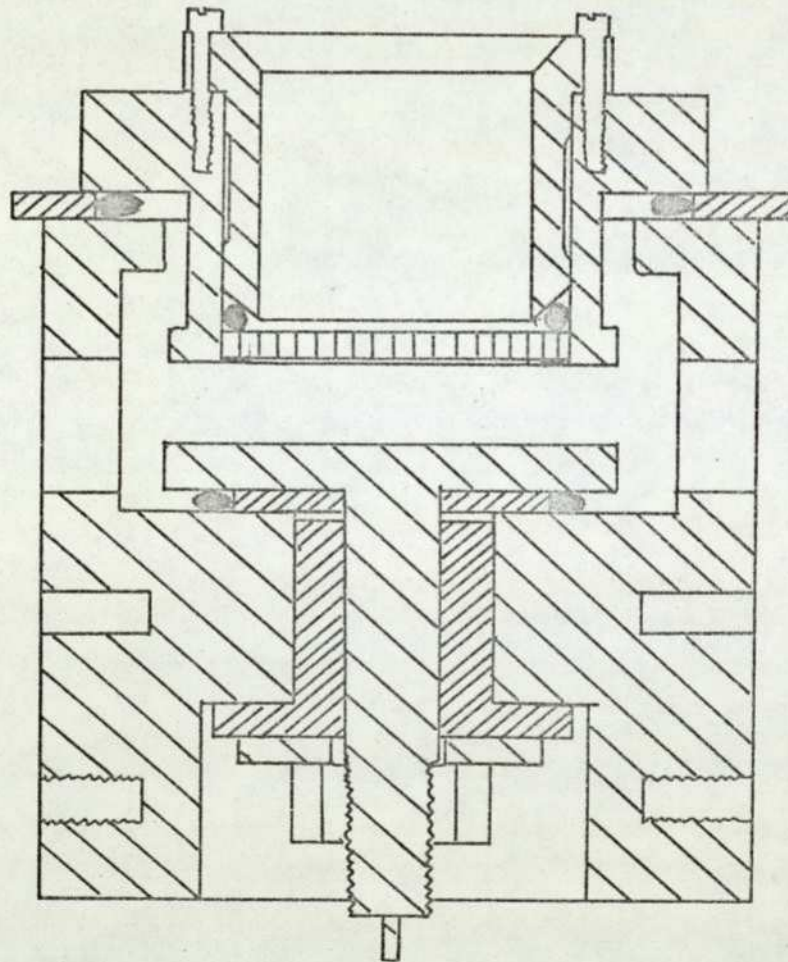
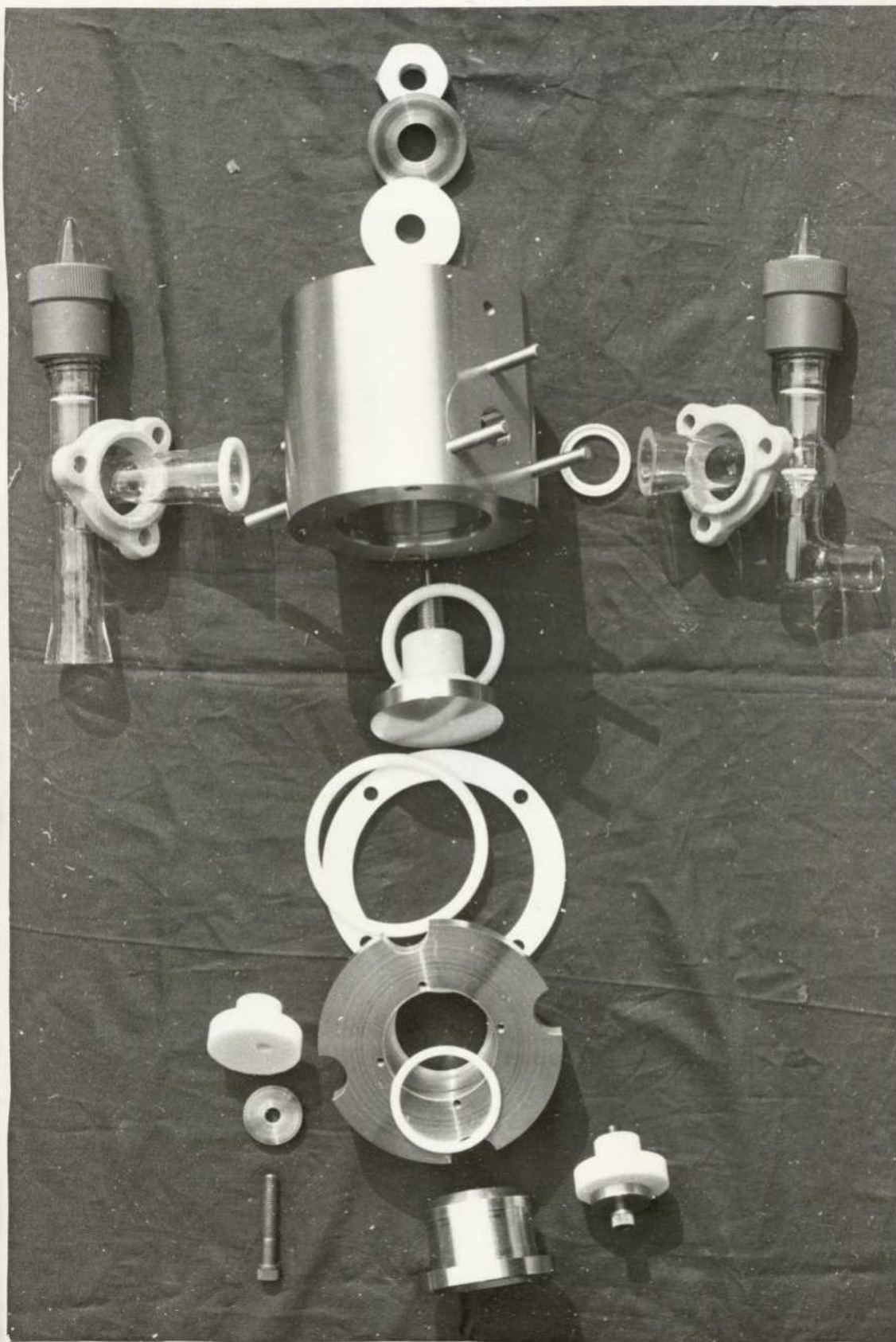
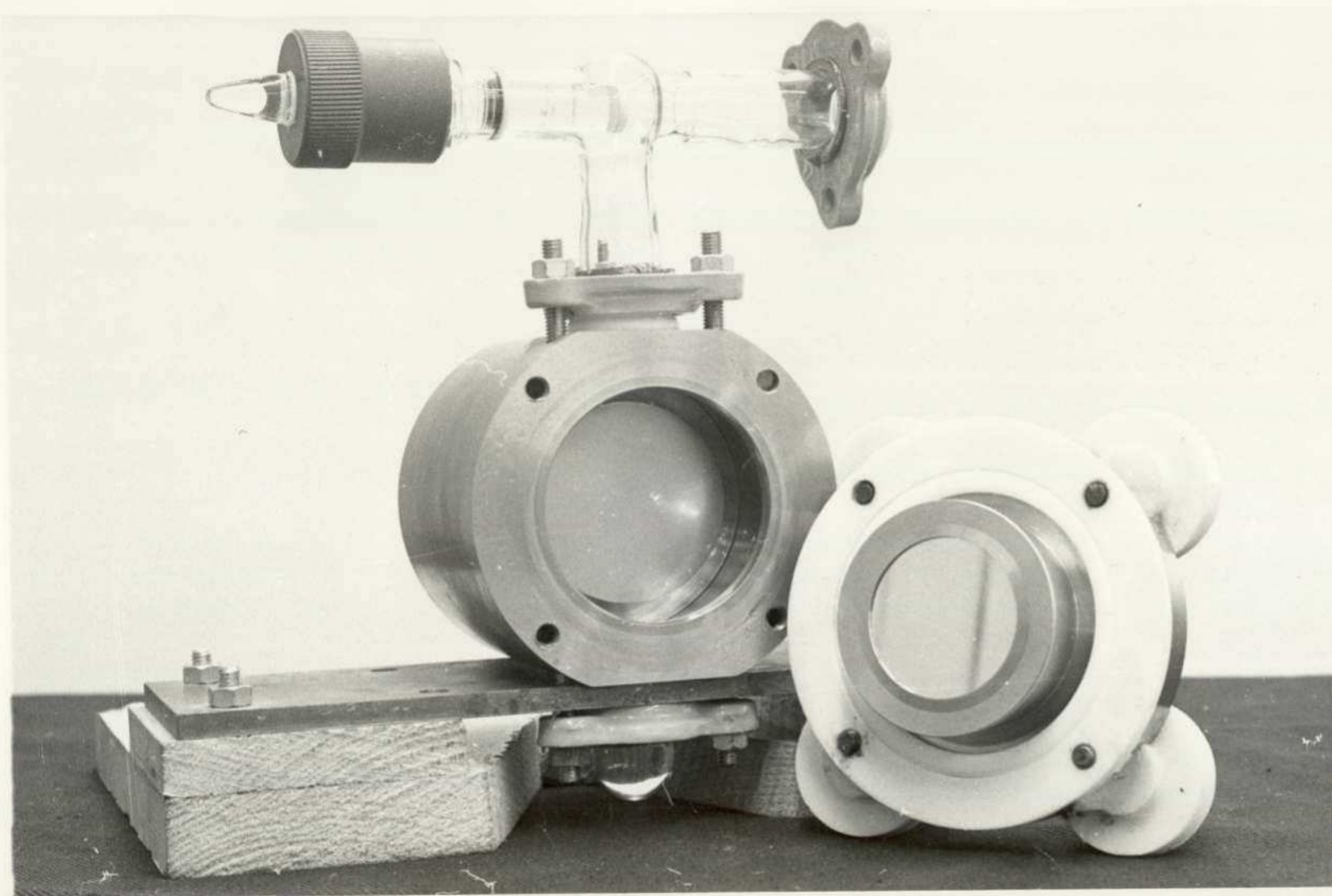


Fig. 5.3 Cross section of test cell.





The vacuum connection from glass to metal was made with a standard Q.V.F. P.T.F.E. joint, a novel method to this field of study. Two apertures were provided in the cell body order to allow the use of Schlieren techniques. They will also allow an assessment of possible bulk generation of carriers.

5.5 PHOTOCATHODE FILM DEPOSITION

The quartz discs were held four at a time for deposition in a thin film unit. This allows deposition to a measured thickness under high vacuum.

Swan (1961) suggested the technique of back illumination through the glass, rather than from the liquid side. He argued that if the film was sufficiently thick, then photo injection would occur without appreciable irradiation of the liquid. This idea has been accepted by several subsequent workers but without validation. The following research however, throws light on this theory.

Lea and Mee (1968) gave the relationship between the photoelectric current S_{i_1} , due to a film thickness S_x at a depth of x in the film as

$$S_{i_1} \propto I_1 \cdot e^{-\alpha x} \cdot e^{\left(\frac{-x}{L}\right)} \cdot S_x \quad 5.2$$

where I_1 is the intensity of the incident light

α the absorption coefficient of the incident light

L the attenuation length of the electron

Fig. 5.4 illustrates some of these variables.

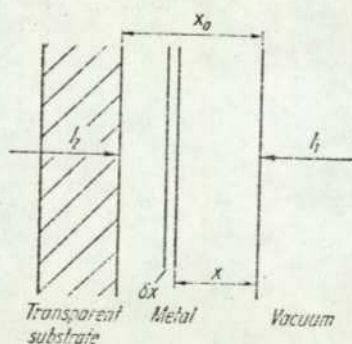


Fig. 5.4 Illustration of variables (after Lea and Mee).

Integrating this equation gives

$$i_1 = \int_0^{x_0} I_1 \cdot \exp(-\alpha x) \cdot \exp\left(-\frac{x}{L}\right) \cdot dx \quad 5.3$$

The radiation which passes through the film without absorption

is

$$I_1 \exp(-\alpha x_0)$$

The problem of determining these constants was solved by comparing the photoelectric currents, for the same thickness film, when illuminated from either side (i.e. vacuum and substrate side).

The simplified relationship is

$$\ln \frac{i_1}{i_2} = \alpha (x_0 - 2L) \quad 5.5$$

where i_1 is current by illumination from the vacuum side

i_2 is current by illumination from the substrate side

So a plot of $\ln \frac{i_1}{i_2}$ against x_0 will give the values for α and L . This was done for a uranium film (fig. 5.5) and the values for radiation of $2483^\circ \text{\AA}$ were found to be

$$\alpha = \frac{1}{128^\circ \text{\AA}}$$

and

$$L = 65^\circ \text{\AA}$$

Fig. 5.6 the relationship of these currents against film thickness

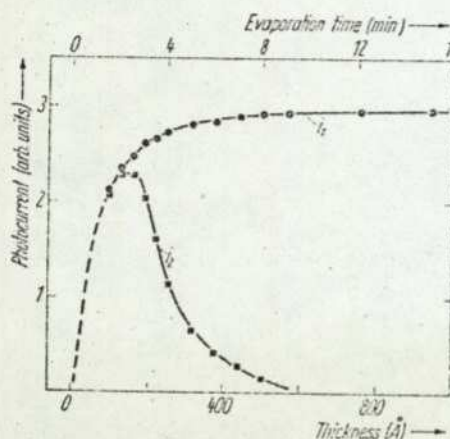


Fig. 5.5

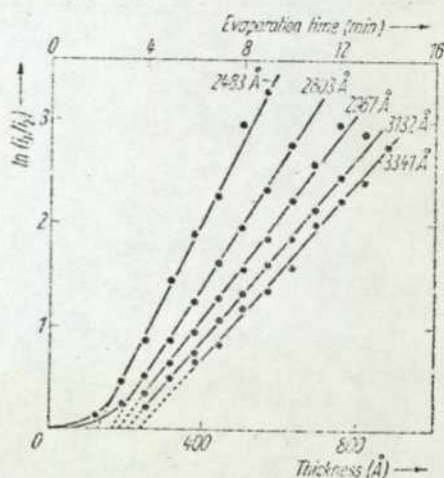


Fig. 5.6

The authors also monitored the electrical resistance of the film during deposition. They found that it reduced in proportion to the log. of the thickness; at a 100°A , it was found to be 400Ω .

To relate these measurements to aluminium films, it is necessary to consult the paper of Batt and Mee (1969) who measured the photoelectric yield of a very pure aluminium film when illuminated from the vacuum side. They found a maximum current at 50°A . But this is related to its purity, for Garron (1965), using a less pure film produced a figure of 520°A .

Batt and Mee also examined electron micrographs of the films which revealed that even a carefully deposited film had a rippled surface similar to a group of islands in water.

To summarise, these papers reveal that the photoelectric effect is a bulk phenomena and imply that it may be possible to find an optimum film thickness.

In this work, thickness from 50°A to 300°A were used, with the maximum current obtained for 150°A . It was also found that with reversed polarity on the cell, photoinjection occurred from the stainless steel electrode. The ratio of normal to reversed currents was of the order of 3, seemingly independent of film thickness.

The design of the disc holder allowed the centre to be masked, allowing the deposition of an extra thick anular ring. This was to provide good electrical contact. However, the research cited in this section suggests that this is unnecessary and it was dispensed with at times with no apparent effect.

5.6 LIQUID PREPARATION

5.6.1 Introduction

Impurities such as unsaturates, benzene, water and dissolved gases must be removed from the test liquid, n hexane, before use in an experiment.

If this purification was sufficiently good, then the measured charge transport would be characteristic of the real liquid, rather than of the impurities. To achieve this would require rather complicated and difficult methods and would result in the fast carriers discussed in section 3.2.

However, most of the experimental work in the past has used intermediary methods of purification resulting in transport of an ionic nature. How as this project is as much a study of method of current transient analysis, as an investigation of liquid insulants, such methods were adopted.

To assess the effects of purification on the resulting transient shape; various improvements were carried to the preparation method during the duration of the experimental work.

5.6.2 Purification

Purification was carried out by the use of activated silica gel and a molecular sieve. According to Potts (1952) the silica gel should remove purities, (such as benzene) which would cause U.V. absorption over the range of $2000\text{\AA}$ to $2500\text{\AA}$ (fig. 5.2).

The molecular sieve, (Union carbide type 4A), is so called because the material, a silicate (or zeolite), is riddled with molecular sized holes, rather like a sponge. For the sieve used, these have a dimension of about $4\text{\AA}$ diameter. The sieve action occurs because of the tendency of molecules smaller than this (such as water) to accumulate in these holes, whereas larger molecules such as hexane will not.

This effect is even stronger for polar molecules, such as water.

It is claimed by B.D.H. that a molecular sieve has a better performance at lower impurity concentrations than other methods and this is why the silica gel treatment is followed by this method.

These materials were contained in two 20" long by 2" diameter pyrex tubes, mounted on top of one another. Each contained had a 1 μ glass filter which also kept the contents in place.

Several authors have suggested the need for freshly activated silica gel. This can be achieved by using freshly crushed coarse crystals or by heating the material in an oven, preferably under vacuum. But since the saturation content of water in hexane is of the order of 100 p.p.m. freshly prepared gel can purify a lot of hexane before saturating. As it is no easy matter to replenish these materials, they were used for several purification cycles.

The resulting purified liquid was tested for U.V. absorption in an Hitachi Perkin Elmer Spectrophotometer (APS 3T). The U.V. curve (fig. 5.2) was very similar to those published by Bloor (1970), Rahman (1970) and Buttle (1969).

Once, after changing the materials, it was found that the conductivity of the liquid was inferior to the usual value and was not steady. The measured current transients were indicative of bulk ionisation and when the liquid was analysed, it was found to have very poor U.V. transmittance. A possible explanation is that very fine dust from these replenished purification materials found its way through to the cell.

Because of this, it seemed sensible to adopt the common procedure of purging the materials in situ by heating under vacuum. This was achieved by using Electrothermal heating tapes, which were wrapped around the outside of the pyrex tubes. The system would then be heated for

up to 12 hours while the top, or input, end of the system was connected, via a cold trap, to a backing pump.

Fig. 5.7 shows a schematic view of the system before modification was undertaken.

5.6.3 Liquid degassing method

Initially the degassing system was exactly as described by Rhodes (1971). Plate 3 shows it in the middle of a degassing cycle.

Although the system worked reasonably well, it was felt that some improvements were necessary. The first of these concerns the pressure at various points in system, compared to that indicated by the Penning gauge.

Now the lowest pressure which can be achieved is effectively determined by the worst conductance in the system. Hence there is no point in going for a diffusion pump with an enormous speed unless the diameter of the glassware is sufficiently large to make use of it.

The conductance of an orifice is proportional to the square of its diameter, and for a long tube to the cube of the diameter. Now the degassing system as it stood used a $\frac{1}{2}$ " diameter tube to evacuate the test cell. A calculation showed that the pressure in the cell would be an order of magnitude lower than that indicated on the system's penning gauge. This was measured by replacing the test cell by another gauge and was found that the ratio was in fact 30 times. So clearly this needed improvement.

A second problem was that of controlling the flow of evaporated hexane from the distillation flash to the degassing spheres. It is desirable to be able to halt the flow leaving a small amount of hexane in the flask and also, it is hoped, the heavier impurities and dust. To achieve this in the system as it was required critical timing of the cessation of replenishment of liquid nitrogen in the degassing spheres. If this went wrong, it was possible to end up with

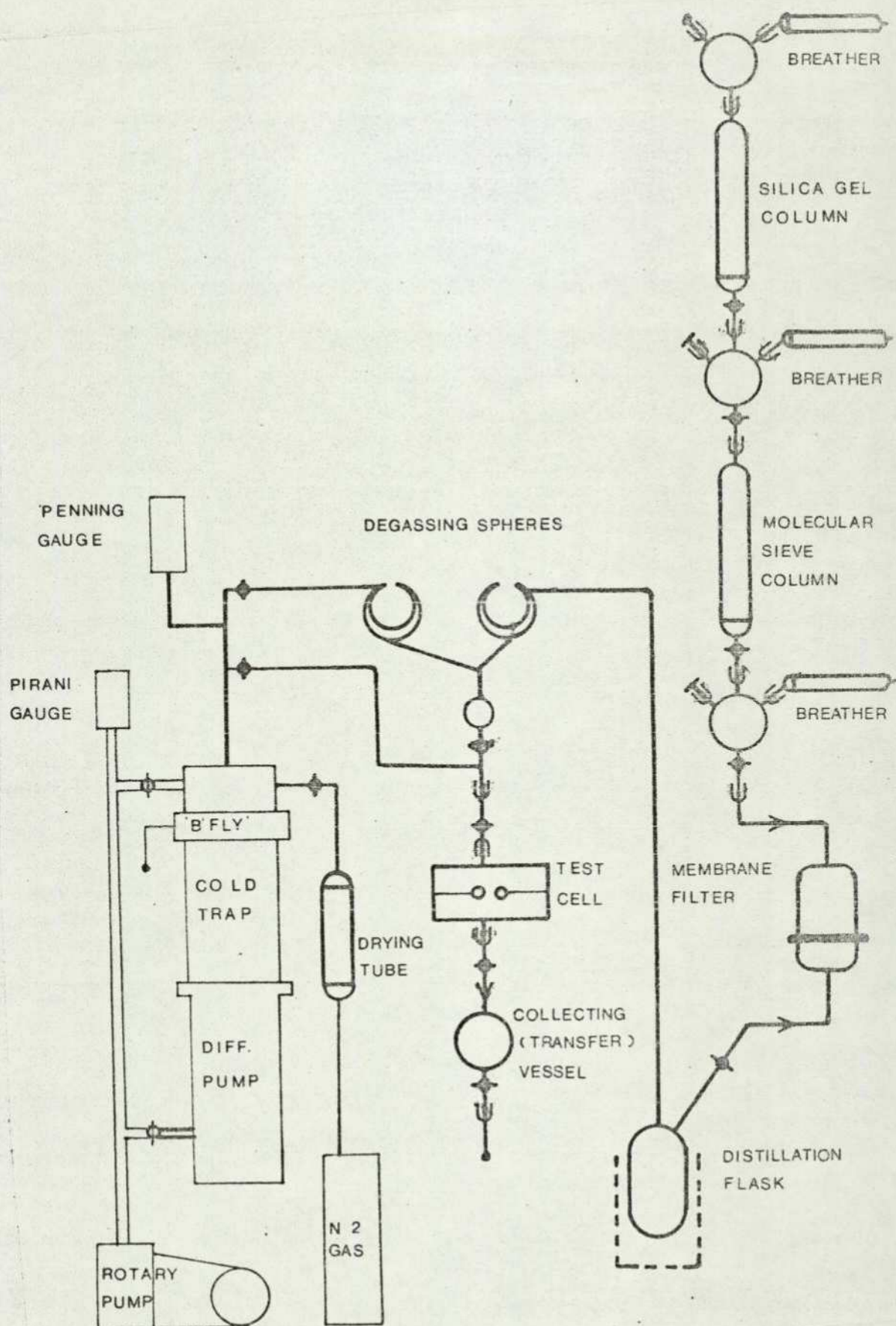
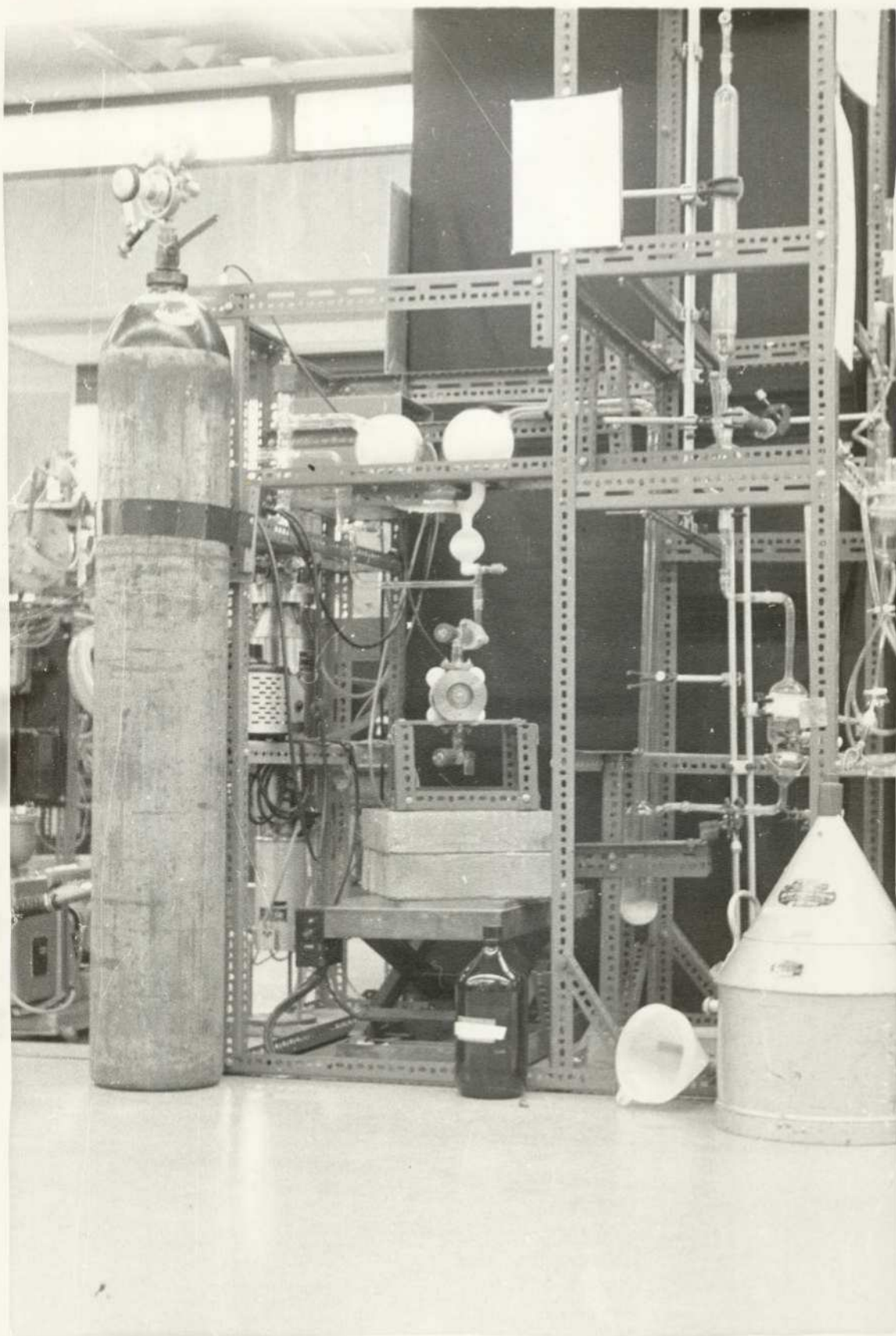


Fig. 5.7 (after Rhodes)



all the hexane out of the flask, or alternatively, with it all back in.

It was impossible to remove this end flask for cleaning. The solution to these two problems was to separate the degassing system from the purification system and use a flask with an integral valve as the transfer vessel. Hence this tap could control the distillation, and the flask could easily be cleaned. Where possible the "Young's" pushfit type joints were replaced by the more positive and safer Quickfit joints.

The diffusion pump had been using Edwards type 705 oil but this was replaced by the superior "Santovac 5" oil. This oil is intended for use in "clean systems" and has a very much lower vapour pressure than earlier oils.

In view of these charges, it is convenient to give here, a full description of the operation of the new system. The first step is to connect the transfer flask to the bottom of the purification column, evacuate it, and then fill it with purified hexane. The flask is then sealed and is transferred to the degassing system which is then brought down to 10^{-6} torr. The hexane is then frozen by the application of liquid nitrogen, in a dewar, around the flask.

The tap is then opened, removing residual gases. A number of these freeze-thaw cycles are carried out before the hexane is allowed to slowly evaporate upto the top spheres.

After sufficient hexane had been so collected, the transfer was stopped by closing the end flask's valve. At this time the top spheres were closed off from the rest of the system and the hexane was allowed to warm up to room temperature. While this happened, the test cell was kept evacuated by the auxilliary vacuum path (now of larger diameter). Finally, the entire system was shut off from the pumps and the hexane was allowed to enter the cell under gravity.

At this point two methods were possible. The first was simply to seal off the cell, by its integral valve, at the vapour pressure of hexane. Alternatively, the pressure could be raised slightly higher than atmospheric, by the use of oxygen free nitrogen from a compressed gas cylinder. As there was a length of hexane in the tubing above the tap, there was no possibility that this would diffuse into the cell.

This pressure differential would prevent air from entering the cell. But as the vapour pressure of hexane is quite high, it is perhaps unnecessary and in practise, no difference could be detected between the methods.

5.7 PICOAMMETER DESIGN TECHNIQUES

5.7.1 Introduction

From a study of earlier work, the current levels to be measured were not expected to be greater than about 100pA. It is desirable to be able to measure currents at the lowest possible levels in order to detect any possible liquid motion and to determine if the transients change with current magnitude.

The design of picoammeters involve a compromise between sensitivity, noise and bandwidth. Now in order to achieve sufficient sensitivity, past experimentors have often resorted to integrating the currents in order to overcome noise problems. This technique is unfortunately similarly effective in making interpretation difficult. If the transients were of the predicted shape, this would suffice, but section 3.8 shows this not to be so.

As several experimental studies have been marred by the poor current measuring techniques, it seems worthwhile to do a thorough study of the constraints governing this subject. This will reveal several useful techniques, some of which are unavailable on commercial equipment.

5.7.2 Risetime

The bandwidth of picoammeters can be more suitably expressed as a risetime for this application. This is defined as the time taken, t_r , for the output to change from 10% to 90% of its final value, in response to a step current input. Another alternative is to express the speed in terms of the time constant of an equivalent CR network. An approximate expression relating these is given by Keithley (1972)

$$t_r = \frac{0.35}{f_{3dB}} = 2.2 CR \quad 5.4$$

where f_{3dB} is the 3dB bandwidth.

The straight forward method of measuring a current is to pass it through a sufficiently large resistor and monitor the voltage drop. This is known as the shunt method and requires a high input impedance voltage follower if the subsequent circuitry is not to load the resistor. The major problem concerns the capacitance present at the input, which forms a CR circuit. So for a sensitivity of 1V/pA, i.e. a $10^{12} \Omega$ resistor and stray capacitance of 10pF, the time constant would be 10 sec.

Another approach is to monitor this voltage drop with a vibrating capacitor electrometer. This can have a sensitivity of 10mV with a $10^{16} \Omega$ input impedance but sadly the instrument itself exhibits a risetime of about a second.

The alternative is to use the "feedback" method. This, as the name suggests, places the high value resistor in the feedback loop of a high gain amplifier. This is illustrated in fig. 5.8.

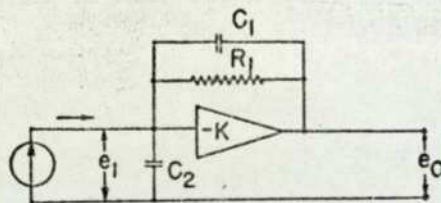


Fig. 5.8 Circuit using feedback (after Presley)

C_2 the total effective capacitance across the input, and C_1 is self capacitance of the resistor R_1 . The amplifier, gain K will need to be of the "electrometer" class - namely having very small input bias currents so as not to swamp the current being measured.

The output voltage e_o , expressed as a function of time, in response to step current input of magnitude i , has been shown to be (Praglin and Nichols 1960)

$$e_o = - \frac{i R_1}{1 + \frac{1}{k}} \left[1 - \exp \frac{-t}{R_1 \left\{ \frac{C_2}{1+k} + C_1 \right\}} \right] \quad 5.5$$

Hence, for a sufficiently large k , the time constant is now unaffected by the input capacitance and is only dependent on the self capacitance of the resistor. Typically, this would be 1pF, giving an improvement in the time constant over the previous example from 10 sec. to 1 sec.

This self capacitance can be minimised by the inclusion, in the feedback loop, of a compensation circuit as shown in fig. 5.9.

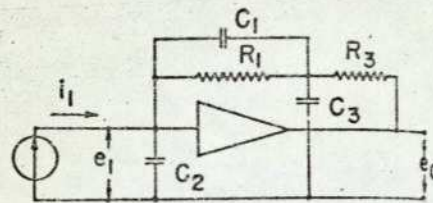


Fig. 5.9 Circuit using compensated feedback (after Presley)

When this CR network has been adjusted so that

$$C_3 R_3 = C_1 R_1$$

then the new effective time constant has been shown by Praglin and Nichols to be

$$\frac{R_1 \cdot (C_1 + C_2)}{k+1}$$

This implies that the response speed would now be virtually unlimited. Sadly, the above analyses assume that the resistor's self capacitance can be lumped together as C_1 , but in practice it

is distributed, thus the component acts as a delay line.

The final improvement is to minimise this distributed capacitance. One approach by Kendal and Zabielski (1970) is to apply feedback to the middle of the resistor, thus making use of the distributed capacitance. With optimum adjustment, a fivefold improvement in risetime is claimed.

An alternative and perhaps sounder approach, has been given by Presley (1966). This method establishes an electric field to match the voltage gradient along the resistor. This is illustrated in fig. 5.10.

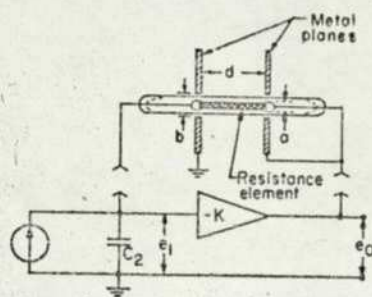


Fig. 5.10 Presley's method of increasing risetime.

The concept is to eliminate along the resistor, any stray capacitance to earth. In practice, this is achieved by mounting the resistor between two large metal plates. One is at earth potential (to match the virtual earth input), and the other is driven by the output of the amplifier.

5.7.3 Noise

There are three major sources of noise:

- (a) Johnson noise current i_j , in the feedback resistor.
- (b) Amplifier current noise i_a
- (c) " voltage " e_a

The R.M.S. Johnson noise current is given by

$$i_n = \sqrt{\frac{4kT\Delta f}{R}}$$

where k is Boltzman's constant

T is the absolute temperature

Δf is the bandwidth

and R is the resistance.

This expression assumes that the picoammeter's bandwidth has a sharp cut off, but in practice it will have a 6dB/octave roll off. Also the resistors used never reach their theoretical noise performance, so an empirical expression often used (Weinberger 1971) is

$$i_n = \frac{\pi \times 2.2 \times}{2} \sqrt{\frac{4kT \times 3.5}{R \tau_r}}$$

This expression, for a 10^{10} resistor and a risetime of 1ms gives a noise current of about 10^{-13} A.

For current transient analysis, the R.M.S. value of the noise is of less interest than the peak to peak value. It can be shown that the magnitude of the noise is less than $2\frac{1}{2}$ times the R.M.S. value for 99% of the time, so the following relationship is used

$$I_{pp} = 5 I_{rms} \quad 5.10$$

The amplifier current noise depends on the particular amplifier and the overall risetime, but for modern electrometers, it would typically be 10^{-15} A, and can hence be ignored.

The amplifier voltage noise unlike the other sources of noise, is unlimited by the overall bandwidth and depends on the amplifier's open loop bandwidth. This noise voltage can be expressed as an equivalent noise current, when it is referred to the input. A typical noise spectrum is shown in fig. 5.11.

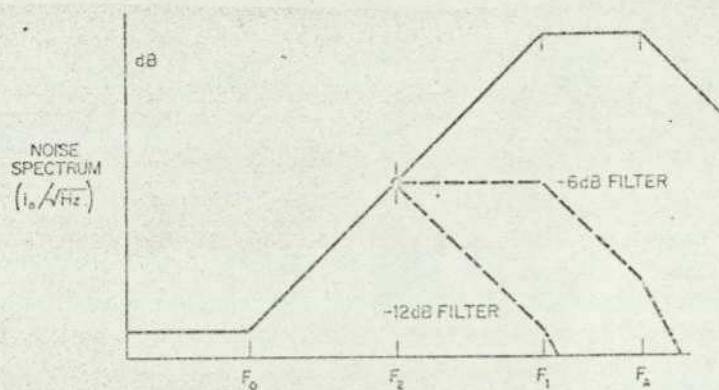


Fig. 5.11 Effect of post filtering on noise spectrum (after Cath and Peabody)

Here F_0 represents the bandwidth of the feedback resistor and the input capacitance, F_1 that of the overall picoammeter and F_A that of the electrometer.

Now the total noise is given by the area under the curve and this noise spectrum raises several points. The first is that the spectrum is flat upto the limits of the electrometer, typically 1MHz. Hence a filter is required on the output to limit this and its effects are shown for 2 different filters acting at a frequency F_2 . It can be seen that a 6dB/octave filter is insufficient and that a 12dB/octave filter is essential.

Fig. 5.12 shows the effect of an increase of input capacitance.

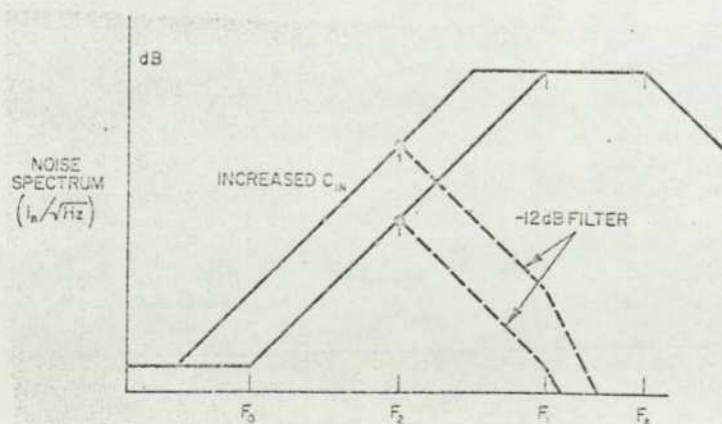


Fig. 5.12 Effect of input capacitance on the noise spectrum (after Cath and Peabody).

This shows the importance of minimising this capacitance, even though it has no effect on the risetime of the amplifier in the feedback mode.

Other sources of noise include electrostatic interference and microphonic effects caused by changes in the input capacitance. The remedy is to have good shielding and to make all input connections rigid.

5.7.4 Sensitivity

For a given value of feedback resistance, and hence risetime, it is possible to gain an increase in sensitivity at the expense of the signal to noise ratio. This can be achieved by the use of post amplification or by a potential divider in the feedback loop, shown in fig. 5.13.

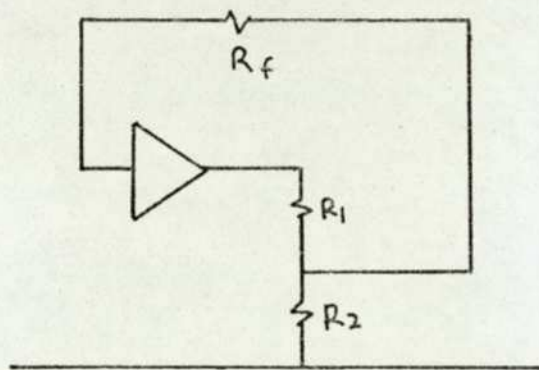


Fig. 5.13 Circuit

A simple analysis will show that the current to voltage gain has been increased by a factor of

$$\frac{R_1 + R_2}{R_2}$$

5.7.5 Risetime measurement

To measure the risetime of a picoammeter it is necessary to apply a step current of very low magnitude. This cannot be simply achieved by applying a step voltage across a large resistor because the time constant of the resistor and its self capacitance would be

in excess of the amplifier. Praglin and Nichols (1960) suggest the circuit shown in fig. 5.14.

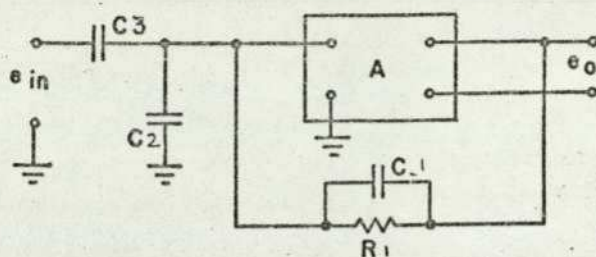


Fig. 5.14 Circuit for measuring risetime (after Praglin and Nichols)

The method involves applying a triangular wave through a small capacitor C_3 (typically 5pF) to the amplifier. Essentially, this triangular voltage wave is applied to a series CR network, producing a square wave current. In this case the resistor is the input impedance of the amplifier, which in turn is a function of the feedback resistor.

For the circuit shown, the output voltage is (Praglin and Nichols)

$$e_o = - \frac{a R C k}{k+1} \left[1 - \exp \left\{ \frac{-t}{R \left[\frac{C_3}{k+1} + \frac{C_2}{k+1} + C_1 \right]} \right\} \right]$$

Thus for a high gain amplifier, the risetime of the output is the true amplifier response and the method is unaffected by the input capacitance.

5.7.6 Performance figures

Figures for the performance of various picoammeters are given here. The Presley design is perhaps the best available at present and the figures represent the best that can be achieved. The Keithley model 417 is a high speed picoammeter available at a cost of £540 and the model 427 is a current amplifier at £410.

The calculated noise figure is the Johnson noise. It can be deduced that this is the main source of noise in the amplifiers, when the bandwidth does not extend to the mains frequency. The slow response figures for the 427 can be obtained with its adjustable 12dB/octave filter which gives an increase of signal noise at the expense of risetime.

<u>R</u>	<u>Presley</u>	<u>417</u>	<u>427</u>		FAST		SLOW	
					noise (pA)	calculated (pA)	noise (pA)	calculated (pA)
10^{14}	500mS	-	-	-	-	-	-	-
10^{13}	50mS	500mS	-	-	-	-	-	-
10^{12}	5mS	30mS	-	-	-	-	-	-
10^{11}	800mS	5mS	-	-	-	-	-	-
10^{10}	200uS	-	1.5mS	.4	.06	330mS	.003	.004
10^9	100uS	-	400uS	2	.4	100mS	.02	.02
10^8	-	-	220uS	10	1.7	10mS	2	.2
10^7	-	-	60 S	50	10	1mS	2	2

5.8 MEASURING APPARATUS

5.8.1 Introduction

The previous section outlined the techniques of low current measurement. It indicated that any design necessitates a compromise between speed, noise and sensitivity. For this application, it is desirable to be able to adjust these parameters at will. For example, if a ramp current is being measured, signal to noise is more important than risetime. The system will also have to cope with transit times which can be upto a 100 times the longest one.

Another problem are the standing currents, in the liquid, due to its conductivity. A method has to be provided for offsetting these to perform the more sensitive measurements.

A block diagram showing a solution to these problems is in fig. 5.15

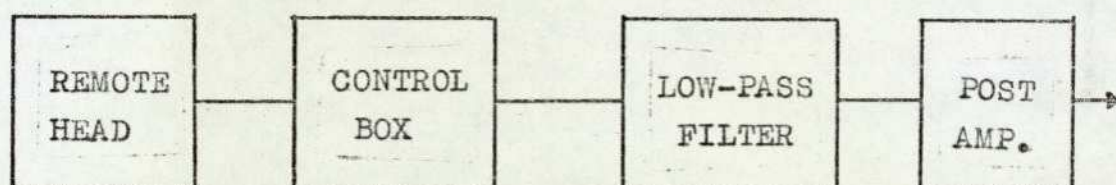


Fig. 5.15 Diagram of measuring apparatus

5.8.2 Picoammeter head

Initial experiments were conducted with an Analog Devices 310 electrometer amplifier, which had been used previously by Buttle (1969). This utilises the varactor bridge principle and features a 10^{-15} A input bias current. Measurements using it showed that if its risetime was speeded up to less than a 100mS, it was prone to spurious noise.

All attempts at shielding could not prevent this and it was subsequently found that such devices are not recommended for operation with a bandwidth greater than 10 Hz .

The alternative electrometer used was an Ancom 15-7c; chosen for its differential input capability, which is was thought might prove useful. This electrometer is specially made for the application of fast current pulses. It features a maximum input bias current of $.1\mu\text{A}$, which doubles for every 10°C increase in temperature.

Finding suitable resistors for the Presley method was a problem. This requires resistors, whose resistance are uniform along their length. Most high megohm resistors commercially available have a wire coil in the middle. This is because they are made from two separate resistors joined together to achieve a good tolerance. They often have a spiral cut in the resistive film which effectively increase their self capacitance. So use was made of Victoreen resistor, which were also used by Presley.

The final circuit of the remote head is shown in fig. 5.16 showing the methods used to achieve the flexibility. This was constructed in two die cast boxes, joined end to end, with the electrometer mounted through the common wall. This was to screen the output from the input, in order to avoid ringing and instability problems.

The feedback resistor was mounted using miniature crocodile clips between two plates, approximately $5''$ by $3''$. One of these was a side of the die cast box (hence at earth potential), and the other was mounted parallel to it, and was driven from the amplifiers output. The clips allowed easy changing of feedback resistors to change ranges.

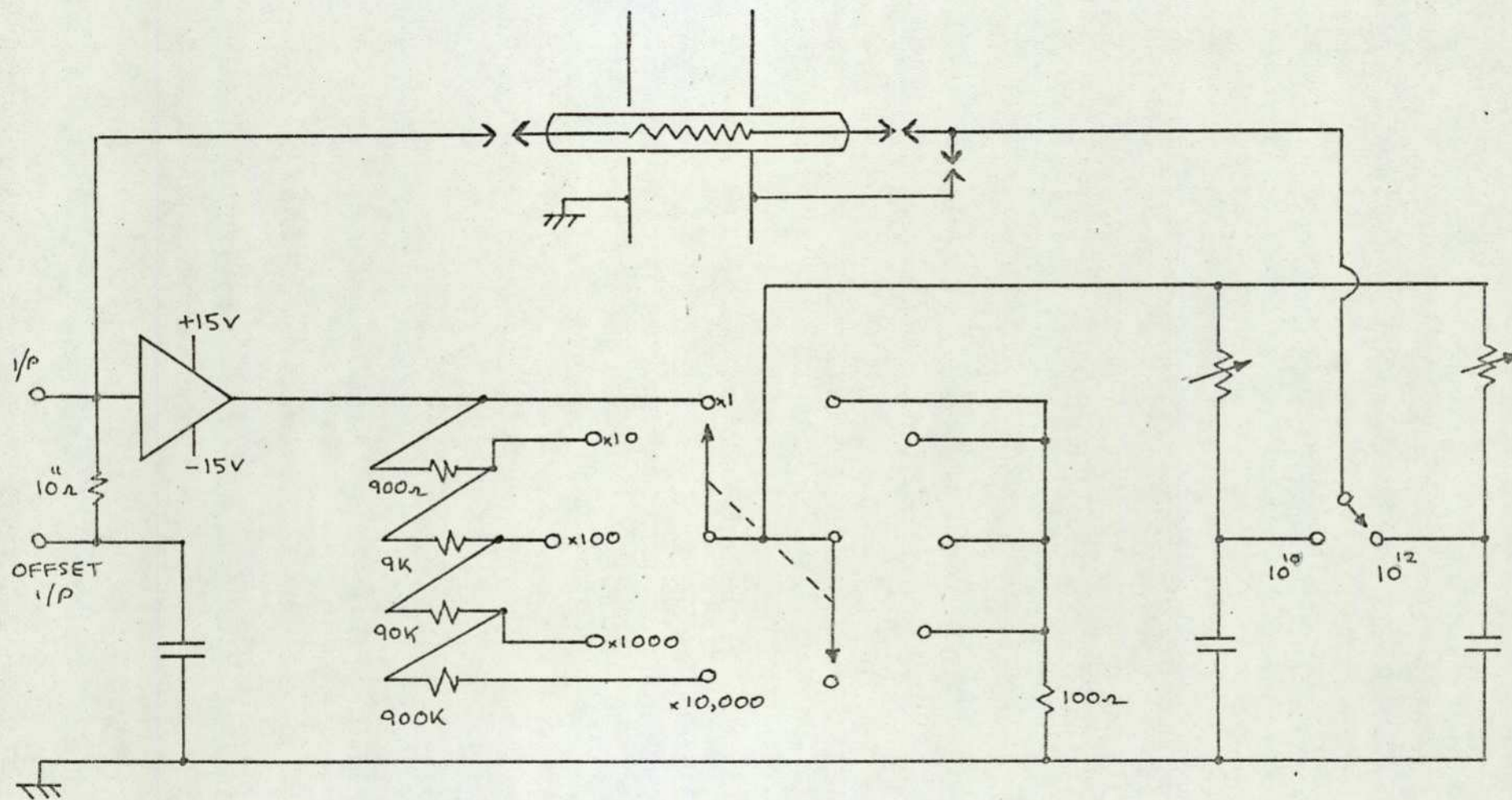


Fig. 5.16 Circuit of picoammeter head.

To match this, the circuit includes a switched choice of compensation circuits, incorporating a variable resistor to enable the circuit to be doped when required.

Further flexibility was provided by a switched potential divider (section 5.7) enabling the sensitivity to be changed in decades from $\times 1$ to $\times 10,000$.

The offset voltage from the control box is fed through a resistor connected to the input.

5.8.3 Control box and associated equipment

The control box contained the power supplies for the picoammeter remote head and housed the offset adjustment. Connections between the two were made by 20 ft. of screen cable using 5 pin DIN type plugs. This carried the +15, 0 and - 15v supplies, the offset voltage and the output.

The offset voltage was derived from 4 potentiometers, connected across the voltage supplies. The least sensitive could provide 150pA offset and the most sensitive .15pA. Such a range was found necessary to cope with the extremes of sensitivity of the picoammeter.

The power supplies were stabilised using two Fairchild μA 78015 three terminal regulators. These had a 1A current capability and a regulation such that the output voltage would change 7mV for a current change of 100mA. Stable supplies are essential in this application.

A Kenlow low pass filter was connected between the control box and the post-amplifier. This had a slope of 24dB/octave and could be varied down to almost d.c. The post amplifier was a Tekronix type 1A7 d.c. amplifier, housed in a storage scope. This provided a wide choice of gain and the scope was useful for offset adjustment.

5.8.4 Performance of the picoammeter

Two values of feedback resistor was used, $10^{10} \Omega$ and $10^{12} \Omega$, and the performance is listed below.

<u>SENSITIVITY (pA/V)</u>	<u>NOISE (pA)</u>	<u>RISETIME</u>
<u>$10^{10} \Omega$</u>		
100	.5	200uS
10	1	200uS
1	.2	500uS
.1	.05	
<u>$10^{12} \Omega$</u>		
1	1	10mS
.1	.05	
.01	.005	

These figures bear out the compromise between noise, speed and sensitivity. It should be noted that approximate peak to peak noise is not merely Johnson noise, but includes mains pick up and vibration effects.

5.8.5 Processing of current transients

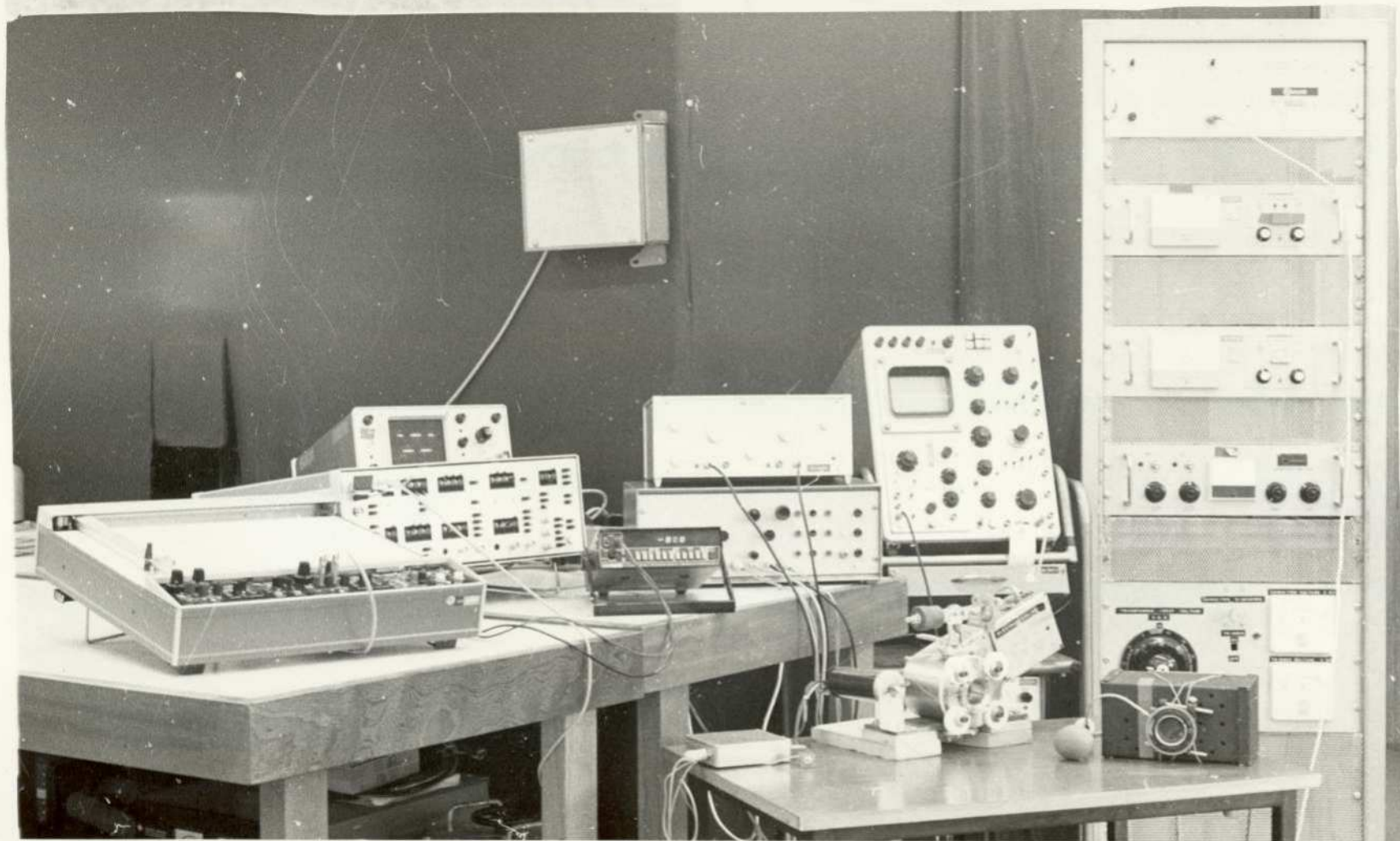
The original intention was to store the transients in the computer's disc store for ease of analysis and comparison. This intention was thwarted by the absence of the computer for a year and the lack of a disc store; because of the repercussions of a fire at the Ferranti works at Bracknell. Fortunately, this mishap coincided with the arrival of a Biomation transient recorder, which averted recourse to a storage scope and a camera.

The transient recorder is essentially a 100MHz A/D convertor coupled to a 2K of 8 bit store. Its power is aided by the elaborate

arming and triggering facilities it offers. This enables it to be used to store and display information which occurred before it was triggered.

The information in store can be viewed continually on a normal scope and can be output on a normal X - Y plotter. In this case a Hewlett Packard plotter was used which had an adequate risetime.

In practice, this proved to be a powerful means of capturing current transients. Plate 4 shows all the equipment which was used.



5.9 HIGH VOLTAGE SUPPLIES

A Brandenburg type 507R supply was used for voltages between 1 and 5kV and a Brandenburgh photocathode supply was used for lower voltages. A multiple stage RC filter was employed for smoothing, and the voltage measurements were taken from the meter of each supply. The calibration was checked by connecting a Pye electrostatic voltmeter to the test cell and the agreement was within experimental error. This checks the possibility of voltage dropping in the filter.

CHAPTER 6

RESULTS

6.1 INTRODUCTION

The following points were noted by the author when reviewing the literature containing liquid state current transients.

The presentation of these transients in the papers, and particularly in the theses, was such that any interpretation apart from the author's was made very difficult. For example, most transients did not even have a scale to the current magnitude. This prevents an assessment of possible space charge effects and liquid motion. As conduction in dielectric liquids is not well understood it is unfortunate for it prevents subsequent investigators from making use of the earlier results. There has also apparently been a tendency for authors to be selective, and to discard, or ignore, results which are not compatible with their particular hypotheses.

In view of this, an attempt will be made to present the results in this chapter with sufficient information, so as to enable the reader to easily make his own comparisons and interpretations of the transients.

These experimental measurements were conducted without any hypotheses in mind. The object was to obtain transients for various values of the possible variables; and to leave the interpretation to a later time. These parameters include liquid purity, current magnitude, field strength, temperature, injection rates, size of illuminated photocathode area, filters etc.

A certain amount of subjectivity must be involved in the selection of these results. Nevertheless, it will become apparent that there is a reasonable consistency to them, which reduces this possibility.

The emphasis in the presentation of the results in this chapter will be to allow the transients to speak for themselves.

6.2 BACKGROUND TO THE RESULTS

All the results in this chapter will be of a graph of current magnitude against time. The setting of the applied voltage and the transient recorder sampling rate was such that for all these results, a fixed point on the time scale corresponds to a certain mobility, regardless of the applied voltage. For example, a distance of 7.1 cm on the X axis corresponds to a mobility of $2.4 \times 10^{-8} \text{ m}^2/\text{V}\cdot\text{sec}$.

The use of the Tekronix amplifier means that the current density scale is either 1, 2 or 4 pA/cm^2 per cm.; or some factor of 10 of this. Rather than give this figure on the graphs, it is more meaningful to give the maximum current density of a transient as a percentage of the S.C.L. value for that field. This will allow easier assessment of the possibility of space charge effects and liquid motion. To calculate this S.C.L. value, a value of the carrier mobility is needed. To this end, a nominal figure of $2.4 \times 10^{-8} \text{ m}^2/\text{V}\cdot\text{sec}$. will be chosen.

Whenever a voltage figure is given, this will indicate the applied voltage. As the gap width was .945 cm., this figure will be close to the actual field strength.

The risetime of the transients were sometimes limited by either the amplifier response (plus the effect of post filtering), or by the time duration of the U.V. radiation. If the latter is the case then the initial part of the curve affected, will be left out. The occurrence of the former will be apparent from the initial rise of the transients. An examination of the results will show that either of these effects can mask a possible sharp decline at the start of the transients.

Appendix G contains a list of the appropriate data for each of the transients, giving the liquid sample, the number of days old, the measurement number of that day, the current density, scaling and the duration of the applied U.V. radiation.

To aid comparison, the transients have been transposed by hand from the XY plotter curve onto graph paper. In certain ways, this is unfortunate, as the human element is notorious for its subjectivity. An assessment of this process can be gained by comparing the transient of fig.6.29(a) to its original, shown in the frontpiece of this thesis.

A total of 14 liquid samples were measured (A to N) and a nomenclature will be used (i.e. B/2/8) to indicate respectively which liquid sample, the number of days old, and the measurement number on that day.

An attempt will be made to present the results with one parameter at a time varied, while the remainder are constant. To this end, most of the transients will be given for samples C and G; on which most measurements were made.

6.3 SUMMARY OF RELEVANT DATA

Here is a summary of data relevant to an interpretation of the following transients. The stability criterion is expressed as a current density percentage of the S.C.L. value. The figures are an approximate interpolation of data supplied by courtesy of T.R. Hewish. Similarly the conduction current density is treated for an assumed conductivity of $10^{-16} \text{ } \Omega^{-1} / \text{cm.}$

gap width	$.945 \pm 3\% \text{ cm}$
illuminated photocathode area	23.4 cm^2
nominal mobility	$2.4 \times 10^{-8} \text{ m}^2 / \text{V. sec.}$

Standard settings of applied voltage and time

V	50V	100V	250V	500V	1kv	2.5kv	5kv
Time sec/cm.	10.4	5.22	2.09	1.04	.522	.209	.104
J S.C.L. pA/cm^2	.013	.54	3.3	13.3	54	335	1,338
J stab %	-	67%	36%	25%	15%	11%	9%
δ %	3.6%	1.75%	.72%	.35%	.17%	.07%	.04%

X distance against mobility ($\times 10^8 \text{ m}^2 / \text{Vsec}$)

dist.(cm.)	1	2	3	4	5	6	7	8	9
μ	17.1	8.6	5.7	4.3	3.4	2.9	2.4	2.1	1.7

6.4 CURRENT TRANSIENTS

6.4.1 Amplifier distortion

Fig. 6.1 (b) shows a current transient taken on the first day of measurements. It exhibits unusual features compared to the other pulse transients, including one made on the same sample (fig. 6.1 (a)).

This puzzled the author for some time, until he realised that the energy used to fire the flash tube was seriously distorting the response of the picoammeter. This dissipation of energy occurs in less than a μs and comes from a bank of large capacitors charged up to about 800V. The coupling was probably due to electromagnetic radiation from a coil in the flash tube circuit being picked up by the extremely sensitive picoammeter input circuitry.

There were two characteristic features on the transients. The first is that the output gives negative for a definite duration ($\frac{1}{2}$ sec.). The second is that this upsets the d.c. balance of the picoammeter and after the duration of the transient, the output does not return to its previous level.

There is hence, the uncertainty that previous experimenters, whose measuring equipment was subject to a similar "shock" might have suffered a comparable distortion. Such distortion would be very difficult to detect if the technique of pulse current integration was used.

6.4.2 Effect of different liquid samples

A total of 14 samples (A to N) were used. The shapes of the transients (both pulse and step if available) are shown in figs. 6.1 to 6.14.

Here follows a summary of relevant information giving details of liquid preparation, approximate conductivity, whether the photocathode had an outer ring (Chapter 5), the film thickness etc.

<u>Sample</u>	<u>No. of measurements</u>	<u>maximum day old</u>	<u>film thickness</u> Å	<u>outer ring?</u>	<u>conductivity</u> $\times 10^{14} \Omega^{-1} \text{cm}^{-1}$
A	147	8	100	X	2
B	91	3	100	X	
C	405	12	148	X	80
D	3	1	148	X	3
E	5	2	200	X	9
F	8	1	74	X	7
G	67	3	74	X	100
H	12	2	111		
I	24	2	37		50
J	10	1	74		13
K	2	1	74	X	
L	3	1	74	X	
M	4	1	122		
N	15	1	122	X	

Sample A

Its U.V. transmittance is shown in fig. 5.2 .

Sample B

Initially, liquid B was tried with a 300 Å film. As there was no photo-current, the liquid was returned to the distillation system, and a new photocathode was fitted. But a certain amount of air entered the cell as well.

Sample D

This was the first liquid prepared on the improved distillation system (Chapter 5).

Sample I

Cell also contained a small quantity of nitrogen.

Sample J

This used redistilled sample I liquid. The design of the apparatus ensured that air was not able to enter the system.

Sample K

Purification materials were replaced before this sample was purified, and the transfer vessel contained zeolite.

Sample L

This used the same photocathode as for sample K.

Sample ~~N~~ M

Purification ^{materials} ~~samples~~ were heated under vacuum by external heating tapes.

Sample ~~M~~ N

Purification ^{materials} ~~samples~~ were thoroughly "flushed" with 3⁰ cut hexane and then with used Koch light hexane. The actual liquid sample was repurified used Koch light, from the previous samples.

6.4.3 Effect of time

Fig. 6.16 shows transients made on sample C after 2 and 12 days. Fig. 6.17 shows transients obtained at the beginning and end of measurements over a day.

6.4.4 Effect of voltage

Fig. 6.15 and 6.28 (a) and (b) reveal the effect, on pulse transient shape, of applied voltage.

6.4.5 Effect of current magnitude

Figs. 6.18 to 6.21 illustrate how the step transient changes with current magnitude for different voltages; similarly fig. 6.22 is for a pulse transient.

Figs. 6.23 to 6.27 makes a comparison between a low level pulse transient and a transient obtained by momentarily blocking the U.V. radiation off from a continuous ON state. In the latter case, the transient is effectively of a layer of no charge, moving across the gap. In this case and with all OFF step transients, they are inverted for easier comparison.

6.4.6 Effect of filtering

Fig. 6.29 shows the effect of filtering the U.V. with an OX7 filter for both normal and reversed applied voltage polarity.

6.4.7 Forward and reverse transients

Fig. 6.29 also shows the difference in shape between transients, due to the same U.V. intensity, with normal and reverse polarity across the cell. Presumably the contribution by bulk ionisation to the transient would be the same in both cases. This was done for both filtered and unfiltered radiation. The U.V. intensity was not constant between these two cases.

6.4.8 Effect of temperature

Measurements were made at different temperatures by placing the cell and light source in a fridge/oven. The effect is shown in figs. 6.8 and 6.9. The large thermal inertia of the heavy metal cell obviated the need to measure the actual liquid temperature. At lower temperatures, care had to be taken to prevent condensation forming on the cell and causing leakage currents. It was also found that if the

cell was subject to a rapid change in temperature, then thermal stress could cause the quartz photocathode to crack.

6.4.9 Linearity of transients

Fig. 6.28 shows transients obtained on sample G to investigate how linear the conduction mechanism was. It shows two low level pulse transients with a ratio of current densities of about 600. The smaller current density was 10^{-15} A/cm^2 .

Step transients were also obtained by masking only part of the photocathode and then suddenly uncovering it. This should show if the transient shape is the same for carriers at either the centre of the cathode or the edge. An OFF step transient is also included.

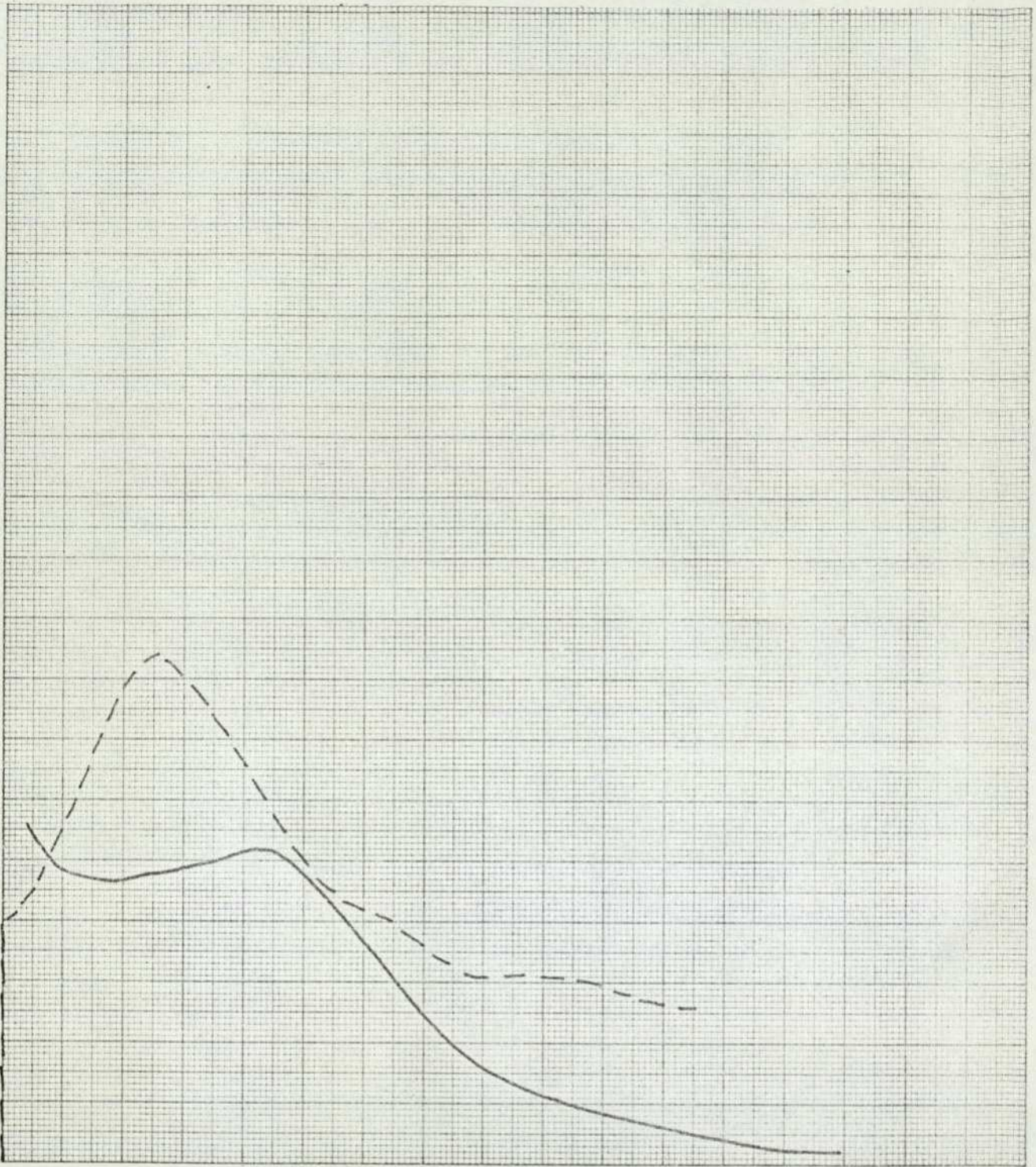


Fig. 6.1 Sample A transients at 1kV

(a) pulse (A/7/72, .9% of S.C.L.) —

(b) distorted pulse (A/1/6) - - -

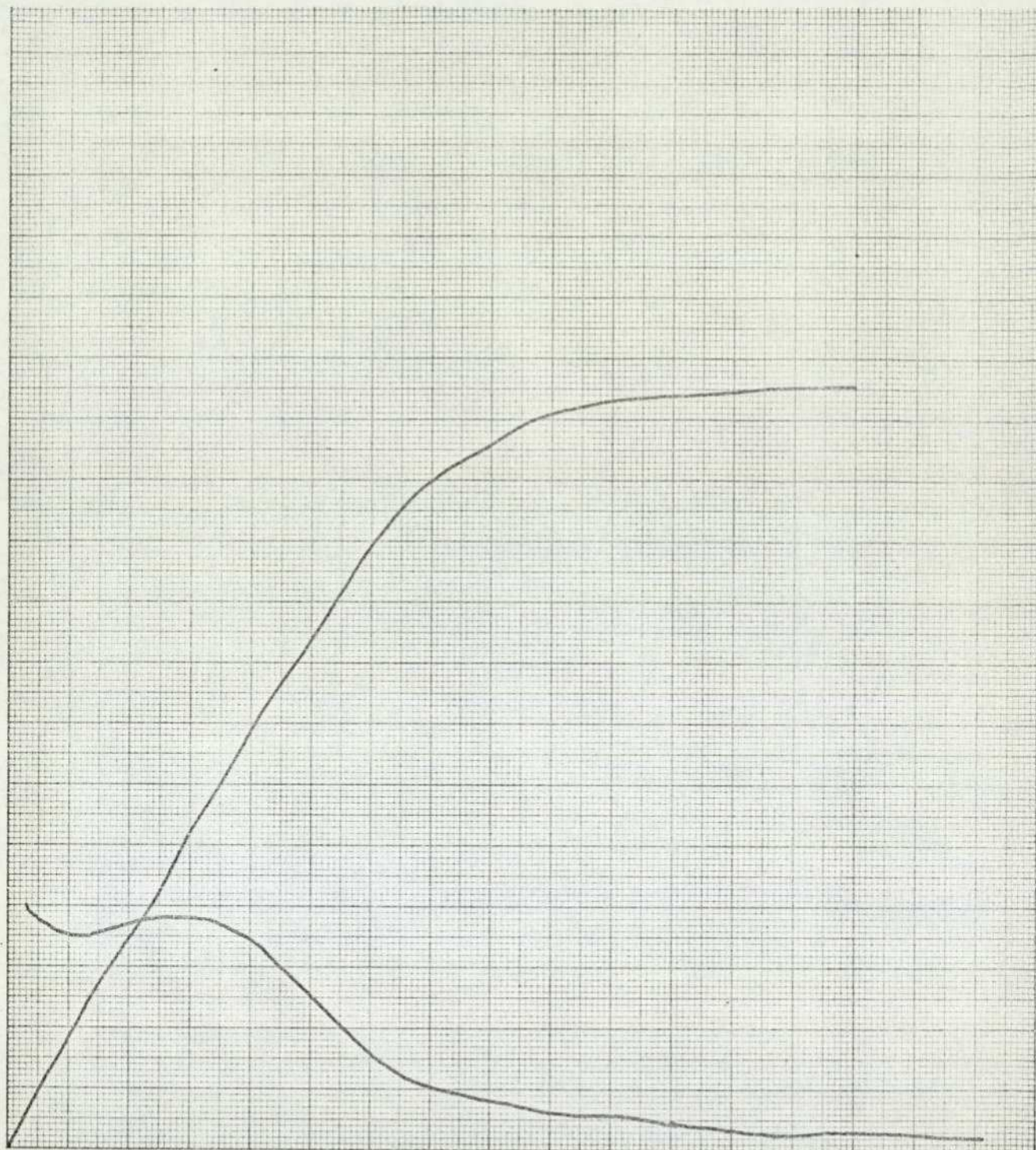


Fig. 6.2 Sample B transients at 1kV

(a) pulse (B/3/2, .7% of S.C.L.)

(b) step (B/3/15, 4.7% of S.C.L.)

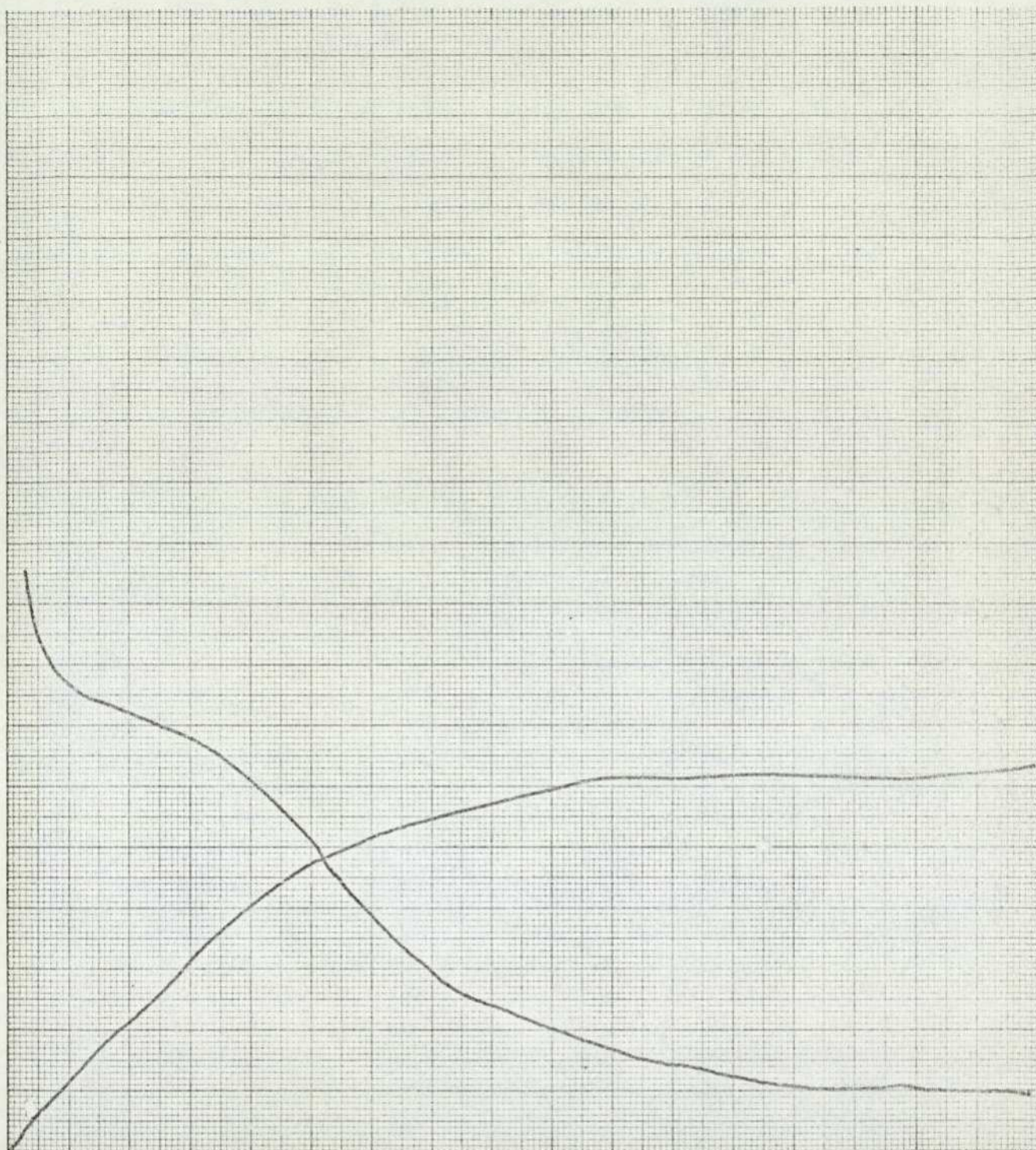


Fig. 6.3 Sample C transients at 1kv

(a) pulse (C/3/2, 1.3% of S.C.L.)

(b) step (C/5/62, 1.2% " ")

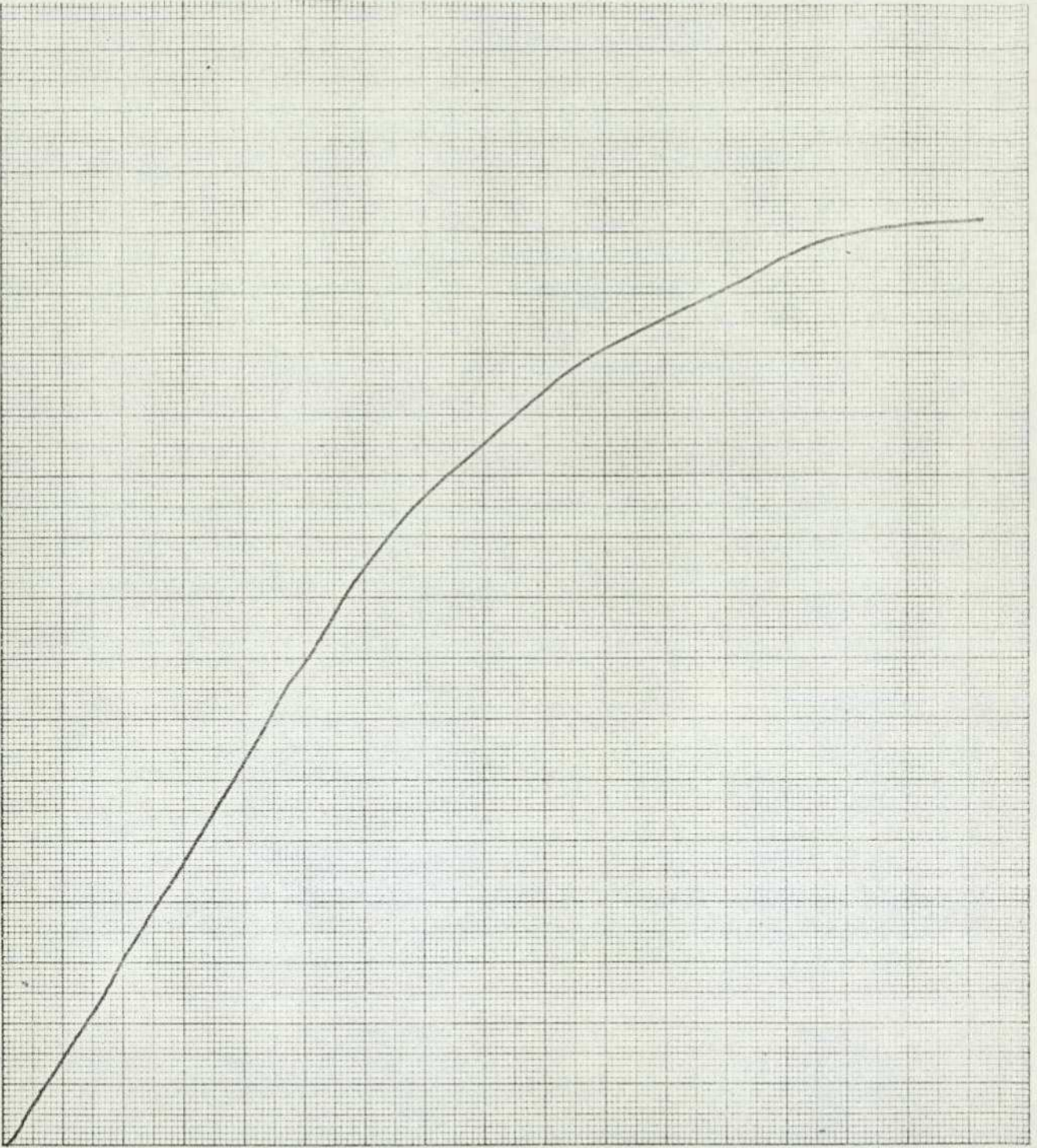


Fig. 6.4 Sample D transients at 2.5kv

(a) step (D/1/3, .09% of S.C.L.)

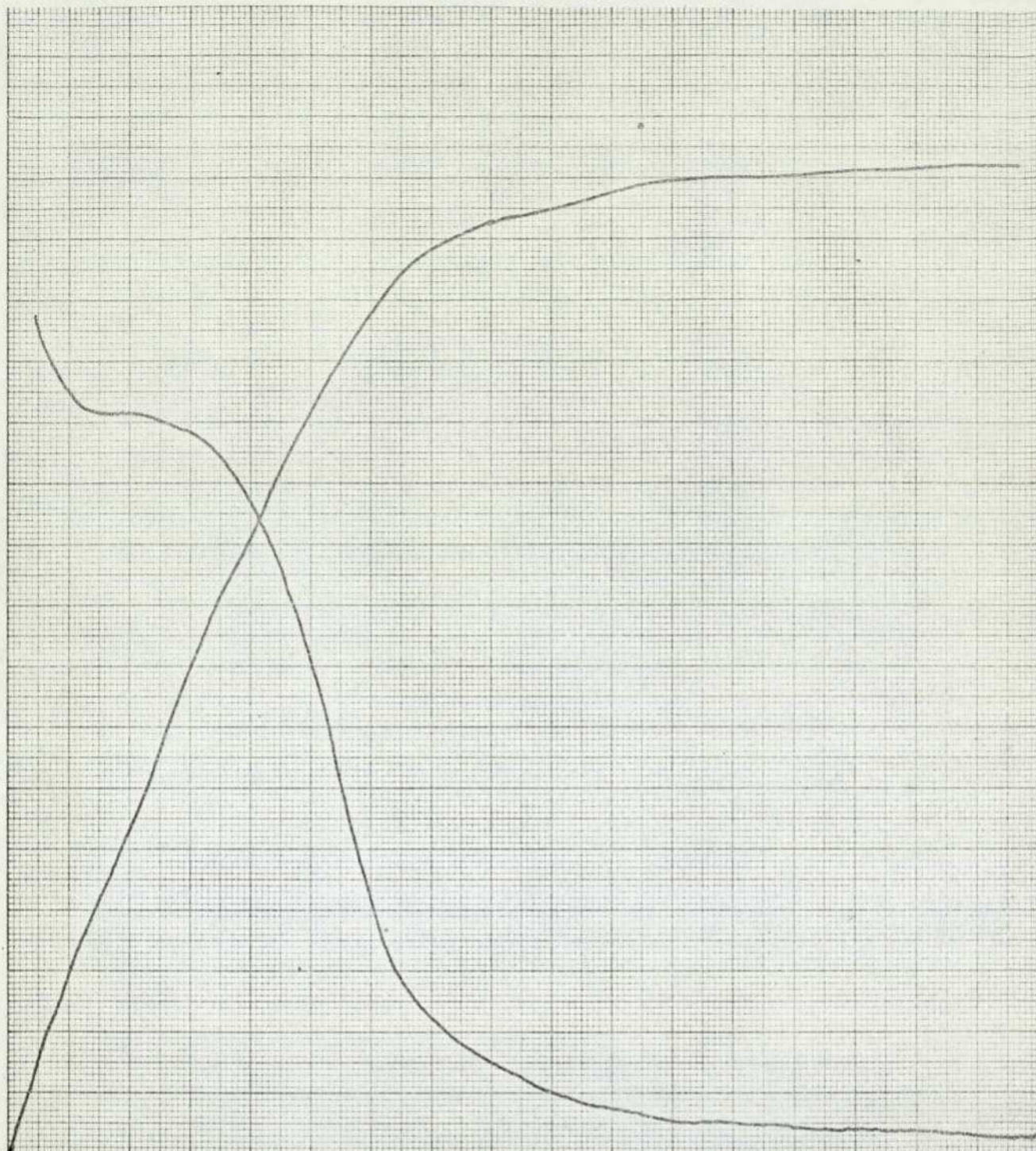


Fig. 6.5 Sample E transients at 2.5kv

(a) pulse (E/2/1, .14% of S.C.L.)

(b) step (E/1/1, .19% " ")

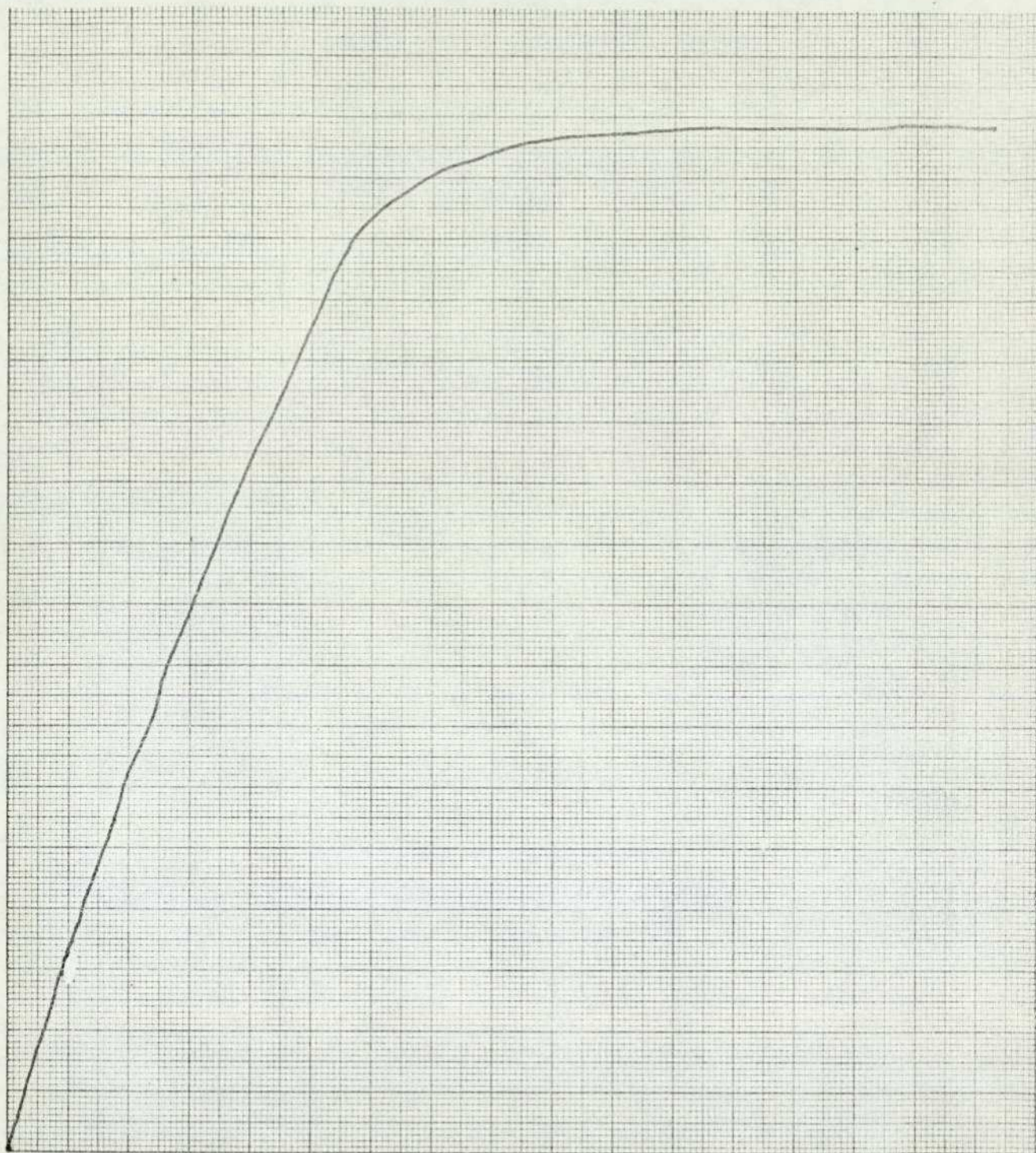


Fig. 6.6 Sample F transients at 2.5kv

(a) step ($F/1/5$, .5% of S.C.L.)

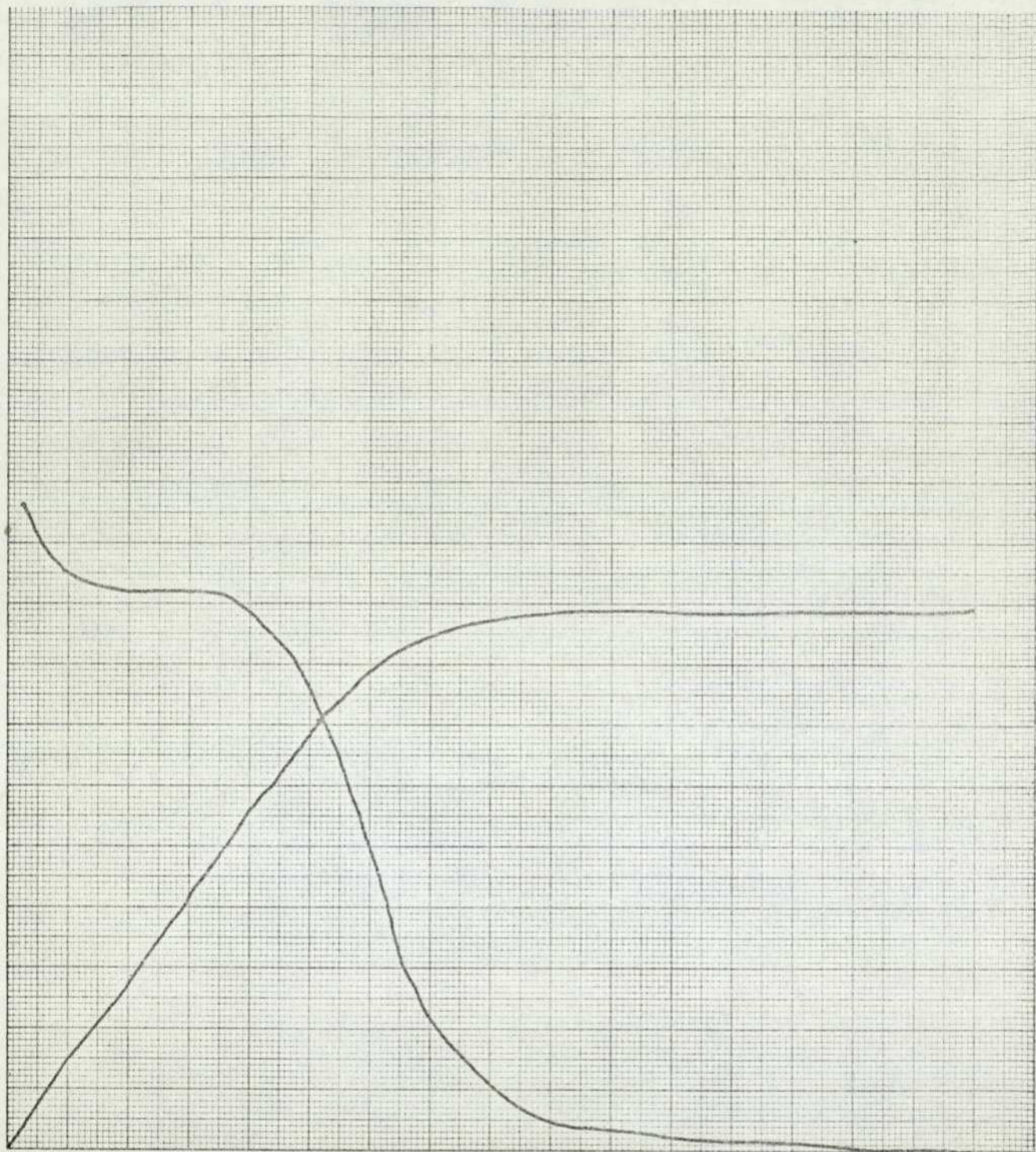


Fig. 6.7 Sample G transients

(a) pulse at 2.5kv (G/1/1, .11% of S.C.L.)

(b) step at 1 kv (G/3/13, .33% " ")

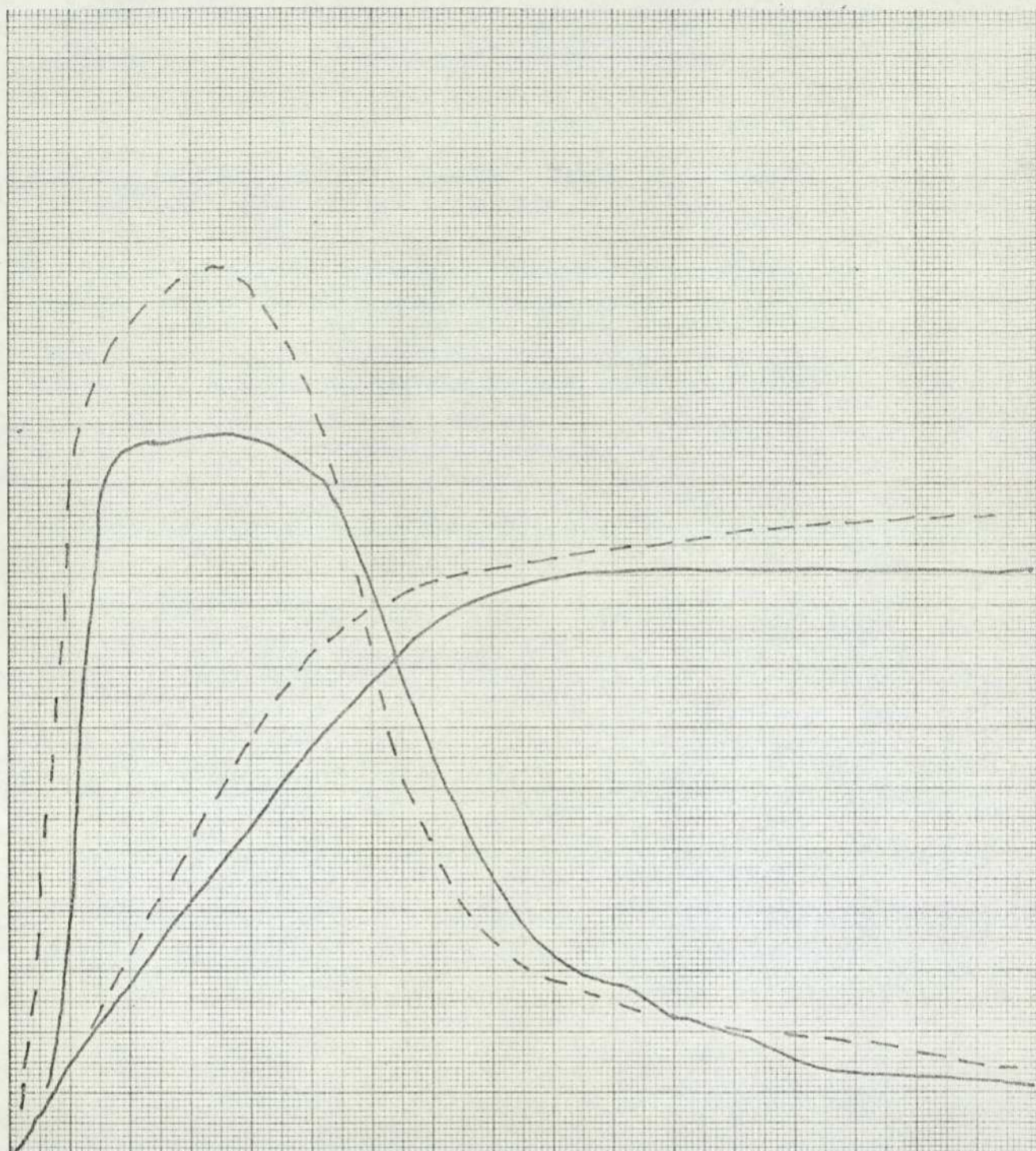


Fig. 6.8 Sample H transients at 1kv illustrating the effect of an increase in temperature.

- (a) pulse at 20°C (H/2/2, .08% of S.C.L.) —
- (b) step at 20°C (H/1/1, .36% " ") —
- (c) pulse at 50°C (H/2/5, .11% " ") - -
- (d) step at 50°C (H/2/4, .08% " ") - -

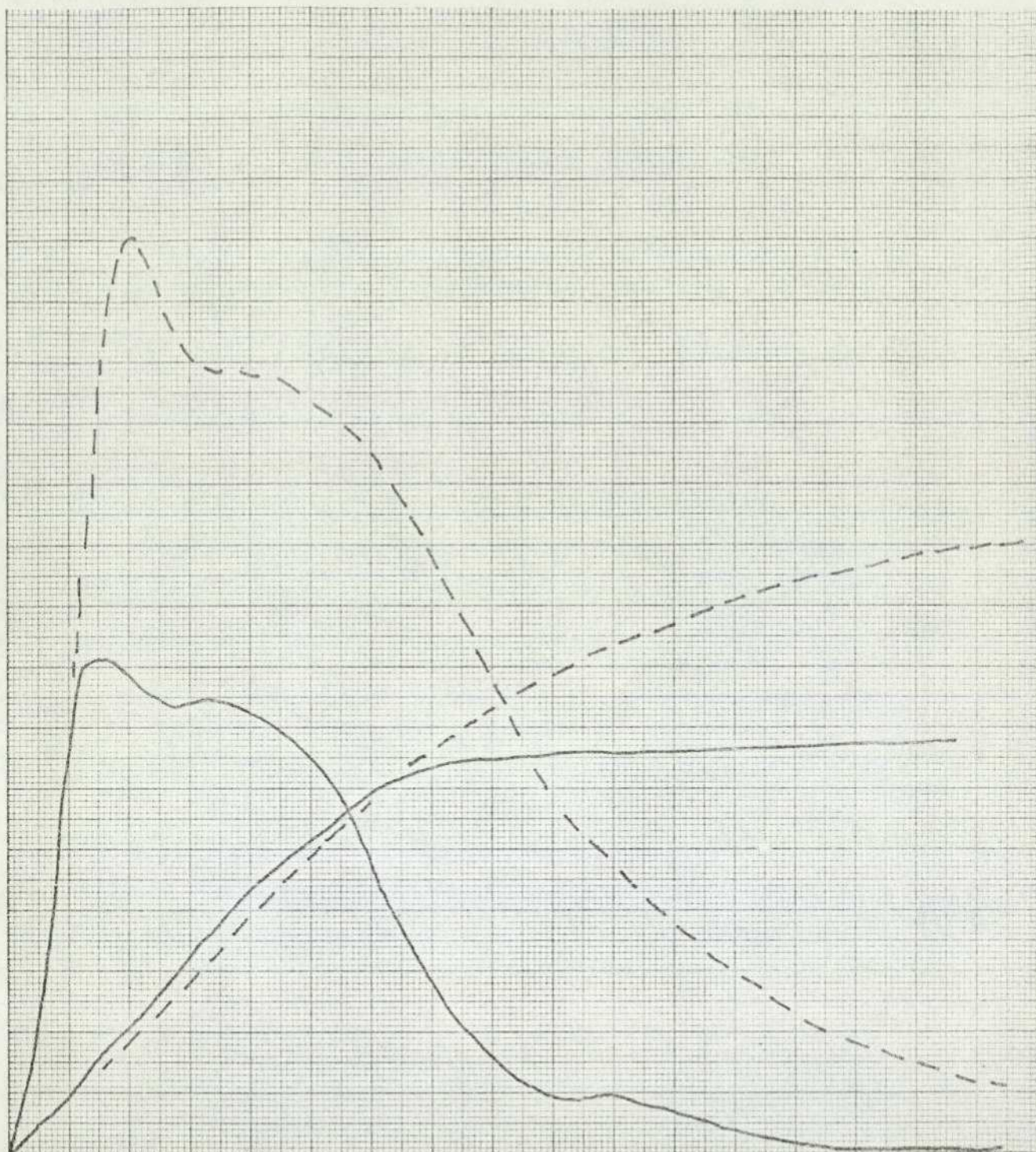


Fig. 6.9 Sample I transients at 1kv illustrating the effect of a decrease in temperature.

- (a) pulse at 20°C (I/1/3, .01% of S.C.L.) —
- (b) step at 20°C (I/1/1, .5% " ") —
- (c) pulse at 8°C (I/2/17, .24% " ") - -
- (d) step at 8°C (I/2/20, .07% " ") - -

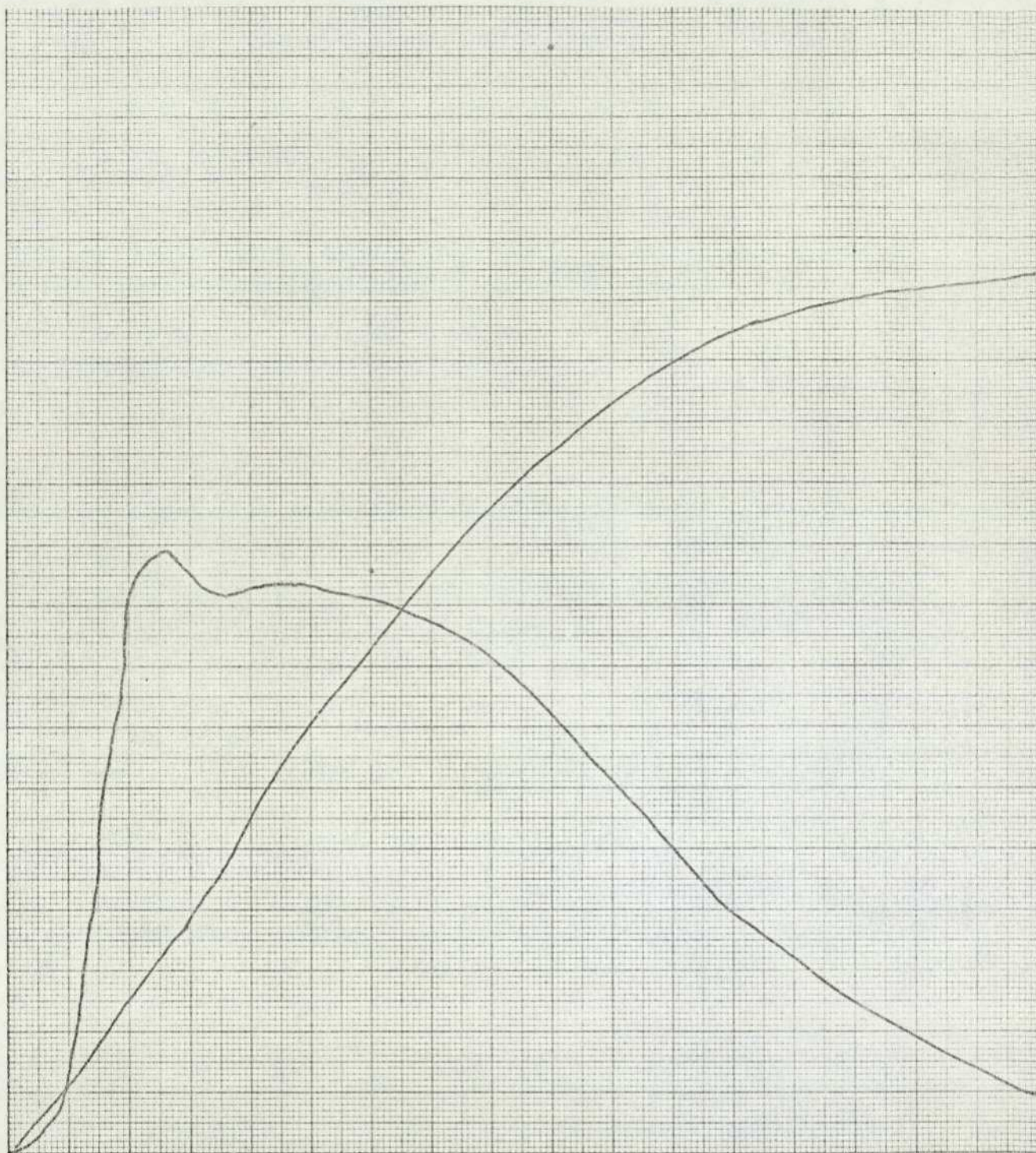


Fig. 6.10 Sample J transients at 1kv (using redistilled sample I liquid)

(a) pulse (J/1/3, .035% of S.C.L.)

(b) step (J/1/1, 1.1% of S.C.L.)

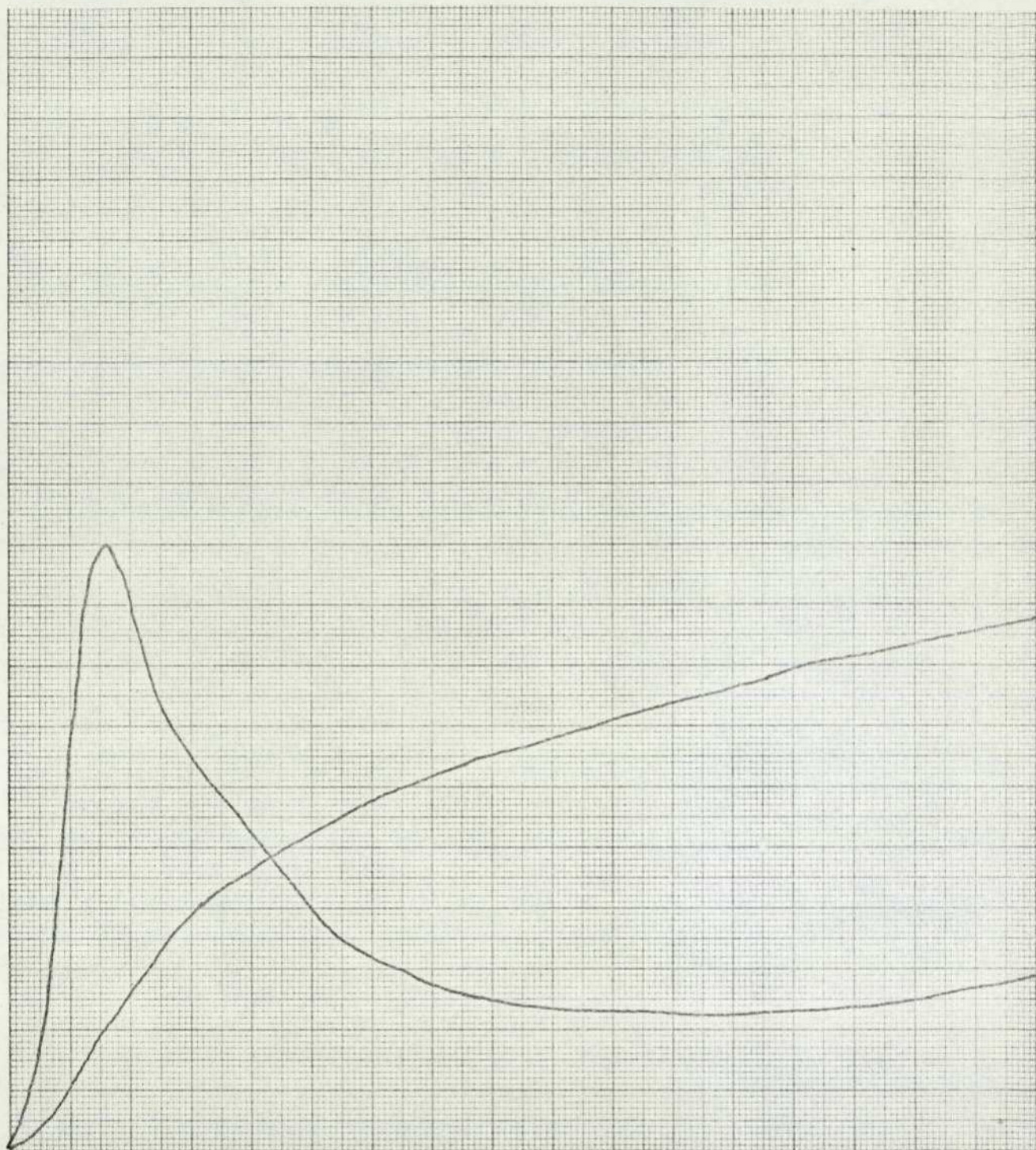


Fig. 6.11 Sample K transients at 1kv

(a) pulse (K/1/1, .19% of S.C.L.)

(b) step (K/1/2, 1.6% of S.C.L.)

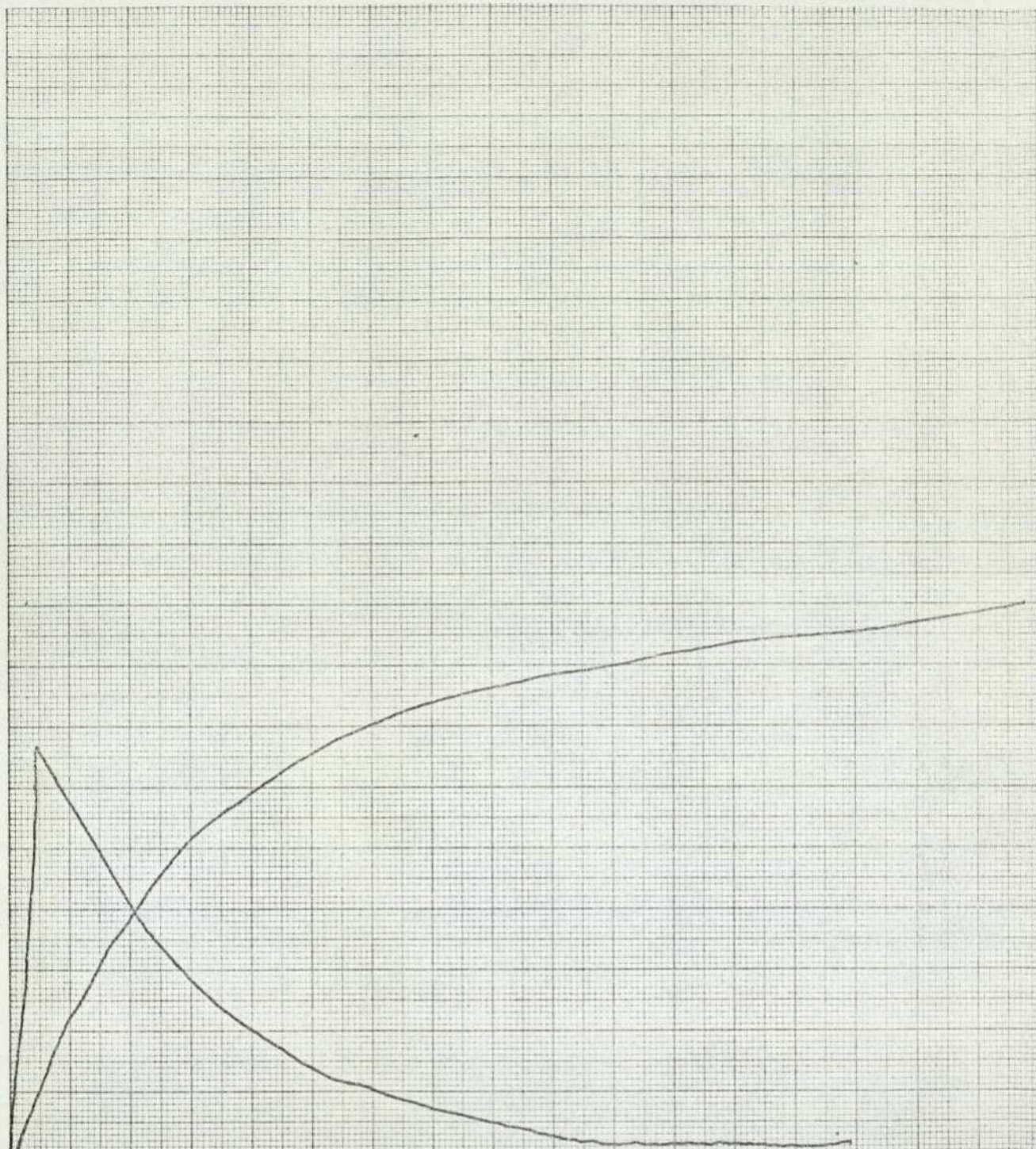


Fig. 6.12 Sample L transients at 1kv

(a) pulse (L/1/3, .12% of S.C.L.)

(b) step (L/1/1, 1.7% " ")

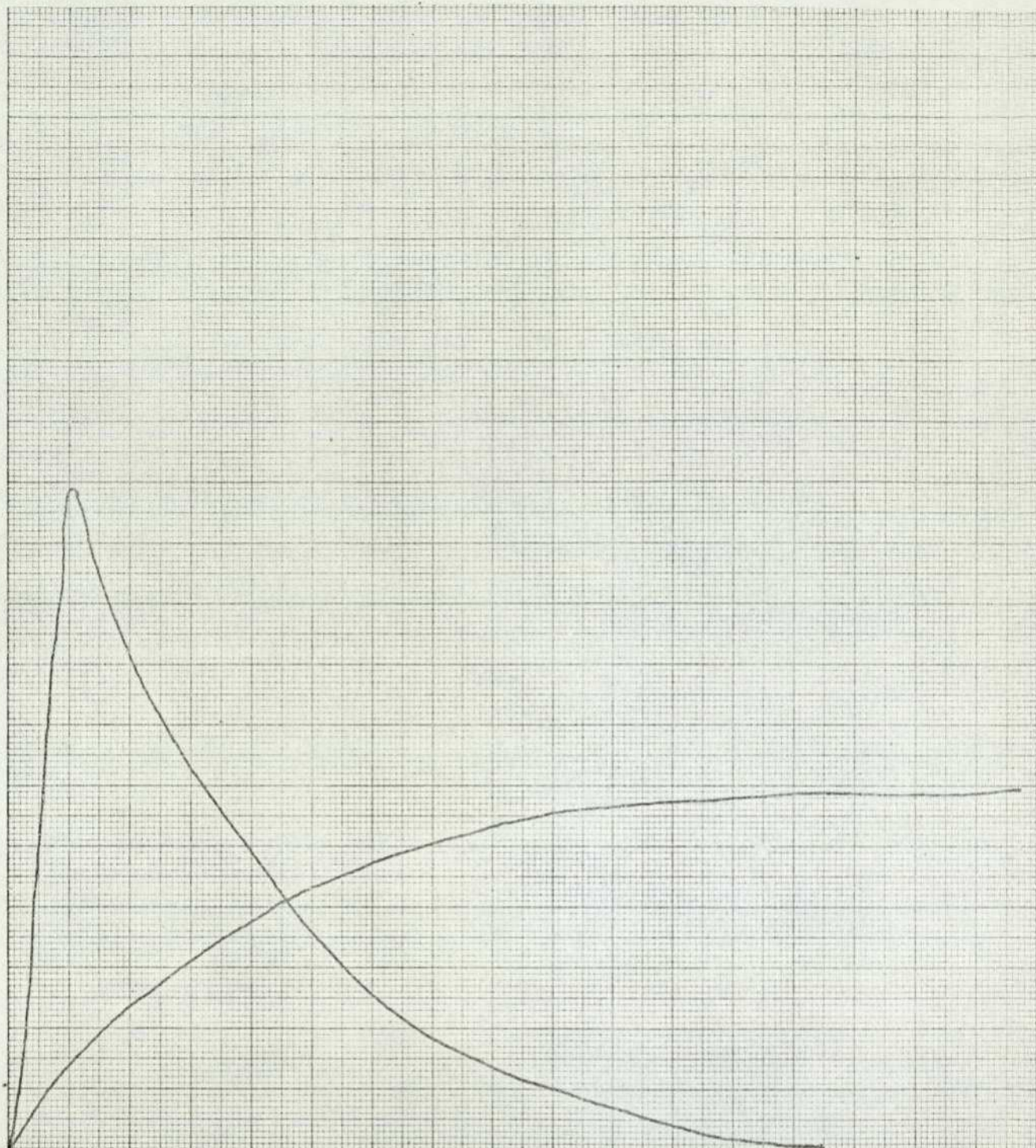


Fig. 6.13 Sample M transients at 1kv

(a) pulse (M/1/1, .08% of S.C.L.)

(b) step (M/1/2, .21% of S.C.L.)

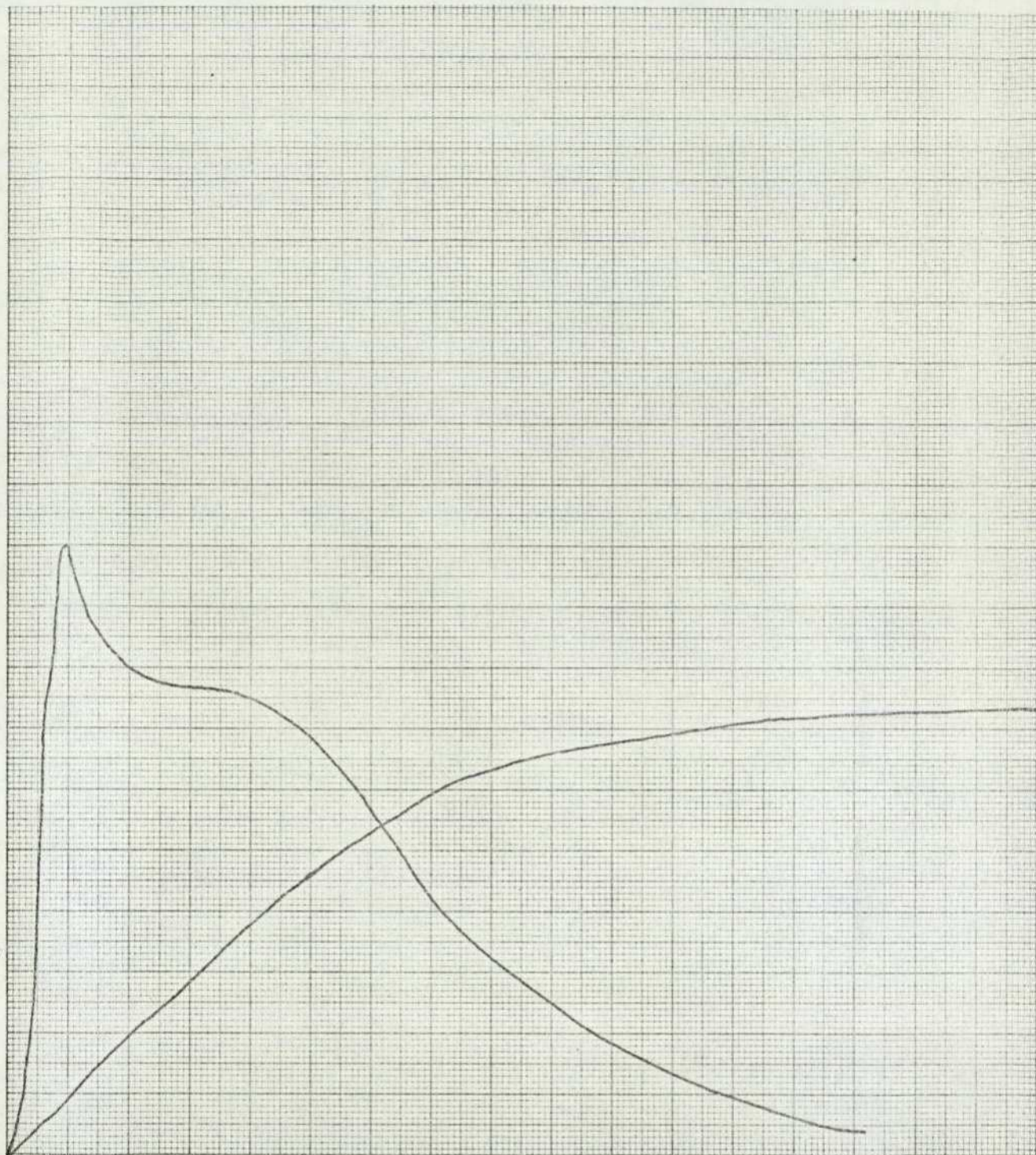


Fig. 6.14 Sample N transients at 1 kv

(a) pulse (N/1/1, .03% of S.C.L.)

(b) step (N/1/3, .54% " ")

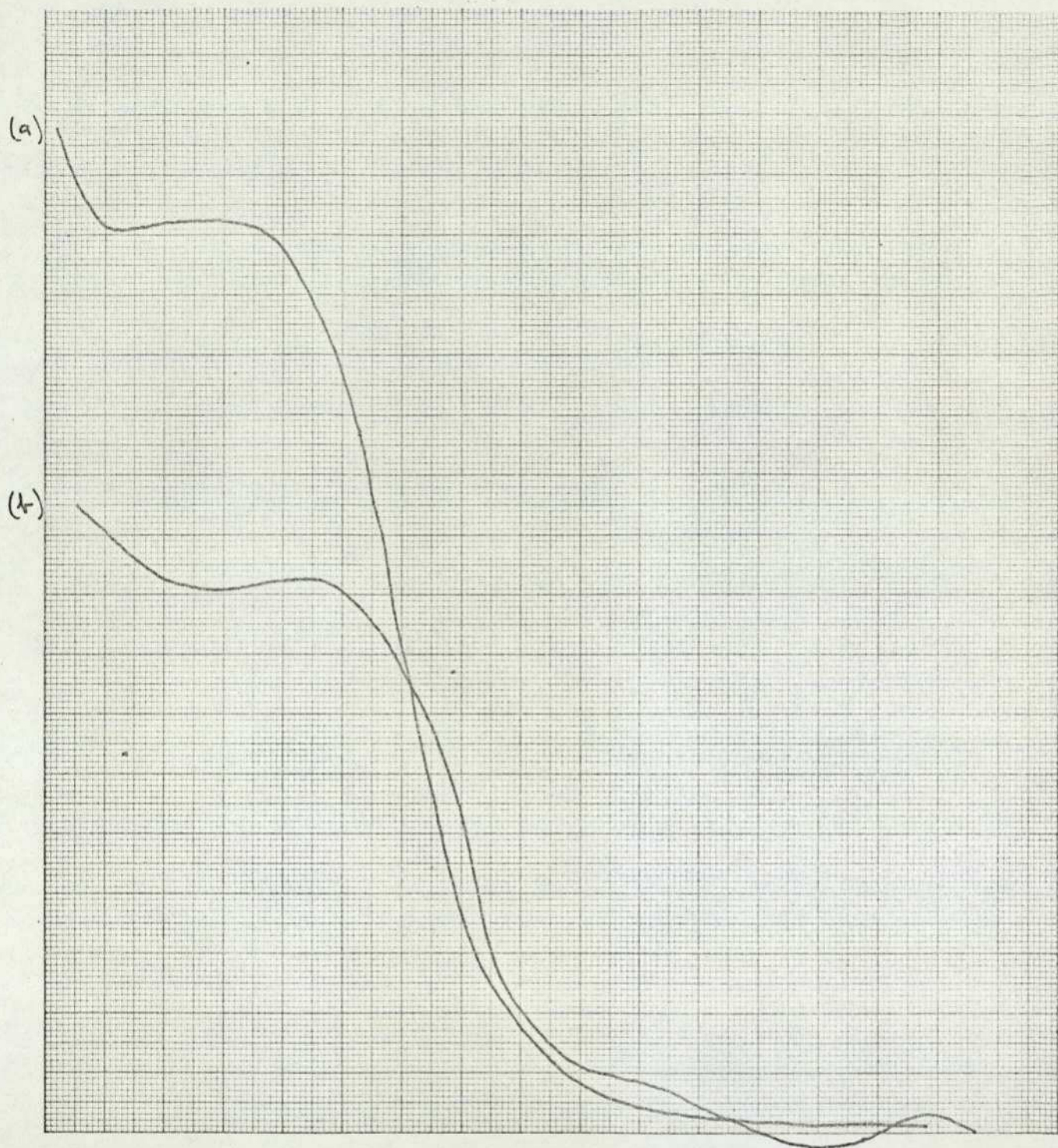


Fig. 6.15 Transient shape against voltage

(a) 2.5kv (G/1/2, .18% of S.C.L.)

(b) 250v (G/2/25, .003% of S.C.L.)

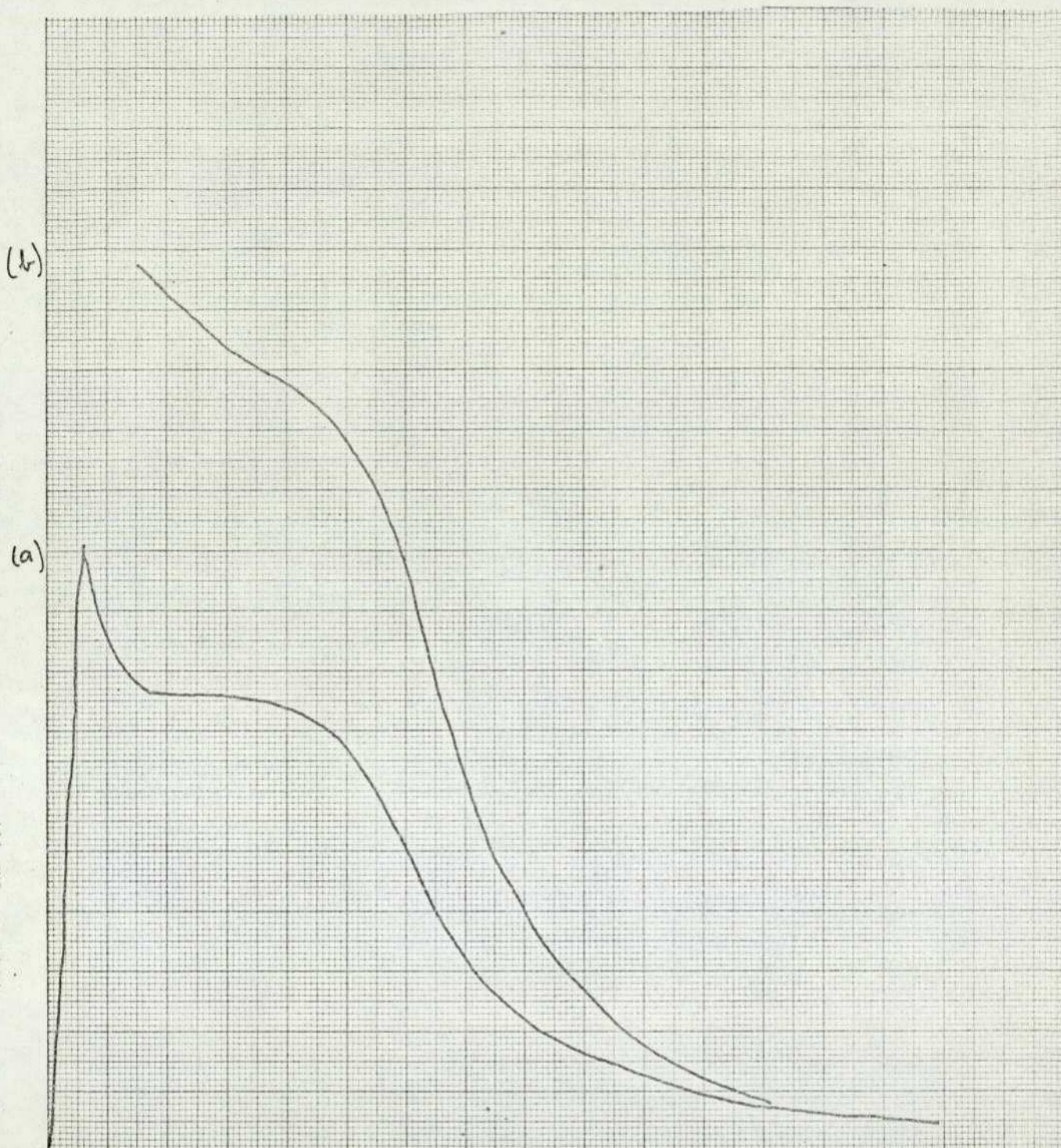


Fig. 6.16 Transient shape against time (at 2.5kv)

(a) 3 days old (G/3/23, .44% of S.C.L.)

(b) 12 days old (G/12/13, 1.6% of S.C.L.)

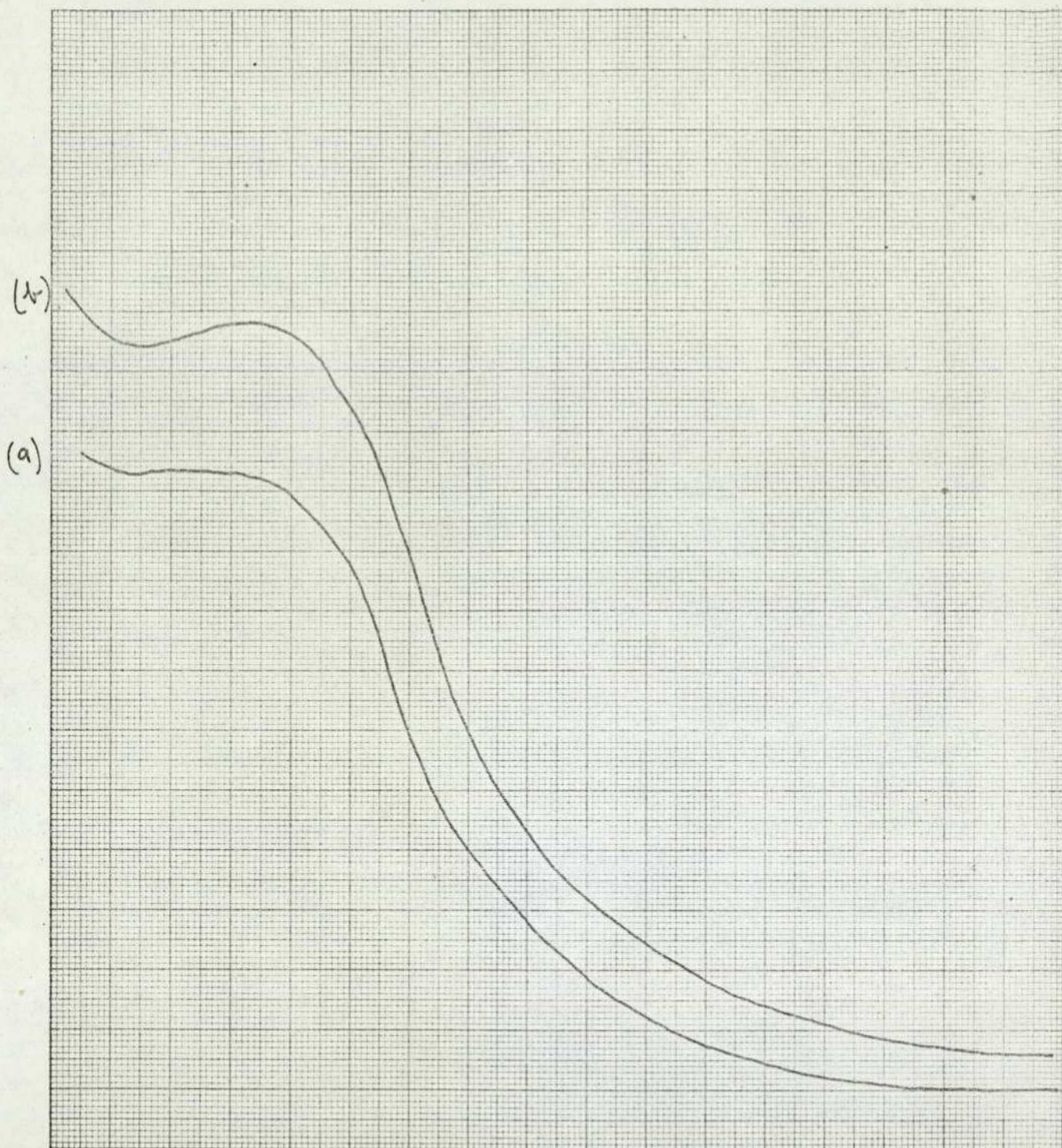


Fig. 6.17 Transient shape at the beginning and end of a day
(at 2.5kv and with OX7 filtering)

(a) at finish (C/4/48, .08% of S.C.L.)

(b) at start (C/4/1, .04% " ")

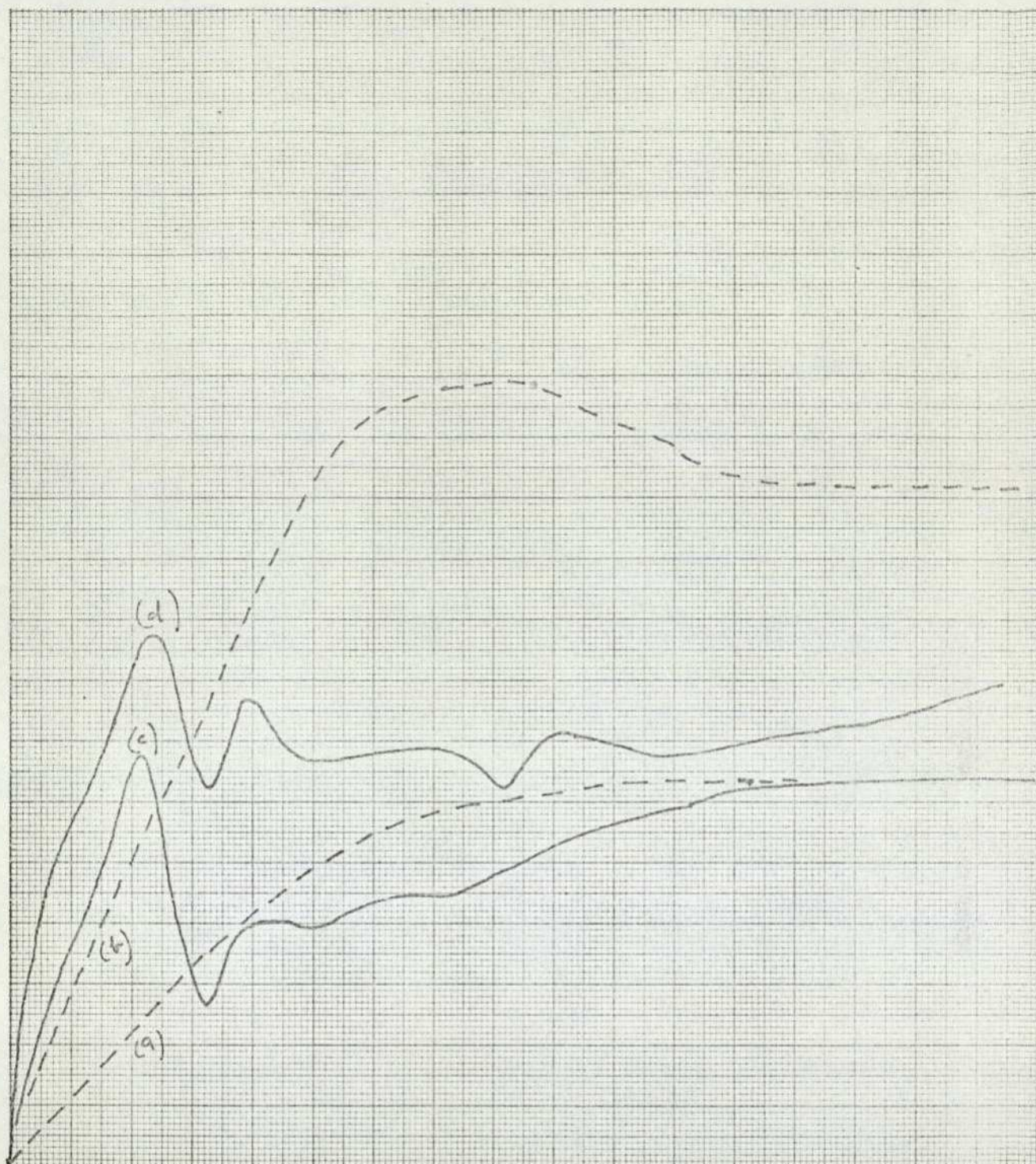


Fig. 6.18 Step transient shape against current density at 100V

- (a) 23% of S.C.L. (C/5/71)
- (b) 41% " " (C/5/70)
- (c) 119% " " (C/4/32)
- (d) 148% " " (C/5/42)

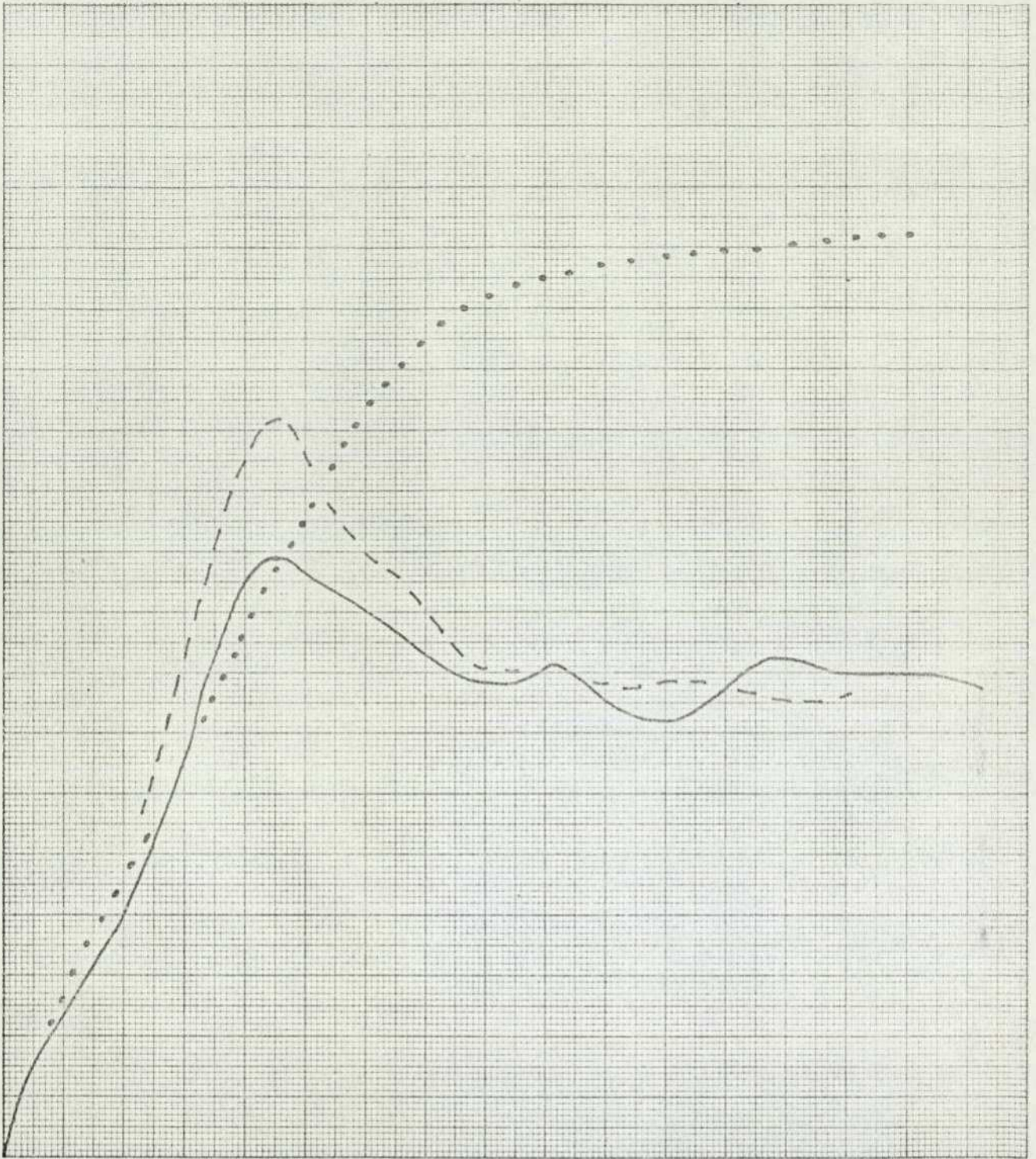


Fig. 6.19 Step transient shape against current density at 250V

- | | | | |
|-----|----------------|----------|---------------|
| (a) | 9.2% of S.C.L. | (C/5/67) | • • • |
| (b) | 46% | " " | (C/2/33) — |
| (c) | 97% | " " | (C/4/5) - - - |

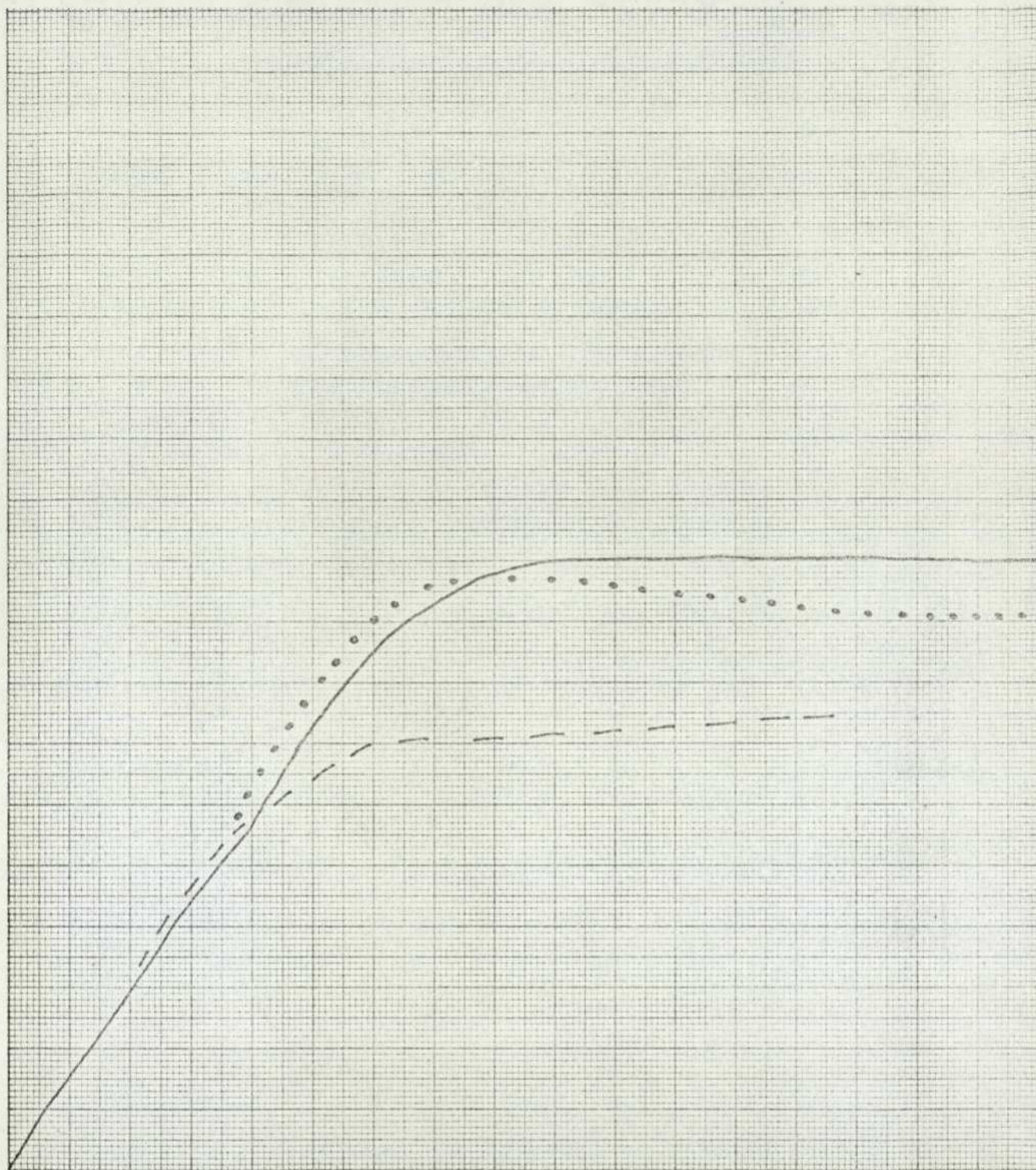


Fig. 6.20 Step transient shape against current density at 1kv, showing ON and OFF transients

- (a) 7.4% of S.C.L., OFF (C/7/3) ———
- (b) 14% " " OFF (C/2/20) - - -
- (c) 17% " " ON (C/2/35) . . .

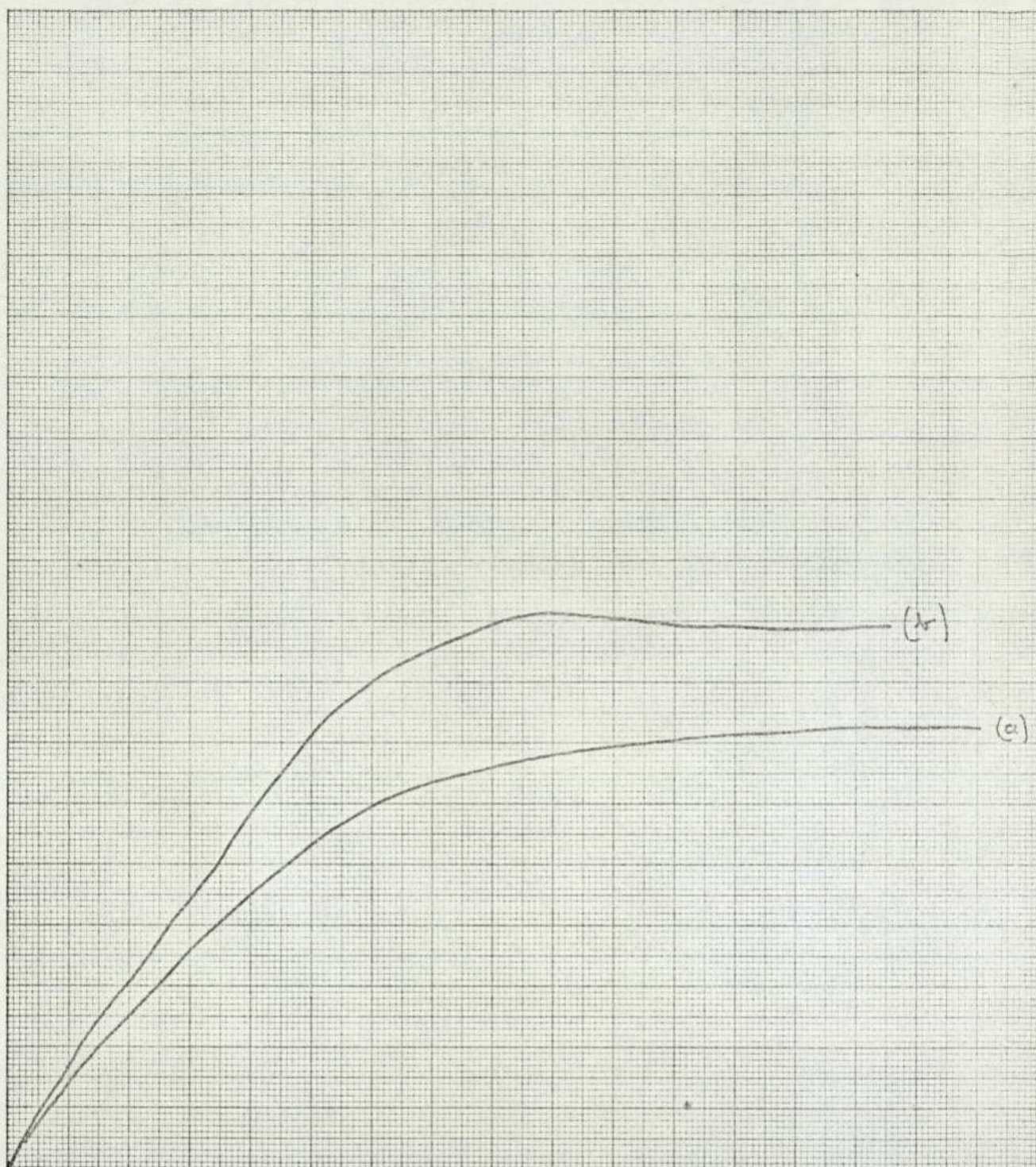


Fig. 6.21 Step transient shape against current density at 2.5kv

(a) .87% of S.C.L. (C/3/29)

(b) 5.3% " " (C/1/36)

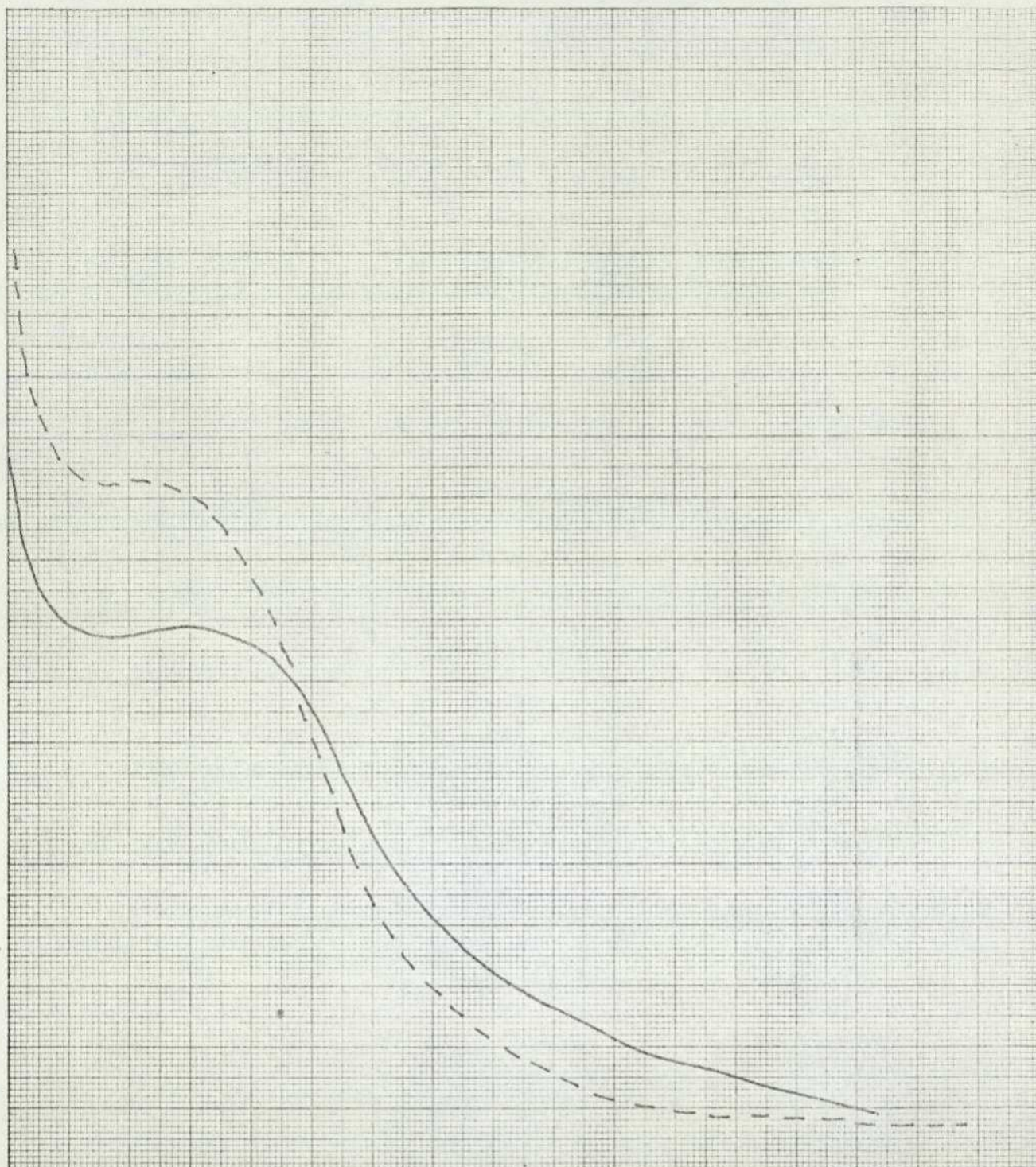


Fig. 6.22 Pulse transient shape against current density at 250V

(a) 2.6% of S.C.L. (C/5/43) ———

(b) 14% " " (C/5/40) - - -

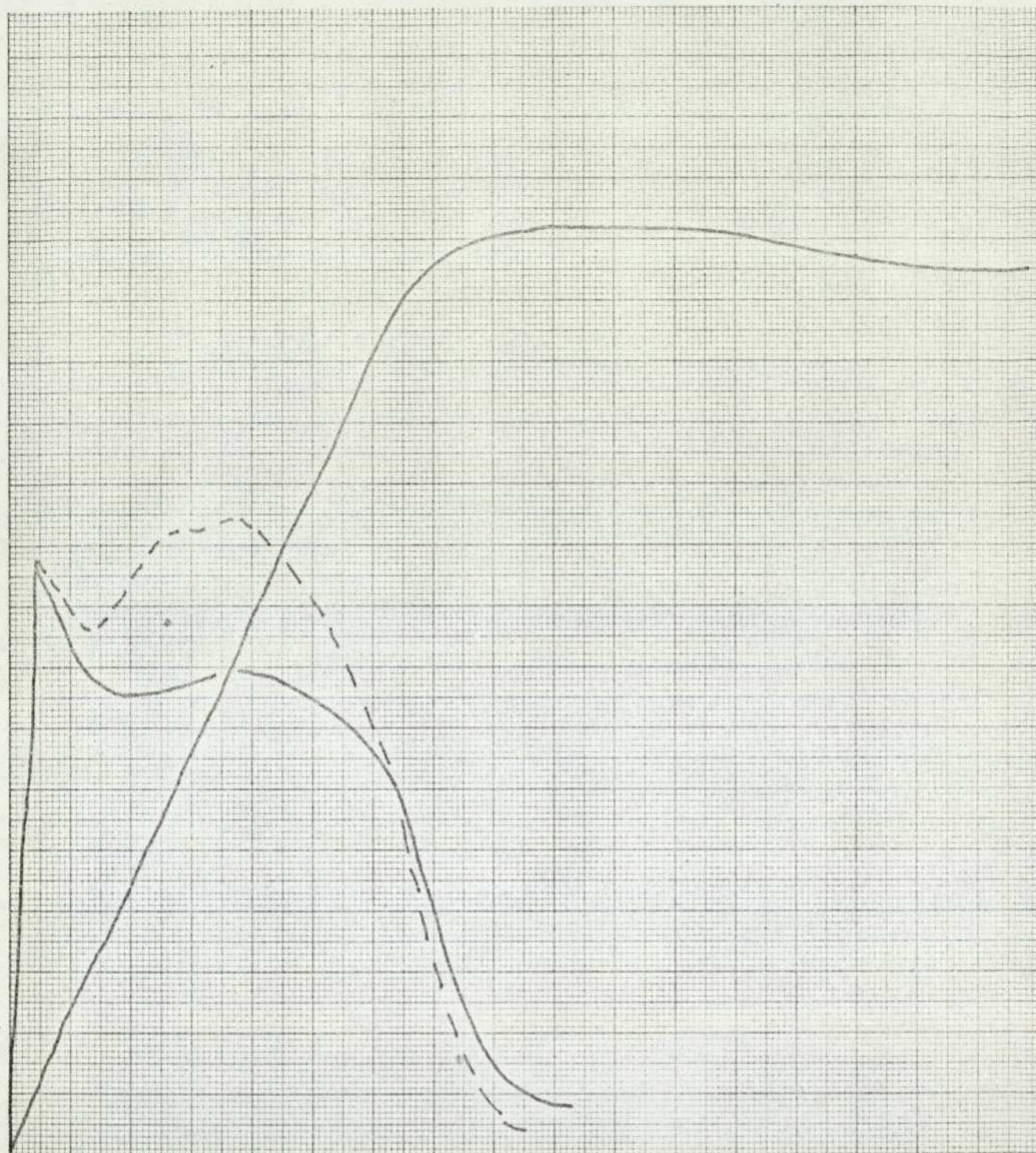


Fig. 6.23 Pulse transient shape at low and high current densities;
 .31% and 4.4% of S.C.L. at 250V

(a) step, 4.4% of S.C.L. (G/2/22)

(b) low level pulse, .23% of S.C.L. (G/2/23) ———

(c) OFF pulse by interrupting U.V. at the level of (a), — —
 .31% of S.C.L. (G/2/24)

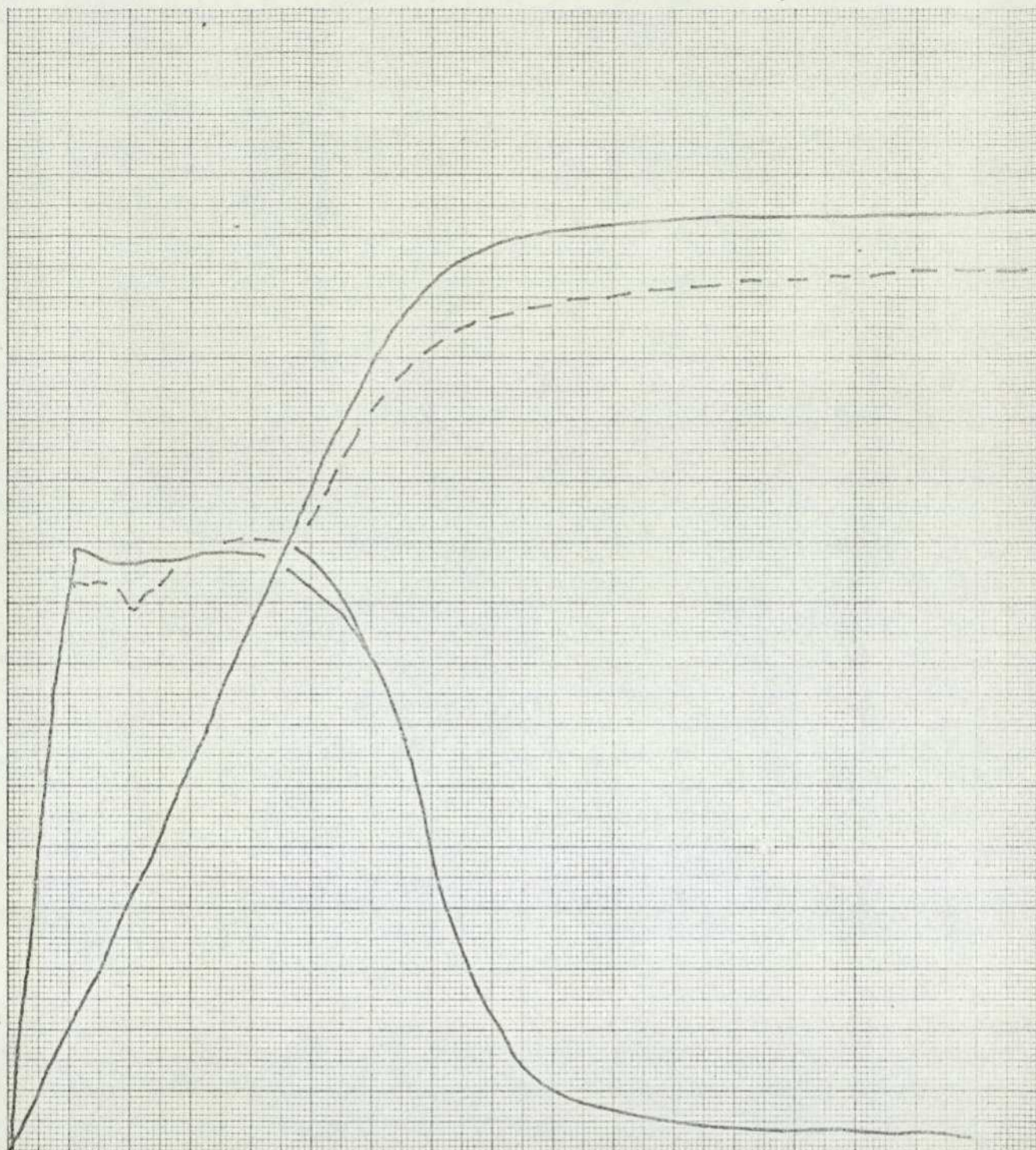


Fig. 6.24 Pulse transient shape at low and medium current densities;
 .15% and 1.2% of S.C.L. at 500V

- (a) ON step, 1.2% of S.C.L. (G/2/18) —
- (b) OFF " 1.1% " " (G/2/17) - -
- (c) low level pulse, .15% of S.C.L. (G/2/21) —
- (d) OFF pulse by interrupting U.V. at the level of (a), - -
 .15% of S.C.L. (G/2/16)

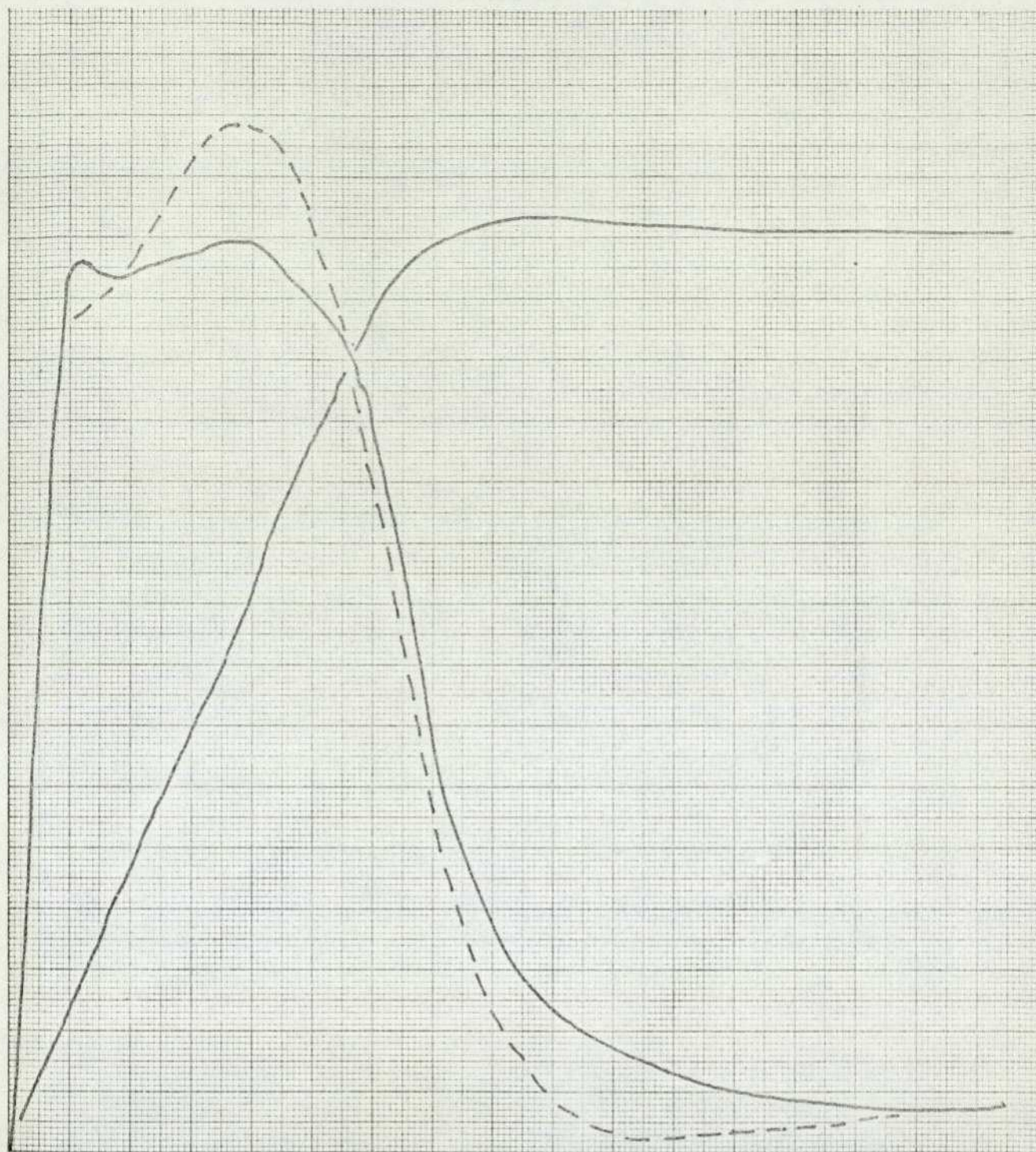


Fig. 6.25 Pulse transient shape at low and high current densities;
 .22% and 2.3% of S.C.L. at 500V

(a) ON step transient, 2.3% of S.C.L. (G/2/14)

(b) low level pulse, .22% of S.C.L. (G/2/15)

(c) OFF pulse by interrupting U.V. at the level of (a), — —
 .25% of S.C.L. (G/2/16)

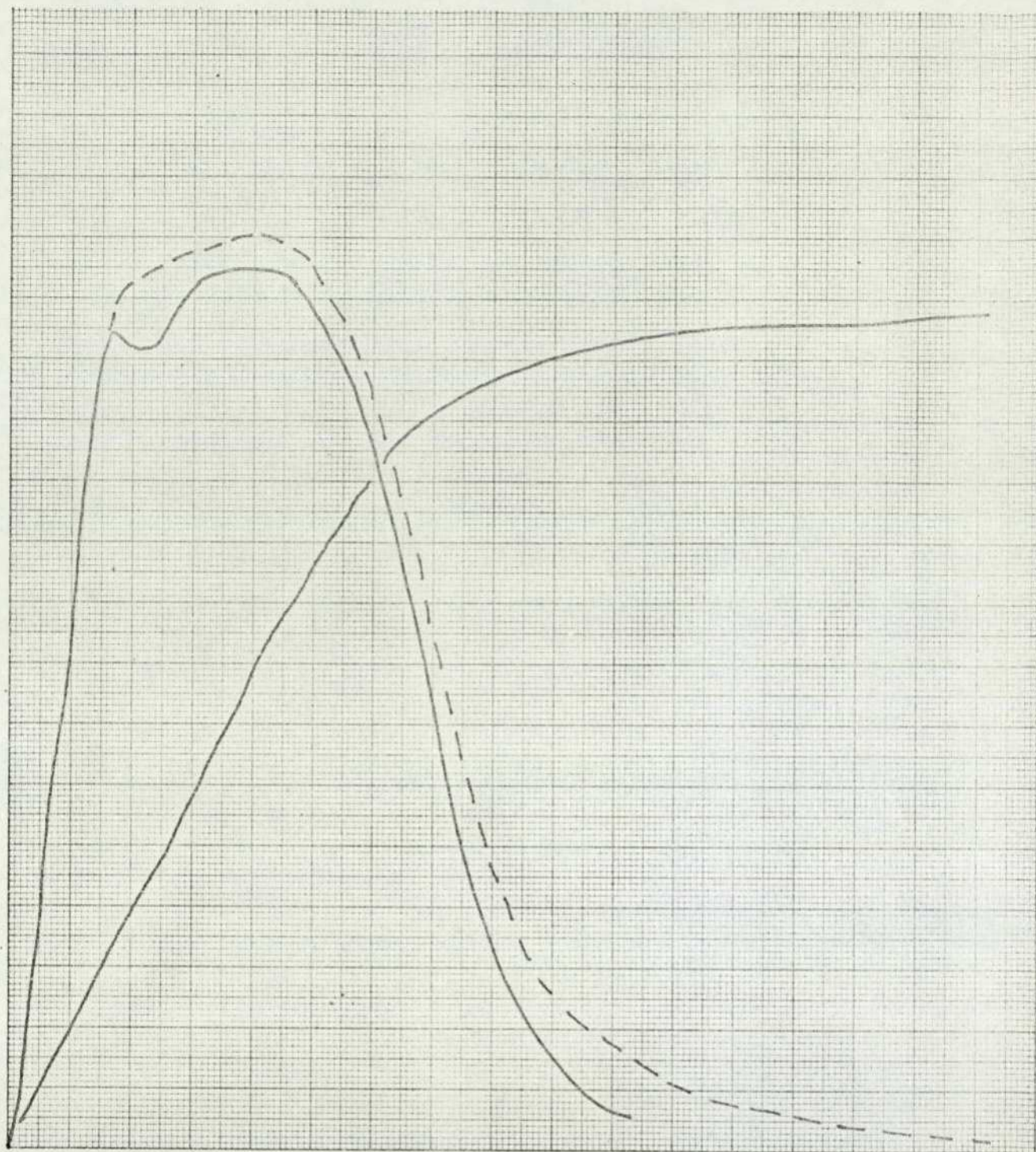


Fig. 6.26 Pulse transient shape at low and medium current densities;
 .05% and .26% of S.C.L. at 1kv

(a) ON step, .26% of S.C.L. (G/2/11)

(b) low level pulse, .05% of S.C.L. (G/2/12)

(c) OFF pulse by interrupting U.V. at the level of (a), — —
 .06% of S.C.L. (G/2/13)

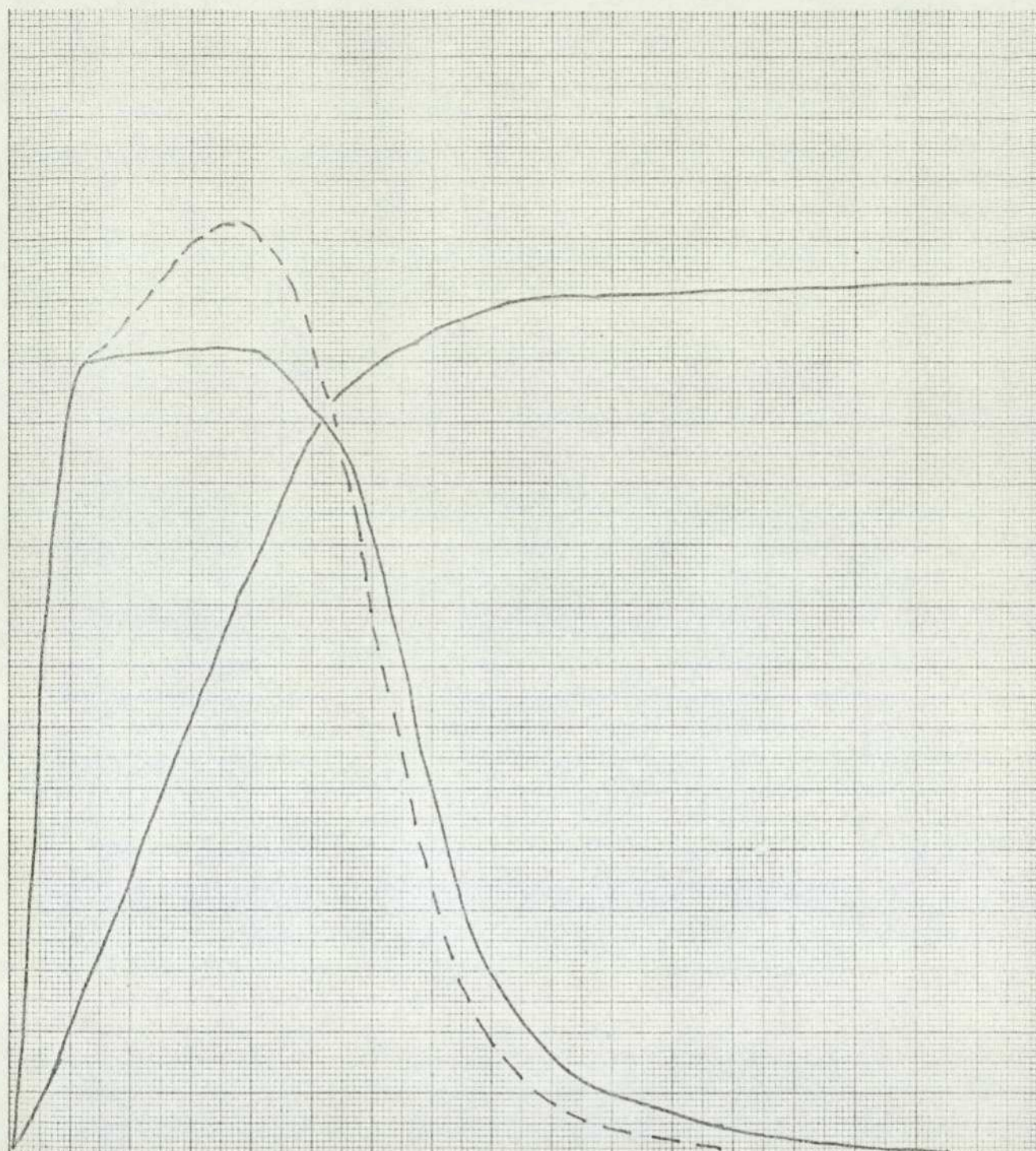


Fig. 6.27 Pulse transient shape at low and high current densities, .1% and 1.1% of S.C.L. at 1kv

(a) OFF step, 1.1% of S.C.L. (G/2/7)

(b) low level pulse, .1% of S.C.L. (G/2/9)

(c) OFF pulse by interrupting U.V. at the level of (a), .11% of S.C.L. (G/2/10)

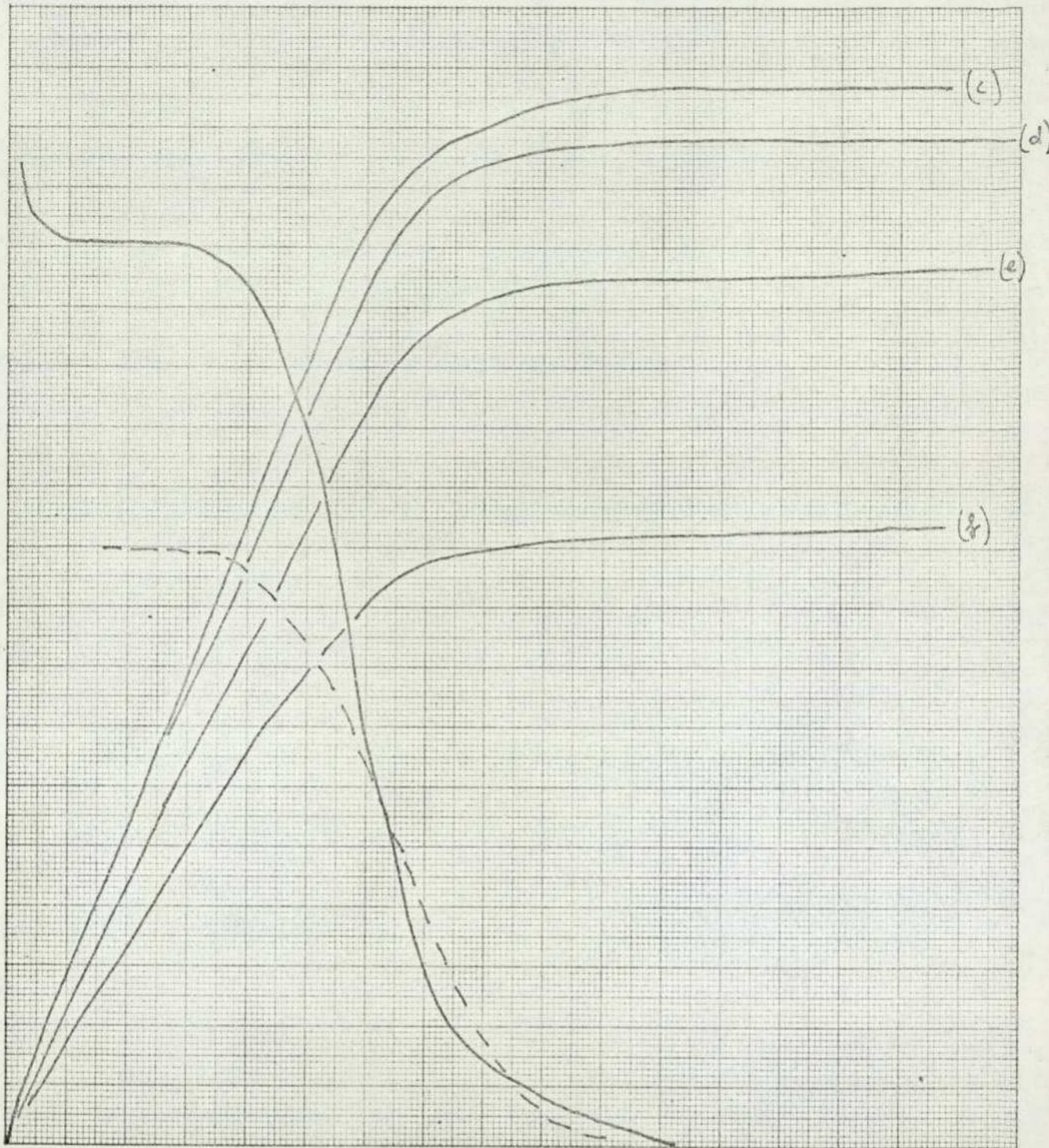


Fig. 6.28 Investigation of linearity of transients, the application of a U.V. step to part of the photocathode when the rest is ON

- (a) pulse at 2.5kv, .18% of S.C.L. (G/1/2) ———
- (b) " " 1kv, .002% " " (G/3/11) — —
- (c) ON step at 1kv, .33% " " (G/3/21)
- (d) centre portion ON at 1kv (G/3/18)
- (e) edge " " " " (G/3/24)
- (f) OFF step at 1kv, .76% of S.C.L. (G/3/23)

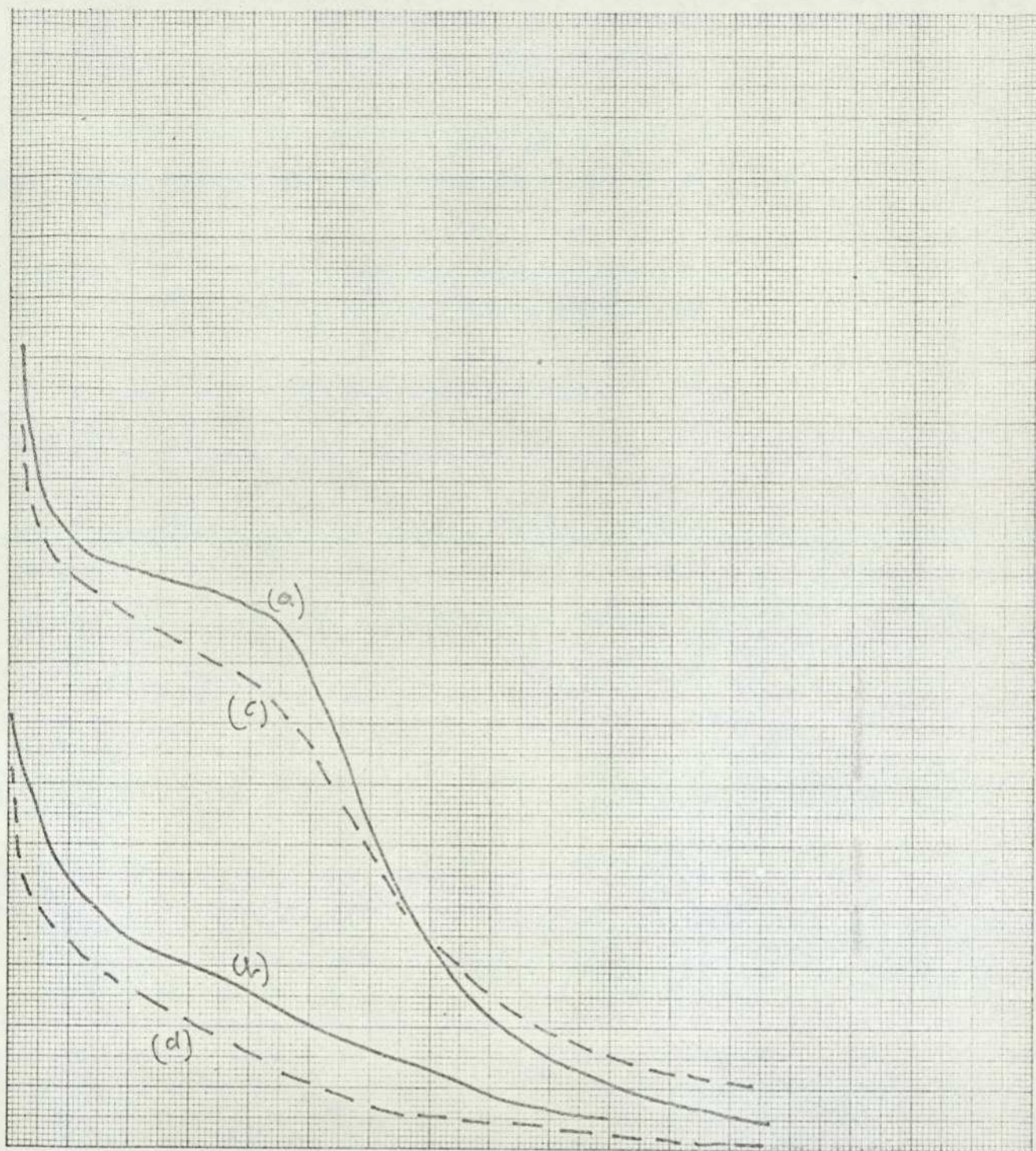


Fig. 6.29 1kv forward and reverse polarity pulse transients

(a) normal polarity, .6% of S.C.L. (C/5/13)

(b) reverse polarity, same U.V. intensity as (a) .19% of S.C.L. (C/5/14)

(c) normal polarity, OX7 filter, .5% of S.C.L. (C/5/11)

(d) reverse " " " , same U.V. intensity as (c),
OX7 filter, .12% of S.C.L. (C/5/12)

CHAPTER 7

DISCUSSION

7.1 DISCUSSION OF THE EXPERIMENTAL CURRENT TRANSIENTS

7.1.1 Variation between liquid samples

The pulse current transients, apart from those of samples K, L and M, exhibit decay tails of various shapes and time duration. The exceptional samples show features typical of bulk ionisation and these will be discussed in the next section.

This tail effect seems to be a consequence of liquid impurity and hence of liquid preparation technique, rather than of the type of photocathode, or the amount of liquid degassing. The most important evidence for this is the extra long tail of sample J, which was reprocessed sample I.

7.1.2 Bulk Ionisation

Samples K, L and M yield transients which seem to be the consequence of a bulk carrier generation, for the shape of the pulse and step transients compare well with the curves of section 2.2. The cause of this has been suggested in section 5.6.2.

When the risetime of the measuring apparatus was sufficiently fast, and the time duration of the U.V. was sufficiently short, the other liquid samples gave transients which had a sharp initial drop (e.g. 6.29).

This could be due to either bulk carrier generation or the trapping of faster ionic carriers into slower ones. Further insight can be gained from the transients in fig. 6.29, which are the forward and reverse polarity transients for the same U.V. intensity. It would be reasonable to expect the size of the photoinjected current from the stainless steel cathode to be different from that from the thin film, but contribution by bulk carrier generation to be the same.

As the current density was low, the shape of the photoinjected current transient should be given by the difference in the above transients. This is shown in fig. 7.1, together with its parent transients.

The transient exhibits an impressively flat top, apart from a small initial drop of doubtful consequence. This confirms that the initial drop in the other transients is a consequence of bulk carrier generation. It also implies that the stress distribution across the gap is constant (section 2.6), that liquid motion is not occurring at this current level, and that the effective mobility of the carriers is constant.

7.1.3 Linearity of the transport mechanism

Although each sample has a characteristic transient shape, it would appear that the phenomenon is linear in each case, because the step transient matches the shape of the pulse transient. It was also found that, providing the current density was low enough, the transient shapes were also not a function of voltage or current level.

The transients in fig. 6.28 for sample G, show that the phenomenon is not a function of electrode area, voltage, current density (if sufficiently small); that the OFF match the ON transients and that the step is the integral of the pulse transient. Such linearity is compatible with the charge transport of carriers with a range of effective mobilities. The possibility that surface trapping is occurring (section 2.5) can be discounted because the transients have a fast initial rise (where shown).

7.1.4 Near S.C.L. current transients

The transients of figs. 6.18 to 6.27 show distortions from the low level transients. The important question is whether this is due to S.C.P. effects or liquid motion. It is impossible to prove that

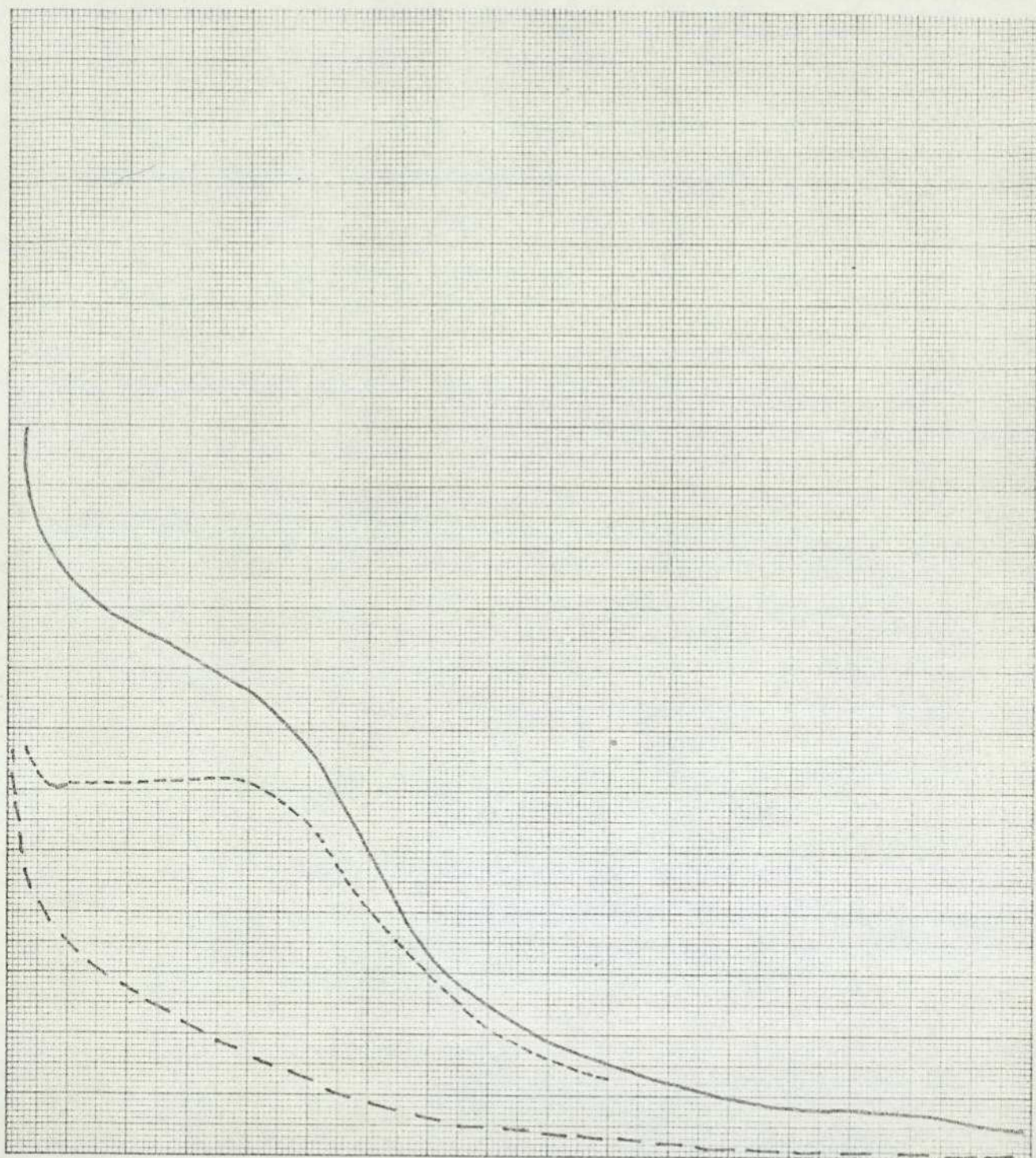


Fig. 7.1 Elimination of bulk ionisation from transients

(a) normal polarity ———

(b) reverse polarity — — —

(c) difference of the two - - - -

liquid motion is occurring from the shape of transients, this can only be inferred.

To consider this, the hallmarks of step injected S.C.P. transients must be examined. Fig. 4.22 shows S.C.P. transients for step charge injection rates of .1, .5 and S.C.L. There are two major points to be noted.

The first is that S.C.P. effects only occur if the final current is in excess of 1. When this does occur, the slope of the curve changes from an initially large value to a smaller one. Conversely, liquid motion would take time to build up and hence the slope should increase.

The transients of samples C and G can be considered with these points in mind. The distorted transients have an increasing slope indicating that liquid motion is occurring. Additional evidence can be gained from fig. 6.18, where the peak of the transient occurs in approximately a 1/3 of the usual time. The maximum change due to S.C.L. effects is .787 of the normal transit time. The curve further has a peak current level of 148% compared to an S.C.L. maximum of 136%. Finally fig. 6.20 shows that the OFF transient occurs in a shorter time than the ON transient and is more linear.

It is possible to make an assessment on the current densities at which such distortions occur.

For sample C, the transients suggest that this is less than 41% at 100V, 46% at 250V, 14% at 1kv and 14% at 2.5kv.

It is possible to be a bit more precise with the transients of sample G because they have less of a tail than those of C. Also the peak in the OFF pulse transients, in figs. 6.23 to 6.27, when compared to the ON transients, serve to give a measure of the magnitude

of this motion. The currents are too small for this peak to be a consequence of S.C.P. effects. The figures for G are that the onset of motion is at less than 4% at 250V, about 1% at 500 (cf figs. 6.24 (a) and (b)) and .26% at 1kv.

This decrease with increasing voltage is analogous to the figures for Allen and Moreau's theory (section 6.3) although their theory is applies to steady state conduction.

Fig. 6.22 shows the corresponding transients for pulse injection, which suggest that the effect of liquid motion on pulse mobility determination is negligible. This confirms the results of Buttle (1969).

7.1.5 Effect of temperature

If the tail effect is a consequence of an effective spread of carrier mobilities, then the whole transient (including the tail) should shrink or stretch with a change in temperature. The transients in figs. 6.8 and 6.9 reveal that this is so and the change in "mobility" is compatible with that expected from the activation energy of the liquid viscosity.

7.1.6 Evaluation of the range of mobilities

Earlier in this thesis the author has been rather critical of the way previous workers have produced elaborate theoretical explanations for features of their transients. It is perhaps unfortunate that they devoted so much effort to such discussions and not to improving their inadequate experimental techniques: for this tendency has caused much duplicated effort and unnecessary controversy. An example of this is Bloor's thesis (1970) which provided a good investigation into the nature of the measured distorted transients but produced invalid conclusions because it was based on poor experimental measuring methods.

With such thoughts in mind, the present author is reluctant to elaborate on the physical nature of the explanation of the "tail effect" in terms of an effective spread in carrier mobilities. This would only be worthwhile when experiments have been performed on slow carrier mobilities in "ultra pure" liquids which have a controlled quantity of known charge scavenger added. For it must be remembered that the explanation has not been positively proved but stands in the absence of any valid alternative. Other theories such as liquid motion, effects of bulk ionisation, non-uniform stress distribution and changes in carrier mobility during transit have been dismissed in

7.1.2. An additional explanation is that poor charge neutralisation at the anode could cause a build up of charge, which could in time distort the transients. This would require the ON and OFF transients to differ in shape, and hence is disproved by the results of 7.1.3. These show the ON and OFF transients to be compatible and that the transport mechanism is independent of time.

However it is of interest to determine the mobility distribution, $f(\mu)$, necessary to produce the measured transients. The following analysis requires that all of the carriers have equal charge.

Assume $f(\mu)$ is the distribution of mobilities

$f(\tau)$ is the distribution of transit times

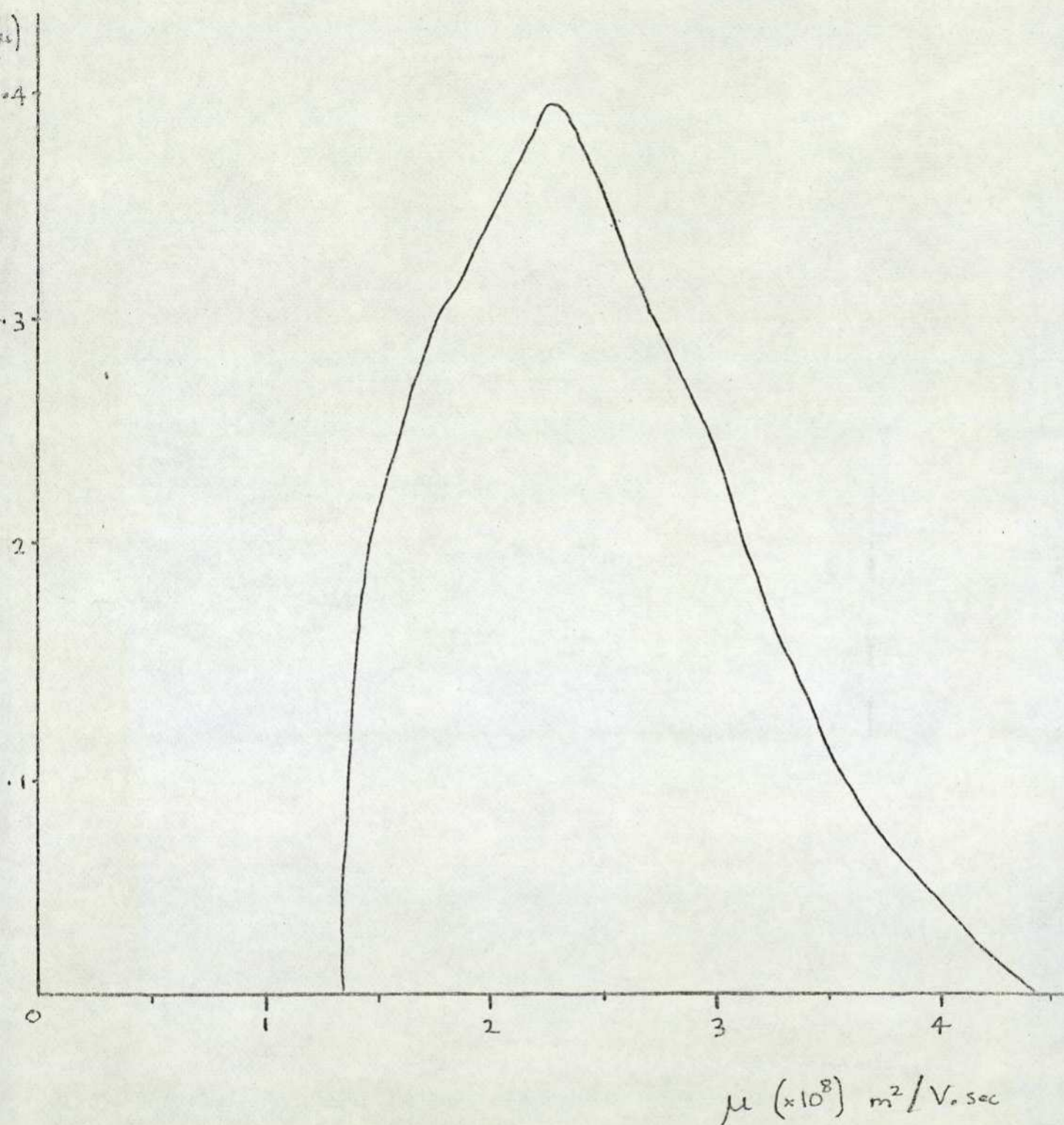


Fig. 7.1c. Mobility distribution of carriers for sample C (derived from current transient of Fig. 7.1a.)

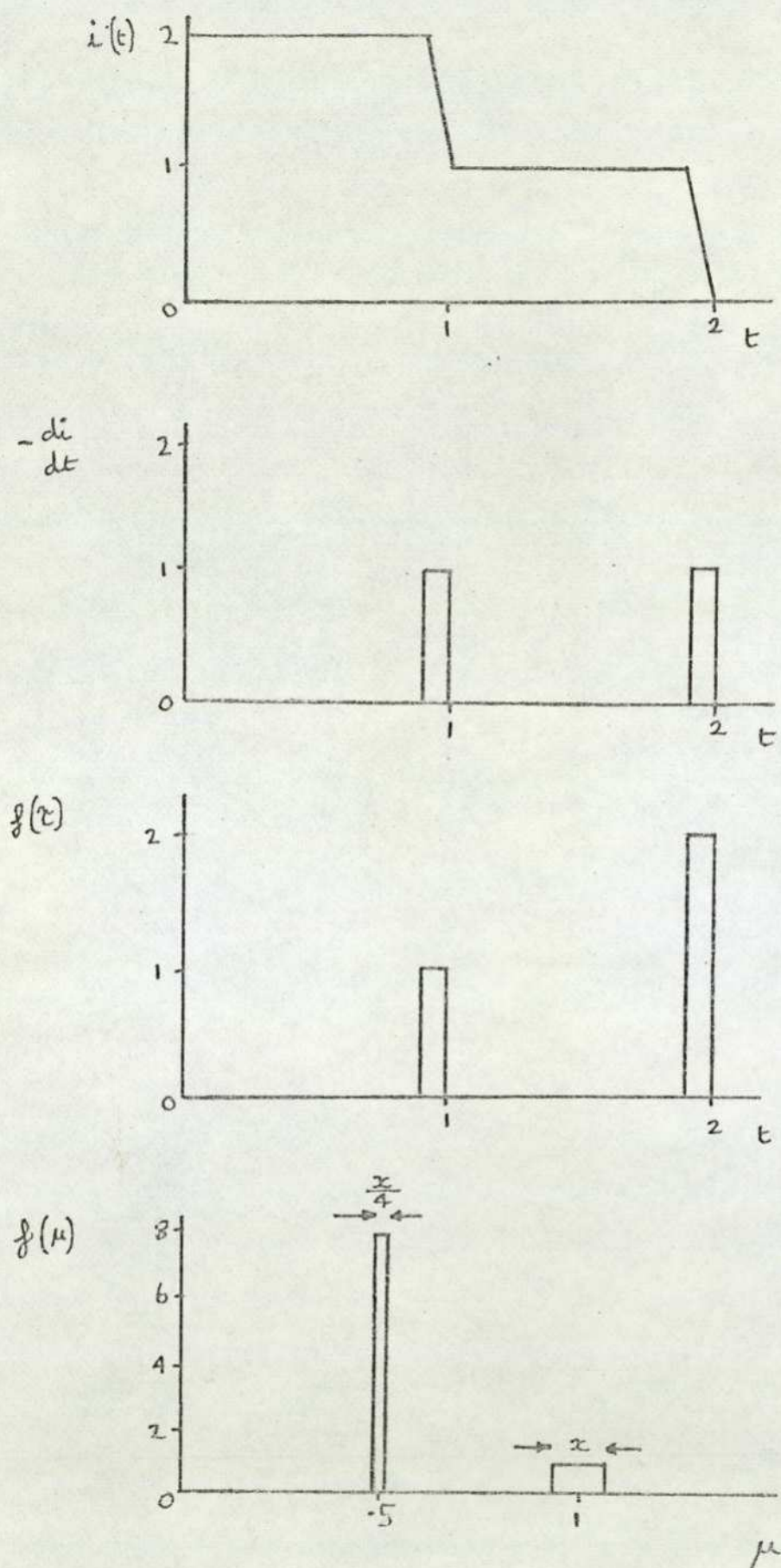


Fig. 7.1b. Illustration of the derivation of a mobility distribution from a current transient.

It is interesting to compare this with the pulse and step current transients for sample C in fig. 6.3. A comparison reveals that it would be very difficult to reach the correct mobility distribution, owing to the masking effect of bulk ionisation. However if Bloor's method of working out the mobilities μ_z , μ_i and μ_m is applied to the step transient (cf. fig. 3.16); then the following values are obtained

$$\mu_z \text{ is } 4.9 \times 10^{-8} \text{ m}^2/\text{V.sec}$$

$$\mu_i \text{ is } 2.85 \times 10^{-8} \text{ m}^2/\text{V.sec}$$

$$\mu_m \text{ is } 1.71 \times 10^{-8} \text{ m}^2/\text{V.sec.}$$

Most previous workers use the time at which the pulse current has fallen to half its peak value and this results in a mobility of $2.71 \times 10^{-8} \text{ m}^2/\text{V.sec.}$

The actual range of numerical values is not so important as the point that they are dependent on the method of liquid purification. The computer model could not be used to produce the mobility distribution from the current transient. However its versatility could be employed to calculate the S.C.P. transients which would result from a certain distribution of carrier mobilities. This could be achieved by running the model several times in parallel for a single carrier, each with a different mobility, and using the sum of the individual charge distributions to produce the new overall field distribution. This illustrates the powerful adaptability inherent in the iterative method. Such an application would not be easy, if at all possible, by analytical means.

7.2 COMPARISON WITH PREVIOUS RESULTS

The discussion in the previous section has presented the view that the tail effect is due to a range of carrier mobilities. This is not an unreasonable finding, as there is no reason why there should be a single discrete carrier. This would explain the range of mobilities reported in the literature (section 3.5). It might well be that the major ionic carriers, in the absence of grosser impurities, is, in the case of the -re carrier, a oxygen ion and a hexane ion for the +re ion; with respective mobilities of about 10 and $5 \times 10^{-8} \text{ m}^2/\text{v.s.}$

The shape of the transients is very similar to and, compatible with the curves of Le Blanc (fig. 3.14), Buttle (figs. 3.25 to 3.27), Bloor (figs. 3.17 to 3.22) and Rahman (fig. 3.24).

Additional evidence in favour of this theory is provided by reported observations of Chong and nuishi (1960), Allen and Hummel (1966), Belmont and Secker (1973) and Gzowski (1962) who all report the effect of impurities. Fig. 7.2 from Gzowski's paper shows how the addition of ethyl alcohol alters a linear ramp rise into a distorted curve.

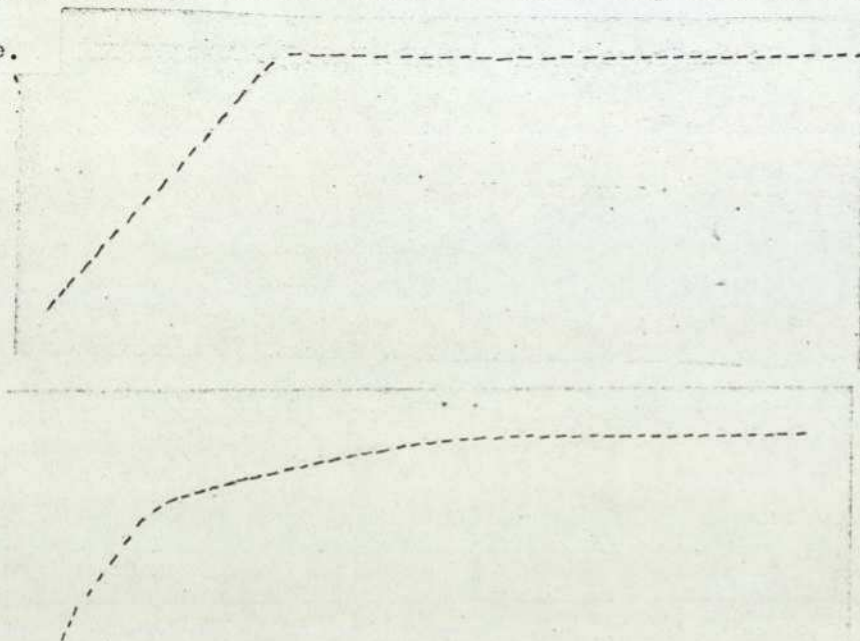


Fig. 7.2 Effect of addition of ethyl alcohol on transients (after Gzowski)

A comparison of the pulse transients of samples K, L and M with the other samples presents a good analogy to the similar work in solids shown in fig. 2.19. This provides support for the interpretation of section 7.13 in terms of bulk ionisation.

The work of Gray and Lewis (1965), cited in section 3.7, proved that liquid motion can occur in conditions similar to those of this experiment. Several other authors have published transients with an increasing slope and hence suggestive of motion. These include Bloor (fig. 3.22), Rahman (fig. 3.24) and Minday et al (fig. 3.1). This latter transient also exhibits a faster OFF transient which is less distorted than the ON transient.

Fig. 7.3 shows step transients in cyclohexane, measured by Silver et al (1968) obtained with a tunnel cathode.

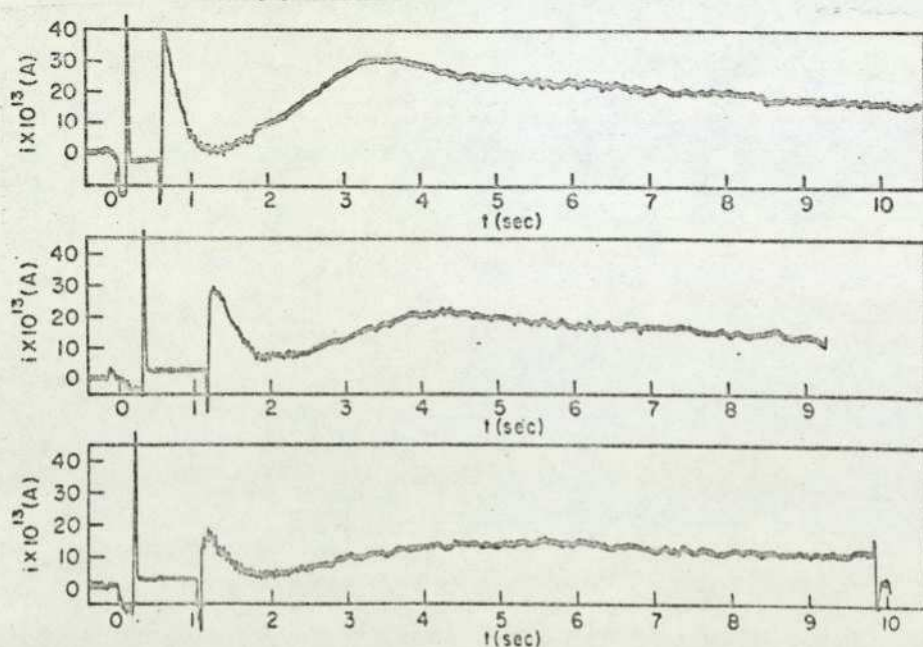


Fig. 7.3 High level step transients in cyclo hexane (after Silver et al)

The authors interpret the distortion in terms of S.C.P. effects and give a mobility figure of $2.5 \times 10^{-8} \text{ m}^2/\text{v.sec}$; the gap width was .45 cm. and the applied voltage was, 144V. However, the discussion in 7.14 would suggest that liquid motion is the main factor.

7.3 A RECENT PAPER ON THE TAIL EFFECT IN SOLIDS

At this point, it is interesting to compare the latest contribution to this question of long tailed transients, in the field of solid dielectrics.

Fig. 7.4 shows some transients in different materials used by Fleming (1974) as the basic material for his paper.

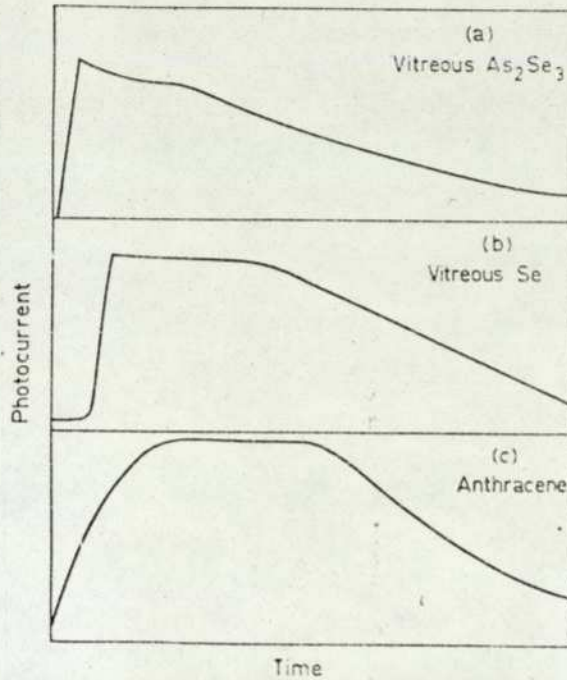


Fig. 7.4 Transients used in Fleming's paper

The author notes that all the transients exhibit a flat top and a long tail and argues against the hypothesis given by Silver et al (section 2.5), because it would require specific values of the trapping parameters which would be unlikely to apply to all the different materials.

Fleming then considers the possibility that certain widely occurring exponential trapped space charge distributions could be the cause.

To test this theory he used numerical methods to show what the resulting transients would be like, but this did not match the experimental transients.

Finally, he argued in favour of a model involving phonon-assisted tunnelling, which it would seem would be equivalent in effect to a range of mobilities. To support this he cites the fact that such a theory is consistent with experimental results which show a mobility that is field dependent.

8. SUGGESTIONS FOR FURTHER WORK

It would seem that there is little point in continuing to do further measurements on ionic transport in liquids when there is such an apparent multiplicity of carrier types. The obvious next step is to perform measurements of the type in Chapter 5, on ultra pure liquids which have been doped with a known quantity of known impurity. This would provide valuable insight into the transport mechanisms.

Correspondingly, it would seem only worthwhile performing studies of liquid motion effects on liquids of intermediate purity. One possible technique would involve shining a continuous U.V. source on test cell; similar to the one used in this work, while using a pulsed spot of U.V. to determine the shape of the resulting current transient at various points across the other beam. It would also be interesting to perform simultaneously Schlieren measurements across the gap.

The true potential of the computer model was not really used in this study. However, it did provide an insight into S.C.P. and trapping phenomenon and acted as an insurance to cope with the possibility of very complex experimental results. However, this technique is full of potential uses and the author feels that it is a pity that it was not programmed in FORTRAN so as to be readily of use to other researchers. For numerous workers have had to resort to producing numerical methods to solve their particular problems, whereas the computer model developed here would be of general use. It does need a processor of at least the "number crunching" power of the Ferranti FM1600B computer if it is to be used in real time.

9. CONCLUSIONS

The application of the techniques of current transient analysis to n-hexane has produced the following conclusions.

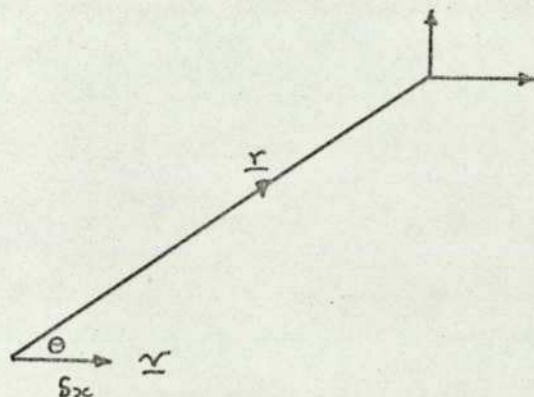
The shape and time duration of the "tail effect" seems to be a consequence of liquid preparation and hence of the impurities in the liquid. This would produce a range of ion sizes and thus mobilities. For any given sample, the spread of mobilities was found to be constant with time, field strength and current level (providing S.C.P. and possible liquid motion was avoided). A test of variation with temperature confirmed this theory, because the whole tail altered in rough accordance with the activation energy of the liquid viscosity.

The use of the best possible picoammeter techniques allowed insight into the distortion of the transients at high current levels. This distortion was interpreted in terms of liquid motion and the measurements suggest approximate current densities at which it starts having effect.

An iterative computer model of the process has been developed of proven accuracy and several applications have been given or suggested. The author feels that this is a very powerful method and that it would be very useful to the corresponding work in solid dielectrics.

APPENDIX A

PROOF OF THE RELATIONSHIP BETWEEN THE DISPLACEMENT CURRENT DENSITY $\underline{J}$ AND THE MOTION OF A CHARGE q WITH VELOCITY $\underline{v}$ AT A DISTANCE $\underline{r}$



To determine the change in D at a distance $\underline{r}$ from the charge q .

If q moves a distance S_x , this can be resolved into two components, $S_x \cos \theta$ in the direction of $\underline{r}$ and $S_x \sin \theta$ perpendicular to $\underline{r}$.

$$\underline{D} = \frac{q}{4\pi \underline{r}^2}$$

Along $\underline{r}$

$$\Delta D = \frac{q}{4\pi} \left[\frac{1}{(\underline{r} - S_x \cos \theta)^2} - \frac{1}{\underline{r}^2} \right]$$

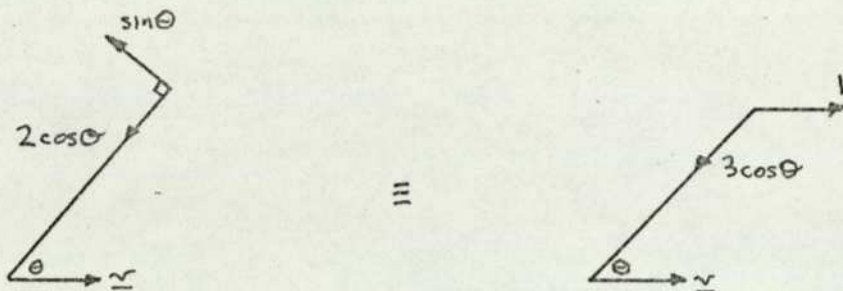
$$= \frac{q}{4\pi \underline{r}^2} \left[1 + \frac{2 S_x \cos \theta}{\underline{r}} - 1 \right]$$

$$= \frac{q}{4\pi \underline{r}^3} \cdot 2 S_x \cos \theta$$

Perp. to $\underline{r}'$

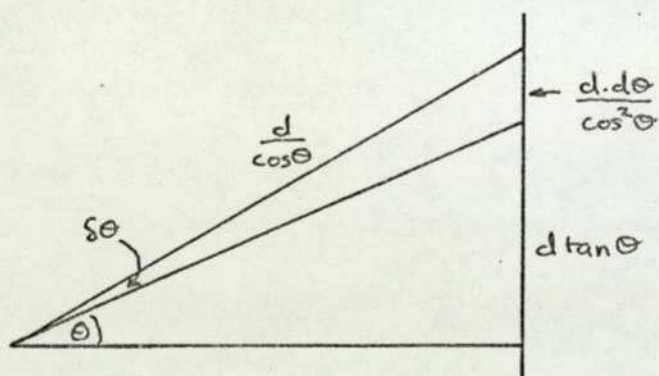
$$SD = \frac{q}{4\pi r^2} \cdot \frac{\delta x \sin \theta}{r}$$

In vectorial notation



$$\underline{J} = \frac{\partial D}{\partial x} = \frac{q}{4\pi |r|^3} \left[3(\underline{r} \cdot \hat{r}) \hat{r} - \underline{r} \right]$$

To show that the total displacement current in free space (ie. without electrodes) is zero.



$$\text{area of annular ring} = 2\pi \cdot d \tan \theta \cdot \frac{d\theta}{\cos^2 \theta}$$

$$\text{component of } \underline{J} \text{ in direction} = 1 - 3 \cos^2 \theta$$

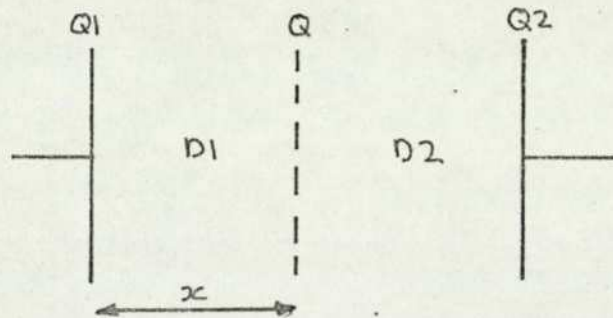
$$\text{Total } \mathbf{i} = qv \int_0^{\frac{\pi}{2}} \frac{\cos^3 \theta}{d^3} \times \frac{\sin \theta d^2}{\cos^3 \theta} \times (1 - 3 \cos^2 \theta) d\theta$$

$$= -\frac{qv}{d} \left[\cos \theta - \frac{3 \cos^3 \theta}{3} \right]_0^{\frac{\pi}{2}}$$

$$= \underline{0}$$

APPENDIX B

PROOF OF THE RELATIONSHIP BETWEEN CURRENT AND CHARGE MOTION IN A DIELECTRIC OF VARIABLE PERMITTIVITY.



Let D_1 and D_2 be the flux densities either side of a charge layer Q at a distance x from one of the electrodes.

If Q_1 and Q_2 are the charge densities on the electrodes then

$$Q_1 + Q_2 + Q = 0 \quad 1$$

$$D_1 = Q_1 \quad 2$$

$$D_2 = -Q_2 \quad 3$$

Let

$$k = -\frac{D_1}{D_2} = \frac{Q_1}{Q_2}$$

$$\frac{1}{Q_2} \text{ gives } k + \frac{Q}{Q_2} + 1 = 0$$

$$\therefore Q_2 = -\frac{Q}{k+1}$$

If the applied voltage $V = 0$

$$D1 \int_0^x \frac{dx}{\epsilon(x)} = -D2 \int_x^d \frac{dx}{\epsilon(x)}$$

$$\therefore k = \frac{\int_x^d \frac{dx}{\epsilon(x)}}{\int_0^x \frac{dx}{\epsilon(x)}}$$

$$1+k = \frac{\int_0^d \frac{dx}{\epsilon(x)}}{\int_0^x \frac{dx}{\epsilon(x)}}$$

$$\therefore Q2 = -Q \cdot \frac{\int_0^x \frac{dx}{\epsilon(x)}}{\int_0^d \frac{dx}{\epsilon(x)}}$$

Suppose the charge Q moves a distance Sx in a time St then

$$i St = \frac{Q \cdot \frac{Sx}{\epsilon(x)}}{\int_0^d \frac{dx}{\epsilon(x)}}$$

$$\therefore i = \frac{Q \cdot v}{\epsilon(x) \int_0^d \frac{dx}{\epsilon(x)}}$$

The theorem of superposition makes this solution valid for all applied voltages.

_____d _____th

_____ large _____

[REDACTED]

[illegible]

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[REDACTED]
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APPENDIX D

CONTROVERSY OF OVER CHARGE MOTION AND THE RESULTANT CURRENT

The purpose of the appendix is to chart the course over the years of the controversy concerning currents due to moving charges in the presence of static space charge.

The fallacious method of deriving the expression

$$i = \frac{E q v}{d} \quad (2.25)$$

and hence in the absence of space charge

$$i = \frac{q v}{d} \quad (2.1)$$

by the principle of virtual work has been discussed by Brignell and Evison (appendix C).

A figure which will help give a picture of the charges and flux densities behind this proof is given here (from Beck 1953).

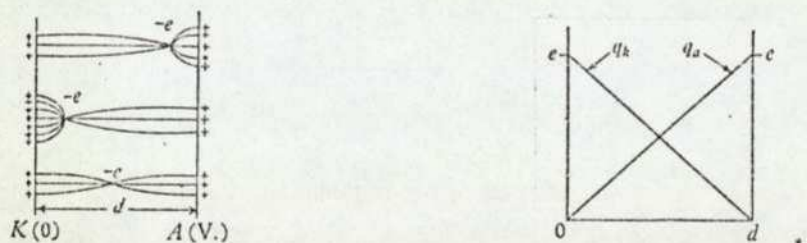


Fig. 1 Fields and currents induced by a moving charge (after Beck)

The important point is that this method gives the correct expression in the absence of space charge. This, at first sight, appears to be a satisfactory method of proving equ. 2.1. Other works which have also used this erroneous method include

Kirslow and Tuffalo (1952)

Beck (1953)

Tove and Falk (1961)

Hemingway (1962)

McMath and Martini (1970)

The method has occurred in several different fields of study and so it is likely that future workers, unaware of this earlier work, will apply this method to the situation with static space charge and result in the wrong formula. So, as discussed in 2.6., this appendix aims to make subsequent workers aware of all the relevant past work.

The bulk of this work has occurred in the field of semiconductor nuclear particle detectors. Here a diode is reversed biased which creates a depletion layer around the junction containing fixed space charge. Incident radiation produces hole-electron pairs, and the motion of these charges under the applied field will produce a current pulse. The relationship between this charge motion and the current is of importance to workers in this field.

But rather than considering each field of application independently, this summary will deal with the contributions in their chronological order. This will serve to illustrate how this controversy has continued in each of the different fields with the various workers unaware of the contributions made by others.

To commence it must be pointed out that the proofs of Shockley (1938) and Ramo (1939) did not consider the effect of static space charge. They both give the following expression

$$i = q_v \cdot E(v) \cdot v \quad 1$$

$E(v)$ is the component of the field at q_v , in the direction of v , which would result if the electrode in question were raised to unit potential and all the other were grounded. If static space charge were present, there would be doubt if $E(v)$ should alter to accommodate it.

In fact the theorem of superposition suggests that the above equation would still apply if static space charge were present, provided $E(v)$ is not altered to take account of it. However, the proof of Jen (1941) confirms this.

Jen worked out the relationship between the instantaneous power supplied to the electrodes and the charge motion. His expression is

$$\sum_{k=1}^n V_k i_k = \int (\rho \cdot \underline{v}) \underline{E} \, d\tau \quad 2$$

where V_k is the potential on the Kth electrode

i_k is the conduction current flowing to it

ρ is the space charge density

$\underline{v}$ is the velocity

and $\underline{E}$ is the field strength.

This equation, in plain words means that the sum of the instantaneous power on all the electrodes equals the rate of change of kinetic energy of the space charge. It is a quasi steady state expression which assumes that the time derivative of E is negligible.

The method derives the $\rho \underline{v}$ term into two parts. V_A due to the applied voltage V_k , and V_B due to the space charge. Correspondingly, the field can be split into E_A and E_B .

then

$$\sum V_{Ak} i_k = \int (\rho \cdot \underline{v}) \underline{E}_A \, d\tau \quad 3$$

and

$$0 = \int (\rho \cdot \underline{v}) \underline{E}_B \, d\tau \quad 4$$

Then by putting

$$V_{A1} = 1 \quad 5$$

and

$$V_{Ak} (k=2 \rightarrow n) = 0 \quad 6$$

we get the Shockly Ramo equation.

This proof was mentioned by Beck (1953), but as stated previously, he also gave the fallacious proof for equ. 2.1.

In 1961, Tove and Falk used the wrong expression. In response to this and other papers in the field of semiconductor detectors, Cavalleri et al (1963) published a paper pointing out the error. So did Gunn (1964), who provided a new proof, similar to a technique in thermodynamics of going round a closed cycle. Sadly, this elegant proof has been missed or ignored by all subsequent authors.

In 1964, Tove and Falk stated their agreement with Cavalleri et al and also gave two further proofs. One was by the use of the virtual work principle and the other by considering both the convection and displacement currents.

However, in spite of this, Golding (1966) in a review of semiconductor detectors, derived the incorrect formula.

Also in 1966 Sirkis and Holonyak proved equ. 2.1. via the incorrect expression (equ. 2.25) from energy considerations. Later on in the paper they derived it correctly by a method of calculating the induced electrode charge. This is similar to that given by Brignell and Evison (1973). Unfortunately, they did not consider what happens when static space charge is present, which would have revealed their inconsistency.

Martini and Ottaviani (1969) published a paper in response to the one by Golding which they hoped would finally resolve this problem. They provide experimented evidence for the correct expression from measurements on a p n detector with a field which increases linearly with distance.

Deshpande (1970) replied by using energy considerations to derive an expression similar to that of Jen's (equ. 3) except that the E term includes the applied field and the space charge field, thus producing the faulty equation.

Vass (1970a, b) gave support to Deshpande with the use of equ. 3 by Jen as a starting point. He also gave a proof to show that Ramo's theory derived from Jen's work is inapplicable to semiconductors with space charge.

In response to these three papers Cavalleri et al (1971) replied proving the correct expression from Jen's work and also pointing out the errors in the above papers. They finally gave an energy balance method as well.

This did not satisfy Strokan et al (1972) who gave experimental proof for the incorrect expression. Subsequently, contradictory evidence against this, was provided by Williams and Lawson (1973) in favour of the correct expression.

Finally El-Kharadly and El-Ibiary (1974) have given a proof based on the perturbation method. They also comment on faults in Strokan's theory and experimental evidence.

APPENDIX E

COMPUTER MODEL TRACE OUTPUT FOR LOW LEVEL PULSE INJECTION

CYCLE NO. 1
 NORM. TIME= +0.500021 E - 1
 TOTAL Q1= +0.000000 E + 0
 TOTAL Q2= +0.000000 E + 0
 TOTAL Q= +0.000000 E + 0
 PREVIOUS JT= +0.000000 E + 0
 CHARGE ADDED= +0.100001 E - 6 (+0.100001 E - 4)
 NEW Q1 1 +0.100001 E - 4
 EQ= +0.100000 E + 1

CYCLE COUNT=- 2

CYCLE COUNT=- 1

1(1)

D1= +0.500012 E + 1
 D2= +0.500012 E + 1
 Q(J)= +0.999984 E - 5 1
 L. 1 V12= 0 FL.= 0
 TIME = +0.199993 E + 0
 EBAR= +0.100000 E + 1
 CT DIST. = +0.500012 E + 1
 JT(CT)= +0.999967 E - 8
 CT= +0.999979 E - 6 TOT.= +0.999979 E - 6 6
 JT(D1)= +0.949957 E - 8
 JT= +0.194996 E - 7
 CTT= +0.999979 E - 6

L. 2 V12= 1 FL.= 0
 TIME = +0.199993 E + 0
 EBAR= +0.100000 E + 1
 CT DIST. = +0.400012 E + 1
 JT(CT)= +0.799992 E - 8
 CT= +0.999979 E - 6 TOT.= +0.199996 E - 5 6
 JT(D1)= +0.849991 E - 8
 JT(D2)= +0.349991 E - 8
 JT= +0.444979 E - 7
 CTT= +0.199996 E - 5

L. 3 V12= 2 FL.= 0
 TIME = +0.199993 E + 0
 EBAR= +0.100000 E + 1
 CT DIST. = +0.300012 E + 1
 JT(CT)= +0.599994 E - 8
 CT= +0.999979 E - 6 TOT.= +0.299996 E - 5 6
 JT(D1)= +0.749992 E - 8
 JT(D2)= +0.749992 E - 8
 JT= +0.654963 E - 7
 CTT= +0.299996 E - 5

L. 4 V12= 3 FL.= 0
 TIME = +0.199993 E + 0
 EBAR= +0.100000 E + 1
 CT DIST. = +0.200012 E + 1
 JT(CT)= +0.399996 E - 3
 CT= +0.999973 E - 6 TOT.= +0.399960 E - 5 6
 JT(D1)= +0.649993 E - 8
 JT(D2)= +0.649993 E - 8
 JT= +0.824943 E - 7
 CTT= +0.399960 E - 5

L. 5 V12= 4 FL.= 0
 TIME = +0.199993 E + 0
 EBAR= +0.100000 E + 1
 CT DIST. = +0.100012 E + 1
 JT(CT)= +0.200009 E - 8
 CT= +0.999979 E - 6 TOT.= +0.499981 E - 5 6
 JT(D1)= +0.549994 E - 8
 JT(D2)= +0.549994 E - 8
 JT= +0.954397 E - 7
 CTT= +0.499981 E - 5

L. 6 V12= 5 FL.= 1
 TIME = +0.199993 E + 0
 EBAR= +0.100000 E + 1
 JT(D1)= +0.000000 E + 0
 JT(D2)= +0.500006 E - 8
 CS= +0.499957 E - 5 TOT.= +0.499957 E - 5 6
 JT= +0.100490 E - 6
 CTT= +0.499981 E - 5

L. 7 V12= 6 FL.= 4
 TIME = +0.610352 E - 4
 EBAR= +0.100000 E + 1
 JT(D2)= +0.000000 E + 0
 CS= +0.610385 E - 9 TOT.= +0.610385 E - 9 7
 JT= +0.100490 E - 6

CHARGE O/P L Q1 Q2
 6 +0.599995 E - 1 +0.499957 E - 5 +0.499981 E - 5
 7 +0.699997 E - 1 +0.610385 E - 9
 TOT. Q= +0.999971 E - 7
 JT= +0.100490 E - 6
 EO= +0.100000 E + 1

CYCLE NO. 2
 NORM. TIME= +0.100004 E + 0
 TOTAL Q1= +0.500013 E - 7
 TOTAL Q2= +0.499976 E - 7
 TOTAL Q= +0.999989 E - 7
 PREVIOUS JT= +0.100490 E - 6
 EO= +0.100000 E + 1

CYCLE COUNT= 2

6(2)

D1= +0.100000 E + 2
 D2= +0.110000 E + 2
 Q(J)= +0.499969 E - 5 6

L. 6 V12= 0 FL.= 0
 TIME = +0.199993 E + 0
 EBAR= +0.100000 E + 1
 CT DIST. = +0.500000 E + 1
 JT(CT)= +0.499938 E - 8
 CT= +0.499960 E - 6 TOT.= +0.499960 E - 6 11
 JT(D1)= +0.499938 E - 8
 JT= +0.999876 E - 8

L. 7 V12= 1 FL.= 0
 TIME = +0.199993 E + 0
 EBAR= +0.100000 E + 1
 CT DIST. = +0.400000 E + 1
 JT(CT)= +0.399962 E - 8
 CT= +0.499960 E - 6 TOT.= +0.999920 E - 6 11
 JT(D1)= +0.499938 E - 8
 JT(D2)= +0.499938 E - 8
 JT= +0.239966 E - 7

L. 8 V12= 2 FL.= 0
 TIME = +0.199993 E + 0
 EBAR= +0.100000 E + 1
 CT DIST. = +0.300000 E + 1
 JT(CT)= +0.299963 E - 8
 CT= +0.499960 E - 6 TOT.= +0.149987 E - 5 11
 JT(D1)= +0.499938 E - 8
 JT(D2)= +0.499938 E - 8
 JT= +0.369946 E - 7

L. 9 V12= 3 FL.= 0
 TIME = +0.199993 E + 0
 EBAR= +0.100000 E + 1
 CT DIST. = +0.200000 E + 1
 JT(CT)= +0.199981 E - 8
 CT= +0.499960 E - 6 TOT.= +0.199984 E - 5 11
 JT(D1)= +0.499933 E - 8
 JT(D2)= +0.499938 E - 8
 JT= +0.489936 E - 7

L. 10 V12= 4 FL.= 0
 TIME = +0.199993 E + 0
 EBAR= +0.100000 E + 1
 CT DIST. = +0.100000 E + 1
 JT(CT)= +0.999904 E - 9
 CT= +0.499960 E - 6 TOT.= +0.249979 E - 5 11
 JT(D1)= +0.499938 E - 8
 JT(D2)= +0.499938 E - 8
 JT= +0.599903 E - 7

L. 11 V12= 5 FL.= 1
 TIME = +0.199993 E + 0
 EBAR= +0.100000 E + 1
 JT(D1)= +0.000000 E + 0
 JT(D2)= +0.499938 E - 8
 CS= +0.249996 E - 5 TOT.= +0.249996 E - 5 11
 JT= +0.649888 E - 7

L. 12 V12= 6 FL.= 4
 TIME = +0.610352 E - 4
 EBAR= +0.100000 E + 1
 JT(D2)= +0.000000 E + 0
 CS= +0.000000 E + 0 TOT.= +0.000000 E + 0 12
 JT= +0.649888 E - 7

CYCLE COUNT=- 1

6(1)

D1= +0.100000 E + 2
D2= +0.110000 E + 2
Q(J)= +0.499946 E - 5 6
L. 6 V12= 0 FL.= 0
TIME = +0.199993 E + 0
EBAR= +0.100000 E + 1
CT DIST. = +0.500000 E + 1
JT(CT)= +0.499915 E - 8
CT= +0.499931 E - 6 TOT.= +0.299985 E - 5 11
JT(D1)= +0.474938 E - 8
JT= +0.974842 E - 8
CTT= +0.499931 E - 6

L. 7 V12= 1 FL.= 0
TIME = +0.199993 E + 0
EBAR= +0.100000 E + 1
CT DIST. = +0.400000 E + 1
JT(CT)= +0.399939 E - 8
CT= +0.499931 E - 6 TOT.= +0.349979 E - 5 11
JT(D1)= +0.424939 E - 8
JT(D2)= +0.424939 E - 8
JT= +0.222467 E - 7
CTT= +0.999862 E - 6

L. 8 V12= 2 FL.= 0
TIME = +0.199993 E + 0
EBAR= +0.100000 E + 1
CT DIST. = +0.300000 E + 1
JT(CT)= +0.299951 E - 8
CT= +0.499931 E - 6 TOT.= +0.399963 E - 5 11
JT(D1)= +0.374962 E - 8
JT(D2)= +0.374962 E - 8
JT= +0.327436 E - 7
CTT= +0.149981 E - 5

L. 9 V12= 3 FL.= 0
TIME = +0.199993 E + 0
EBAR= +0.100000 E + 1
CT DIST. = +0.200000 E + 1
JT(CT)= +0.199970 E - 8
CT= +0.499931 E - 6 TOT.= +0.449957 E - 5 11
JT(D1)= +0.324963 E - 8
JT(D2)= +0.324963 E - 8
JT= +0.412410 E - 7
CTT= +0.199972 E - 5

L. 10 V12= 4 FL.= 0
TIME = +0.199993 E + 0
EBAR= +0.100000 E + 1
CT DIST. = +0.100000 E + 1
JT(CT)= +0.999847 E - 9
CT= +0.499931 E - 6 TOT.= +0.499946 E - 5 11
JT(D1)= +0.274969 E - 8
JT(D2)= +0.274969 E - 8
JT= +0.477394 E - 7
CTT= +0.249967 E - 5

L. 11 V12= 5 FL.= 1
 TIME = +0.199993 E + 0
 EBAR= +0.100000 E + 1
 JT(D1)= +0.000000 E + 0
 JT(D2)= +0.249969 E - 8
 CS= +0.249984 E - 5 TOT.= +0.499957 E - 5 11
 JT= +0.502387 E - 7
 CTT= +0.249967 E - 5

L. 12 V12= 6 FL.= 4
 TIME = +0.610352 E - 4
 EBAR= +0.100000 E + 1
 JT(D2)= +0.000000 E + 0
 CS= +0.000000 E + 0 TOT.= +0.000000 E + 0 12
 JT= +0.115228 E - 6

7(1)

D1= +0.110000 E + 2
 D2= +0.120000 E + 2
 Q(J)= +0.610370 E - 9 7

L. 7 V12= 0 FL.= 0
 TIME = +0.199993 E + 0
 EBAR= +0.100000 E + 1
 CT DIST. = +0.500000 E + 1
 JT(CT)= +0.000000 E + 0
 CT= +0.610356 E -10 TOT.= +0.610356 E -10 12
 JT(D1)= +0.000000 E + 0
 JT= +0.000000 E + 0
 CTT= +0.610356 E -10

L. 8 V12= 1 FL.= 0
 TIME = +0.199993 E + 0
 EBAR= +0.100000 E + 1
 CT DIST. = +0.400000 E + 1
 JT(CT)= +0.000000 E + 0
 CT= +0.610356 E -10 TOT.= +0.122071 E - 9 12
 JT(D1)= +0.000000 E + 0
 JT(D2)= +0.000000 E + 0
 JT= +0.000000 E + 0
 CTT= +0.122071 E - 9

L. 9 V12= 2 FL.= 0
 TIME = +0.199993 E + 0
 EBAR= +0.100000 E + 1
 CT DIST. = +0.300000 E + 1
 JT(CT)= +0.000000 E + 0
 CT= +0.610356 E -10 TOT.= +0.183107 E - 9 12
 JT(D1)= +0.000000 E + 0
 JT(D2)= +0.000000 E + 0
 JT= +0.000000 E + 0
 CTT= +0.183107 E - 9

L. 10 V12= 3 FL.= 0
 TIME = +0.199993 E + 0
 EBAR= +0.100000 E + 1
 CT DIST. = +0.200000 E + 1
 JT(CT)= +0.000000 E + 0
 CT= +0.610356 E -10 TOT.= +0.244143 E - 9 12
 JT(D1)= +0.000000 E + 0
 JT(D2)= +0.000000 E + 0
 JT= +0.000000 E + 0
 CTT= +0.244143 E - 9

L. 11 V12= 4 FL.= 0
 TIME = +0.199993 E + 0
 EBAR= +0.100000 E + 1
 CT DIST. = +0.100000 E + 1
 JT(CT)= +0.000000 E + 0
 CT= +0.610356 E - 10 TOT.= +0.305178 E - 9 12
 JT(D1)= +0.000000 E + 0
 JT(D2)= +0.000000 E + 0
 JT= +0.000000 E + 0
 CTT= +0.305178 E - 9

L. 12 V12= 5 FL.= 1
 TIME = +0.199993 E + 0
 EBAR= +0.100000 E + 1
 JT(D1)= +0.000000 E + 0
 JT(D2)= +0.000000 E + 0
 CS= +0.305192 E - 9 TOT.= +0.305192 E - 9 12
 JT= +0.000000 E + 0
 CTT= +0.305178 E - 9

L. 13 V12= 6 FL.= 4
 TIME = +0.610352 E - 4
 EBAR= +0.100000 E + 1
 JT(D2)= +0.000000 E + 0
 CS= +0.000000 E + 0 TOT.= +0.000000 E + 0 13
 JT= +0.115231 E - 6

CHARGE O/P	L	Q1	Q2
11	+0.109999 E + 0	+0.499957 E - 5	+0.499946 E - 5
12	+0.119999 E + 0	+0.305192 E - 9	+0.305178 E - 9
TOT. Q= +0.999935 E - 7			
JT= +0.115231 E - 6			
EO= +0.100000 E + 1			

CYCLE NO. 3

APPENDIX F

COMPUTER MODEL TRACE OUTPUT FOR S.C.P. PULSE INJECTION (2 CARRIERS)

CYCLE NO. 1
 NORM. TIME= +0.500021 E - 1
 TOTAL Q1= +0.000000 E + 0
 TOTAL Q2= +0.000000 E + 0
 TOTAL Q= +0.000000 E + 0
 PREVIOUS JT= +0.000000 E + 0
 CHARGE ADDED= +0.100497 E + 1 (+0.100498 E + 3)
 NEW Q1 1 +0.100498 E + 3
 EO= +0.223998 E - 3

CYCLE COUNT=- 2

CYCLE COUNT=- 1

1(1)

D1= +0.112000 E - 2
 D2= +0.602612 E + 1
 Q(J)= +0.100496 E + 3 1
 L. 1 V12= 0 FL.= 1
 TIME = +0.892823 E + 3
 EBAR= +0.223998 E - 3
 CT= +0.833838 E + 1 TOT.= +0.833838 E + 1 1
 JT(D1)= +0.107381 E - 3
 CS= +0.833057 E + 1 TOT.= +0.833057 E + 1 1
 JT= +0.107381 E - 3
 CTT= +0.833838 E + 1

L. 2 V12= 1 FL.= 2
 TIME = +0.892828 E + 3
 EBAR= +0.100516 E + 1
 CT= +0.833838 E + 1 TOT.= +0.833838 E + 1 2
 JT(D2)= +0.879898 E - 1
 CS= +0.833960 E + 1 TOT.= +0.833960 E + 1 2
 JT= +0.880347 E - 1
 CTT= +0.166768 E + 2

L. 3 V12= 2 FL.= 2
 TIME = +0.892823 E + 3
 EBAR= +0.100516 E + 1
 CT= +0.833838 E + 1 TOT.= +0.833838 E + 1 3
 JT(D2)= +0.796490 E - 1
 CS= +0.833960 E + 1 TOT.= +0.833960 E + 1 3
 JT= +0.167744 E + 0
 CTT= +0.250151 E + 2

L. 4 V12= 3 FL.= 2
 TIME = +0.892823 E + 3
 EBAR= +0.100516 E + 1
 CT= +0.833838 E + 1 TOT.= +0.833838 E + 1 4
 JT(D2)= +0.715119 E - 1
 CS= +0.833960 E + 1 TOT.= +0.833960 E + 1 4
 JT= +0.239059 E + 0
 CTT= +0.333535 E + 2

L. 5 V12= 4 FL.= 2
 TIME = +0.332828 E + 3
 EBAR= +0.100516 E + 1
 CT= +0.833838 E + 1 TOT.= +0.833838 E + 1 5
 JT(D2)= +0.629749 E - 1
 CS= +0.833960 E + 1 TOT.= +0.833960 E + 1 5
 JT= +0.302032 E + 0
 CTT= +0.416914 E + 2

L. 6 V12= 5 FL.= 2
 TIME = +0.892828 E + 3
 EBAR= +0.100516 E + 1
 CT= +0.833838 E + 1 TOT.= +0.833838 E + 1 6
 JT(D2)= +0.546360 E - 1
 CS= +0.833960 E + 1 TOT.= +0.833960 E + 1 6
 JT= +0.356559 E + 0
 CTT= +0.500303 E + 2

L. 7 V12= 6 FL.= 4
 TIME = +0.521851 E - 2
 EBAR= +0.100516 E + 1
 CT= +0.217823 E + 0 TOT.= +0.217823 E + 0 7
 JT(D2)= +0.131544 E - 2
 CS= +0.217854 E + 0 TOT.= +0.217854 E + 0 7
 JT(CT)= +0.173997 E + 0
 JT= +0.536957 E + 0

CHARGE O/P	L	Q1	Q2
1	+0.999999 E - 2	+0.833057 E + 1	+0.833838 E + 1
2	+0.200000 E - 1	+0.833960 E + 1	+0.833838 E + 1
3	+0.299997 E - 1	+0.833960 E + 1	+0.833838 E + 1
4	+0.400000 E - 1	+0.833960 E + 1	+0.833838 E + 1
5	+0.499992 E - 1	+0.833960 E + 1	+0.833838 E + 1
6	+0.599995 E - 1	+0.833960 E + 1	+0.833838 E + 1
7	+0.699997 E - 1	+0.217854 E + 0	+0.217823 E + 0
TOT. Q= +0.100482 E + 1			
JT= +0.536957 E + 0			
EO= +0.255036 E - 1			

CYCLE NO. 2
 NORM. TIME= +0.100004 E + 0
 TOTAL Q1= +0.502441 E + 0
 TOTAL Q2= +0.502457 E + 0
 TOTAL Q= +0.100483 E + 1
 PREVIOUS JT= +0.536957 E + 0
 EO= +0.255036 E - 1

CYCLE COUNT= 2

1(2)

D1= +0.127518 E + 0
 D2= +0.196091 E + 1
 Q(J)= +0.333813 E + 1 1

L. 1 V12= 0 FL.= 1
 TIME = +0.734167 E + 1
 EBAR= +0.255036 E - 1
 CT= +0.212610 E + 1 TOT.= +0.212610 E + 1 1
 JT(D1)= +0.106320 E - 2
 CS= +0.198395 E + 1 TOT.= +0.198395 E + 1 1
 JT= +0.106320 E - 2

L. 2 V12= 1 FL.= 4
 TIME = +0.100000 E + 1
 EBAR= +0.275556 E + 0
 CT= +0.204297 E + 1 TOT.= +0.204297 E + 1 2
 JT(D2)= +0.801182 E - 2
 CS= +0.218500 E + 1 TOT.= +0.218500 E + 1 2
 JT(CT)= +0.453782 E - 2
 JT= +0.136127 E - 1

2(2)

D1= +0.196091 E + 1
 D2= +0.379486 E + 1
 Q(J)= +0.833813 E + 1 2
 L. 2 V12= 0 FL.= 1
 TIME = +0.104062 E + 1
 EBAR= +0.275556 E + 0
 CT= +0.149170 E + 1 TOT.= +0.353467 E + 1 2
 JT(D1)= +0.801182 E - 2
 CS= +0.833653 E - 1 TOT.= +0.227380 E + 1 2
 JT= +0.801182 E - 2

L. 3 V12= 1 FL.= 2
 TIME = +0.104062 E + 1
 EBAR= +0.442337 E + 0
 CT= +0.149170 E + 1 TOT.= +0.149170 E + 1 3
 JT(D2)= +0.333774 E - 2
 CS= +0.227319 E + 1 TOT.= +0.227319 E + 1 3
 JT= +0.163498 E - 1

L. 4 V12= 2 FL.= 4
 TIME = +0.442871 E + 0
 EBAR= +0.609100 E + 0
 CT= +0.118567 E + 1 TOT.= +0.118567 E + 1 4
 JT(D2)= +0.662732 E - 2
 CS= +0.180682 E + 1 TOT.= +0.180682 E + 1 4
 JT(CT)= +0.114896 E - 1
 JT= +0.480785 E - 1

3(2)

D1= +0.379436 E + 1
 D2= +0.562866 E + 1
 Q(J)= +0.833813 E + 1 3
 L. 3 V12= 0 FL.= 0
 TIME = +0.557144 E + 0
 EBAR= +0.442337 E + 0
 CT= +0.232275 E + 1 TOT.= +0.381445 E + 1 3
 JT(D1)= +0.833774 E - 2
 JT= +0.333774 E - 2

L. 4 V12= 1 FL.= 1
 TIME = +0.557144 E + 0
 EBAR= +0.609100 E + 0
 CT= +0.232275 E + 1 TOT.= +0.350836 E + 1 4
 JT(D1)= +0.662732 E - 2
 JT(D2)= +0.833774 E - 2
 CS= +0.413048 E + 0 TOT.= +0.221935 E + 1 4
 JT= +0.233030 E - 1

L. 5 V12= 2 FL.= 2
 TIME = +0.557144 E + 0
 EBAR= +0.775879 E + 0
 JT(D2)= +0.333774 E - 2
 CS= +0.201355 E + 1 TOT.= +0.201355 E + 1 5
 JT= +0.316401 E - 1

L. 6 V12= 3 FL.= 4
 TIME = +0.229197 E + 0
 EBAR= +0.242627 E + 0
 JT(D2)= +0.524163 E - 2
 CS= +0.126581 E + 1 TOT.= +0.126531 E + 1 6
 JT(CT)= +0.184422 E - 1
 JT= +0.103401 E + 0

4(2)

D1= +0.562366 E + 1
 D2= +0.746265 E + 1
 Q(J)= +0.833813 E + 1 4

L. 4 V12= 0 FL.= 0
 TIME = +0.380409 E + 0
 EBAR= +0.609100 E + 0
 CT DIST. = +0.304565 E + 1
 JT(CT)= +0.966024 E - 2
 CT= +0.158597 E + 1 TOT.= +0.158597 E + 1 7
 JT(D1)= +0.833774 E - 2
 JT= +0.179977 E - 1

L. 5 V12= 1 FL.= 0
 TIME = +0.380409 E + 0
 EBAR= +0.775879 E + 0
 CT DIST. = +0.204565 E + 1
 JT(CT)= +0.648832 E - 2

5(2)

D1= +0.746265 E + 1
 D2= +0.929639 E + 1
 Q(J)= +0.833813 E + 1 5

6(2)

D1= +0.929639 E + 1
 D2= +0.111304 E + 2
 Q(J)= +0.833813 E + 1 6

7(2)

D1= +0.111304 E + 2
 D2= +0.121518 E + 2
 Q(J)= +0.217815 E + 0 7

CYCLE COUNT=- 1

1(1)

D1= +0.127518 E + 0
 D2= +0.195091 E + 1
 Q(J)= +0.833032 E + 1 1

APPENDIX G

DATA OF THE TRANSIENTS

The following data applies to figs. 6.1 to 6.29. Most of the nomenclature is explained in section 6.2, with the exception of "F" which indicates that the flash tube was used. The absence of information on the U.V. duration indicates that the B, or T setting of the shutter was used. $Y_p A/cm^2$ is the Y axis current density calibration.

Fig.	Sample	days old	No.	Voltage(V)	% S.C.L.	$Y_p A/cm^2$	U.V. duration
1	A	7	72	1K	.92	.1	F
	A	1	6	"			F
2	B	3	2	"	.70	.1	
	B	3	15	"	4.7	.2	
3	C	3	2	"	1.3	.1	F
	C	5	62	"	1.2	.1	
4	D	1	3	2.5K	.09	.02	
5	E	2	1	"	.14	.04	F
	E	1	1	"	.19	.04	
6	F	1	5	"	.50	.1	
7	G	1	1	"	.11	.04	F
	G	3	13	1K	.33	.02	
8	H	2	2	"	.08	.004	
	H	1	1	"	.36	.02	
	H	2	5	"	.11	.004	
	H	2	4	"	.08	.004	
9	I	1	3	"	.01	.001	1/10
	I	1	1	"	.50	.04	
	I	2	17	"	.24	.01	
	I	2	20	"	.07	.004	
10	J	1	3	"	.035	.002	
	J	1	1	"	1.1	.04	
11	K	1	1	"	.19	.01	
	K	1	2	"	1.6	.1	
12	L	1	3	"	.12	.01	1/25
	L	1	1	"	1.7	.1	
13	M	1	1	"	.08	.004	
	M	1	2	"	.21	.02	

Fig.	Sample	days old	No.	Voltage(V)	%S.C.L.	YpA/cm ³	U.V. duration
14	N	1	1	1K	.03	.002	
	N	1	3	"	.54	.04	
15	G	1	2	2.5K	.18	.04	F
	G	2	25	250	.003	.001	
16	G	3	23	2.5K	.44	.2	1/5
	G	12	13	"	1.6	.4	1/50
17	C	4	48	5K	.08	.1	F
	C	4	1	"	.04	.04	F
18	C	5	71	100	23	.02	
	C	5	70	"	41	.02	
	C	4	32	"	119	.1	
	C	5	42	"	148	.1	
19	C	5	67	250	9.2	.02	
	C	2	33	"	46	.2	
	C	4	5	"	97	.4	
20	C	7	3	1K	7.4	.4	
	C	2	20	"	14	1	
	C	2	35	"	17	1	
21	C	3	29	2.5K	.87	.4	
	C	1	36	"	5.3	2	
22	C	5	43	250	2.6	.01	F
	C	5	40	"	14	.04	F
23	G	2	22	"	4.4	.01	
	G	2	23	"	.23	.001	1
	G	2	24	"	.31	.001	
24	G	2	18	500	1.2	.01	
	G	2	17	"	1.1	.01	
	G	2	21	"	.15	.002	
	G	2	16	"	.15	.002	
25	G	2	14	"	2.3	.02	
	G	2	15	"	.22	.002	1
	G	2	16	"	.25	.002	
26	G	2	11	1K	.26	.01	
	G	2	12	"	.05	.002	
	G	2	13	"	.06	.002	
27	G	2	7	"	1.1	.04	
	G	2	9	"	.1	.004	
	G	2	10	"	.11	.004	

<u>Fig.</u>	<u>Sample</u>	<u>days old</u>	<u>No.</u>	<u>Voltage(V)</u>	<u>% S.C.L.</u>	<u>YpA/cm³</u>	<u>U.V.duration</u>
28	G	1	2	2.5K	.18	.04	F
	G	3	11	1K	.002	.0001	1/100
	G	3	21	"	.33	.01	
	G	3	18	"		.01	
	G	3	24	"		.004	
	G	3	23	"	.76	.04	
29	C	5	13	"	.6	.2	F
	C	5	14	"	.19	"	"
	C	5	11	"	.5	"	"
	C	5	12	"	.12	"	"

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Finally, [REDACTED] [REDACTED] must be congratulated for coping with the author's spelling and handwriting.

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