

THE RESPONSE OF PURE

ORGANIC CRYSTALS

TO IONISING RADIATIONS.

A thesis presented for the degree
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by

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I.

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II.

SUMMARY.

The introduction contains introductory remarks on scintillation counters together with a brief description of the exciton theory believed to be the means of primary energy migration within organic crystals.

Part I contains a review of the data published on the relative responses of different organic crystals and the attempted correlations of these results with the molecular structures of the different phosphors. From data which may be accepted as being both reliable and suitable for comparison it is concluded that theories involving correlations between phosphor efficiencies and molecular structures may be tested only if the efficiency data has (i) been extrapolated to give results for crystals of very small thickness and (ii) been obtained at very low temperatures.

Part II contains accurate calculations based on the exciton theory proposed by Birks (1951) to account for the response of crystals to different types of ionising radiations. Satisfactory agreement is obtained with all the available experimental data in the literature.

In part III an account is given of the response of organic crystals observed by the author for low-energy

III.

α - particles, and of the similar behaviour found by Taylor et al (1951) for low-energy electrons. A possible extension of the theory described in part II, allowing for the escape of excitons through the crystal surface is given accounting for this behaviour. An exciton mean free path in anthracene, terphenyl and stilbene of about $3 - 7\mu$ is obtained.

Results were obtained with strong α - particle sources which could not be explained in terms of normal response of the phosphor used. This effect was investigated as described in part IV, and it is shown that photons are produced in air, glass and quartz when these materials are irradiated by α - particles. This luminescence was probably responsible for the "scintillations" which Richards and Cole (1951) have ascribed to thin films of various materials.

Problems requiring further investigation are indicated.

INTRODUCTION.

A scintillation counter consists of a phosphor capable of transforming an appreciable fraction of the incident particle or radiation energy into luminescent light, a photomultiplier tube for converting this light into electrical impulses, and the electronic equipment necessary for recording these impulses in a suitable manner.

The detection and counting of radioactive radiations by means of appropriately designed scintillation counters is a technique which has been rapidly developed since it was first used about six years ago (Coltman and Marshall (1947); Kallman (1947); Broser and Kallman (1947)). As a result of the rapidity with which the scintillation counter has been developed there exists to-day in the hands of the nuclear physicist an extremely powerful tool for the study of many of his problems.

The processes occurring within scintillation phosphors have received but scant attention compared to the problem of their suitability or otherwise for use in scintillation counters designed for various specific purposes. In this work it is proposed to study the response of one of the major groups of phosphors - the pure organic crystals - and the term "phosphor" or "crystal" used in this work shall imply a pure organic crystal phosphor.

The Scintillation Process.

The scintillation process is a complex one and may be divided into three stages : (I) The excitation of electronic energy resulting from the interaction of the incident ionising radiation with the phosphor molecules. (II) The propagation of this excitation energy from molecule to molecule through the crystal and (III) the final emission of part of this light as fluorescence which is detected.

The mechanism by which the excitation energy is propagated through organic crystals is not yet established with complete certainty. It is, however, not the intention of the author to enter into a discussion of the various theories. Excellent reviews of the subject have been given by Bayliss (1951) and Frank and Livingston (1949). It is assumed here that the electronic excitation energy migrates through a crystal by means of a quantum mechanical resonance process to which Frenkel (1931) gave the name "exciton migration". This phenomenon has been discussed by Pringsheim (1949) who claims that

"in a crystal the total lifetime corresponding to exciton migration becomes shorter in the same ratio as the number of identical crystal elements which take part in the resonance process becomes longer. The excitation energy does not belong to an individual element at any moment, but simultaneously to all of them; the probability of emission increases correspondingly."

An alternative definition of the exciton has been given by

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Mott and Sneddon (1948) which corresponds more closely with that of Frenkel's original paper. It is shown that the excited atom hands over its energy to a neighbour in a process which may be repeated; the place where the excitation is at any moment is called the exciton. This is the mechanism preferred by Bowen et al (1949) and Bowen (1951), who have given a formal description of the process of handing on energy from molecule to molecule as follows :

A is a molecule of the phosphor.

(I) Initiation (excitation): $A + h\nu \rightarrow A^*$.

(II) Propagation (resonance transfer): $A^* + A \rightarrow A + A^*$.

(III) Chain ending (fluorescence): $A^* \rightarrow A + h\nu'$.

The phenomenon of exciton migration has been used to account for the fluorescence emitted by mixed crystals, i.e. those containing e.g. both anthracene and naphthalene molecules; under ultra-violet excitation (Bowen et al (1949)) and under α -particle irradiation (Birks (1950a)). It has also been used by Kaskan and Duncan (1950) to interpret the effect of temperature on the lifetime of the fluorescence of solid acetone.

Agno and Cortellessa (1952) have found evidence for the emission of fluorescence within organic crystals at distances from the source several hundred times greater than the range of the α -particles used, which were those from polonium. This fluorescence is probably due to re-absorption by the crystal, with subsequent re-emission, of fluorescent

light which has previously been emitted after exciton migration of electronic energy as described above (See Part III).

PART I.

THE RELATIVE RESPONSES OF DIFFERENT ORGANIC CRYSTALS TO IONISING RADIATIONS.

Factors preventing the comparison of published results.

The various published results (Harding et al (1949) ; Gittings et al (1949); Koski and Thomas (1949); Collinge and Robbins (1950); Hofstadter et al (1950); Ravilious et al (1950); Koski and Thomas (1951); Sangster (1952)) on the relative responses of different organic crystals to ionising radiations are not able to be generally compared. There are several instrumental and experimental factors which have resulted in inconsistencies in results obtained by different observers.

These factors include (i) the response of the photo-multiplier, which may bias the comparison of scintillators which have their emission spectra at different wavelengths.

(ii) The use of different sources and types of radiation.

(iii) The purity of the crystal under investigation.

Harding et al (1949) have found that they were able to make tests of the purity of different samples of the same material by observing the scintillation responses of the samples. The effect of purity of the phosphor on its efficiency, i.e. on the size of the pulse heights produced by it, was also pointed out by Jordan and Bell (1949).

(iv) The geometry of the experimental arrangement, including the optical contact between the phosphor and the photomultiplier and the type of reflector used.

The effect of two further important factors which have hitherto been disregarded, those of crystal thickness and temperature, will be discussed below.

It is certain that a combination of these factors is the cause of the many discrepancies which have occurred between the results of different determinations of the relative efficiencies of phosphors.

Comparison of reliable results.

Both Koski and Thomas (1951) and Sangster (1952) used carefully prepared and purified crystals, and used Co^{60} γ -rays as the ionising radiation. They have also determined the emission spectra of the crystals which they used, thus enabling the observed efficiencies of the phosphors to be corrected for the spectral response of the photomultipliers used. For these reasons the author believes that their results may be compared. The only difference between their

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experimental arrangements and which could lead to different results is that Sangster has used a thickness of about 6.3 mm for all his crystals, while Koski and Thomas used crystals 1.5 mm thick. The comparison of their results was carried out as indicated below.

The observed relative efficiencies were corrected for the spectral response of the photomultiplier used in each case, (an R.C.A. 5819 by Koski and Thomas and an R.C.A. C-7140 by Sangster), using the response curves shown in figure 1. This correction had not been applied by the investigators themselves. Unfortunately Koski and Thomas did not reproduce the emission spectrum of anthracene for a 1.5 mm thick crystal, and that given by Koski (1951) was used for correcting Koski and Thomas' observed efficiency of anthracene. It will be shown later that this could have no effect on the conclusions drawn from a comparison of the results.

The results corrected in this way are given in table 1. For both sets of results the efficiencies of different phosphors are given relative to anthracene = 1.00, as anthracene is generally accepted as being the most efficient phosphor. The efficiencies found for other phosphors will now be directly comparable, since the differences arising from different geometry in Sangsters' and Koski and Thomas' experimental arrangements have been removed by equating the efficiency of anthracene in the two

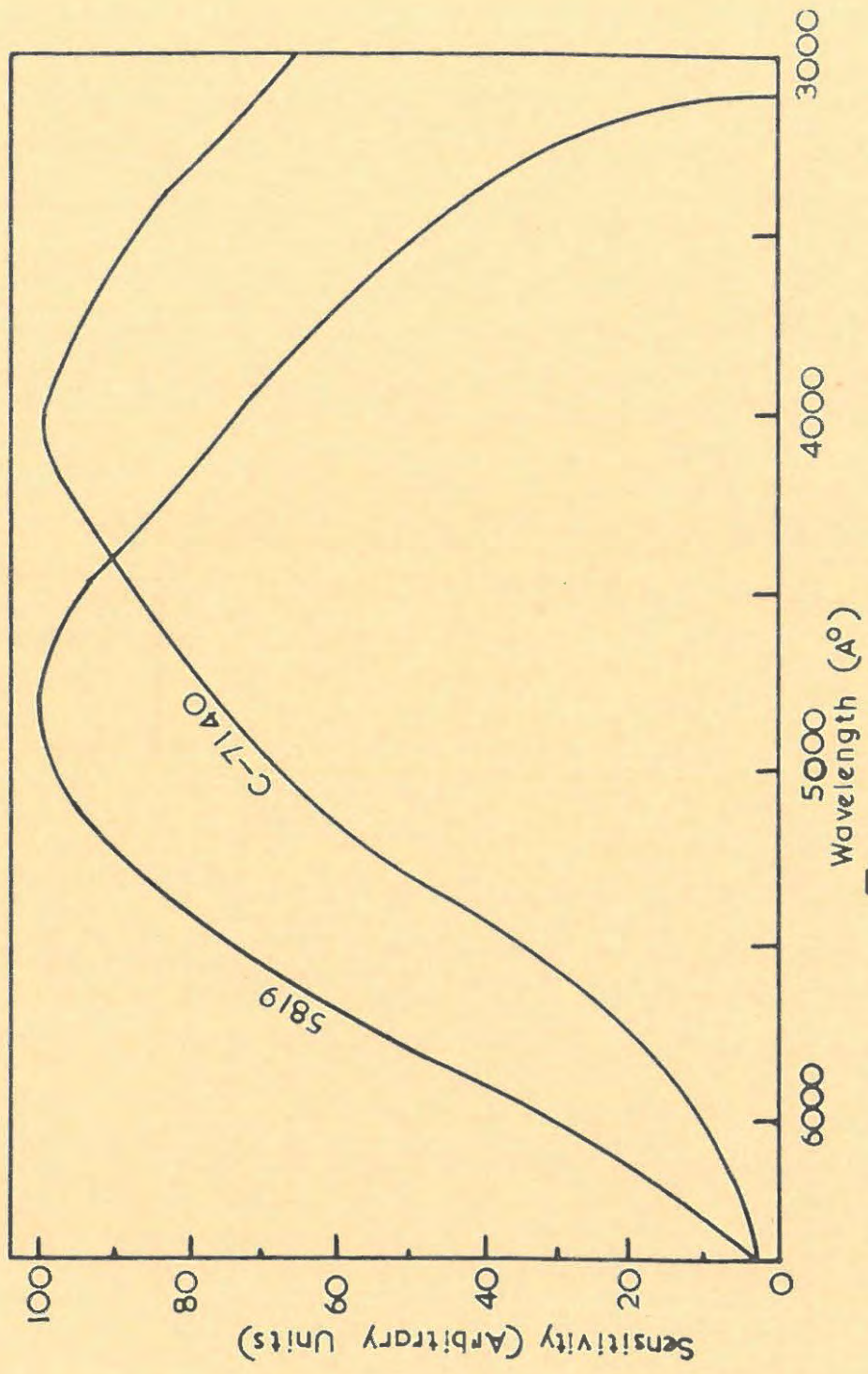


Figure 1

Table 1.

Efficiencies of different phosphors, corrected for photo-multiplier response.

Phosphor	Efficiency (rel. to anthracene = 1.00)	
	Koski & Thomas (1951) (1.5mm thickness)	Sangster (1952) (6.3mm thickness)
Anthracene	1.00	1.00
diphenyl	.32	.04
stilbene	.84	.38
diphenyl butadiene	.78	.61
diphenyl hexatriene	.13	.07
diphenyl octatetraene	.11	.06
diphenyl ethane	.59	.06
diphenyl acetylene	1.10	.22

cases.

It will be noticed that the efficiencies relative to anthracene found by Koski and Thomas appear to be higher than those found by Sangster. A graph showing the efficiency as found by Sangster, for 6.3 mm thick crystals, plotted against the factor by which the efficiency increases as the crystal thickness is reduced to 1.5 mms is shown in figure 2.

In figure 3 are shown the results of Collinge and Robbins (1950) for carefully purified crystals of thicknesses just greater than the ranges of Thorium C and C^{14} α - particles used for irradiation. The observed efficiencies obtained using an EMI 5311 photomultiplier have been corrected for spectral response using approximate spectra data for the crystal thicknesses used. Hence the results will not be as accurate as those of figure 2, but the two sets are nevertheless in general agreement.

Interpretation of results.

From figures 2 and 3 it is evident that phosphors which have low observed efficiencies for large thicknesses show, in general, greater relative increases in efficiency as the thickness is reduced. This may be explained in terms of absorption by a thick phosphor of fluorescence which would be emitted if the phosphor were not so thick.

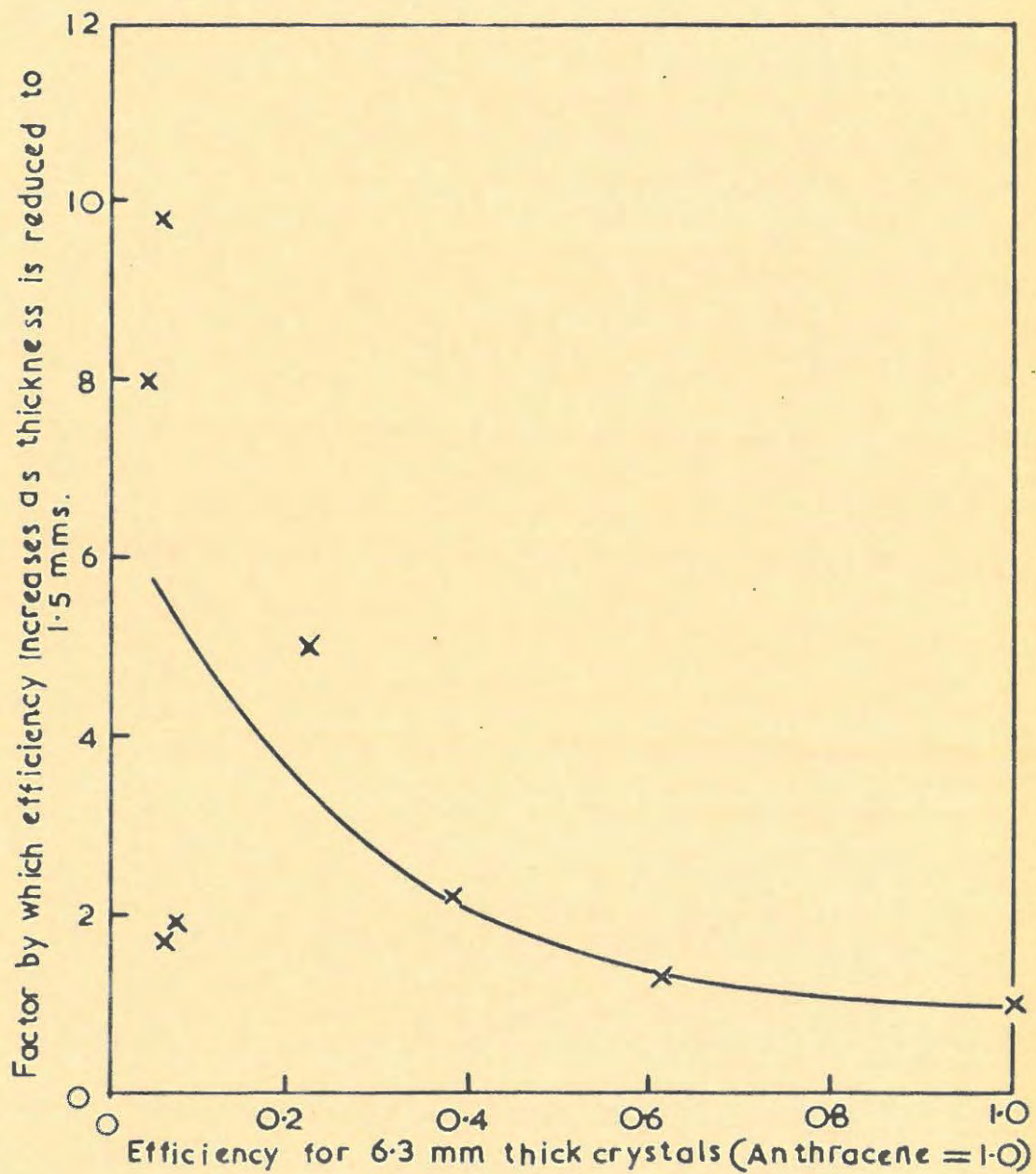


Figure 2

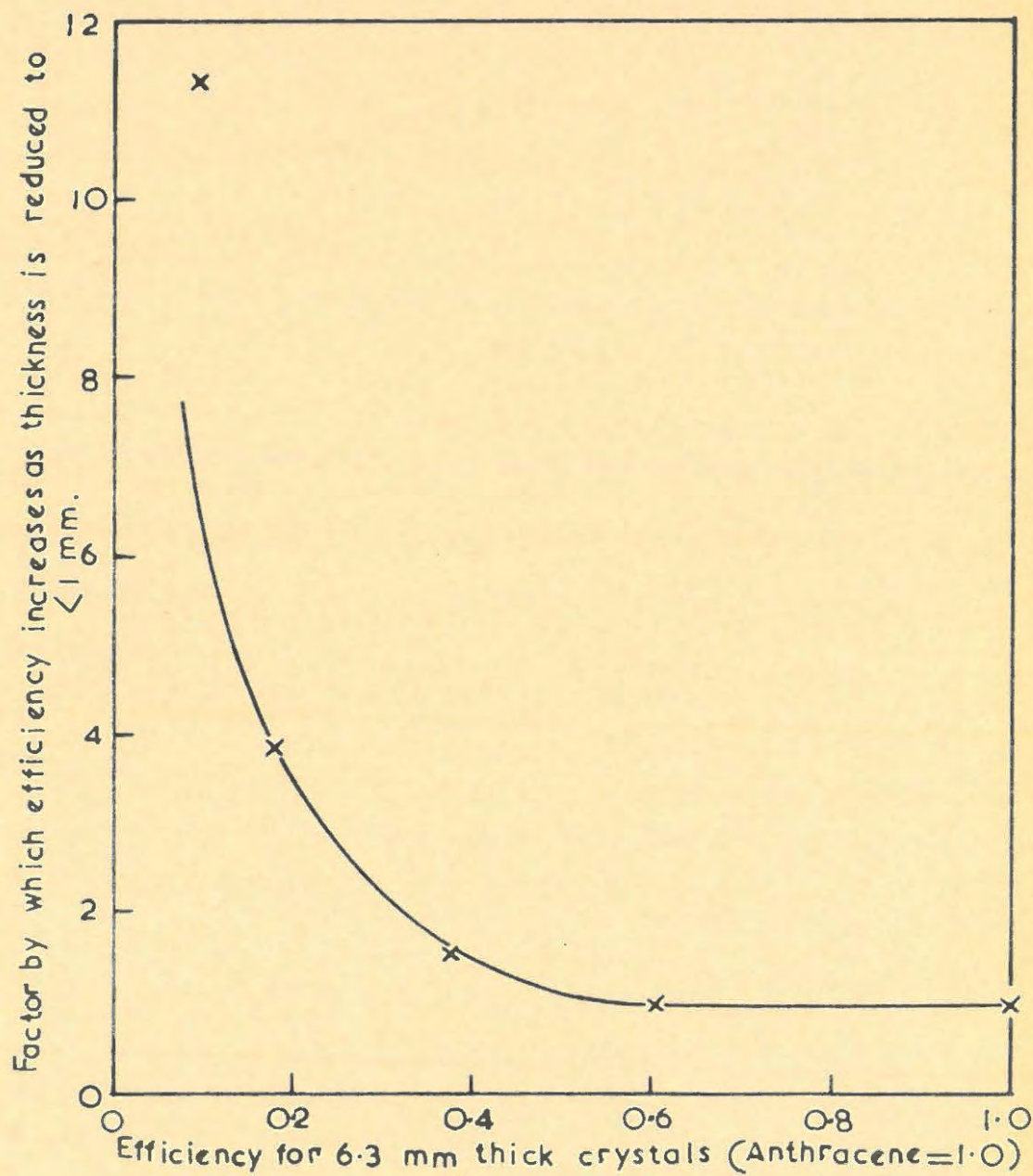


Figure 3

A phosphor which shows a relatively high efficiency at greater thicknesses would possess this high efficiency owing to the fact that little absorption has occurred within the crystal, i.e. there is not much overlap between the fluorescence and absorption spectra. Its efficiency would thus not be expected to increase by a large factor as the phosphor thickness decreases, in agreement with figures 2 and 3.

In the case of phosphors which show poor efficiencies at greater thicknesses, there are two possible reasons for the low efficiencies. Either the phosphor is a poorly efficient scintillator, and will be so for all thicknesses, or the phosphor absorbs an appreciable amount of its own fluorescence, thus reducing its efficiency to the observed low value.

It may be predicted that, owing to absorption, the emission spectra of some phosphors which possess very poor efficiencies when used as thick crystals would be very different from the emission spectra of the same phosphors used as thinner crystals. This is found to be so. The spectra determined by Sangster extend less into the ultra-violet in the case of those phosphors which he found to be poor scintillators than do the spectra found by Koski and Thomas for the same materials. On the other hand, the emission spectra of those materials which Sangster found to be efficient phosphors show no change as the thickness is

reduced. This is illustrated in figures 4 and 5, where the emission spectra of diphenyl butadiene and of diphenyl respectively have been reproduced.

There are large differences between the short wavelength ends of the emission spectra given by Koski and Thomas and by Sangster in the cases of diphenyl (which Sangster found to be the least efficient of the compounds under comparison) and diphenyl hexatriene. There are very noticeable differences in the cases of diphenyl octatetraene and diphenyl ethane (which Sangster found to possess efficiencies of < 0.1) and also of diphenyl acetylene, while there is no real difference in the case of stilbene (Sangster's efficiency = $.38$). In the case of diphenyl butadiene (Sangster's efficiency = $.61$) the spectra coincide.

It is thus believed that in the case of anthracene, which possesses a high efficiency for great thicknesses, the low-wavelength end of the emission spectrum will not alter much as the crystal thickness changes from 6.3 to 1.5 mm. This is probably the case, since Koski (1951) obtained an emission spectrum for an anthracene crystal 4 mm thick identical with that of Sangster for a 6.3 mm thick crystal. Even if some slight shift were to occur as the crystal thickness is decreased from 4 mm. to 1.5 mm., the difference in tube response would be small enough not to affect figure 2 and the interpretation above.

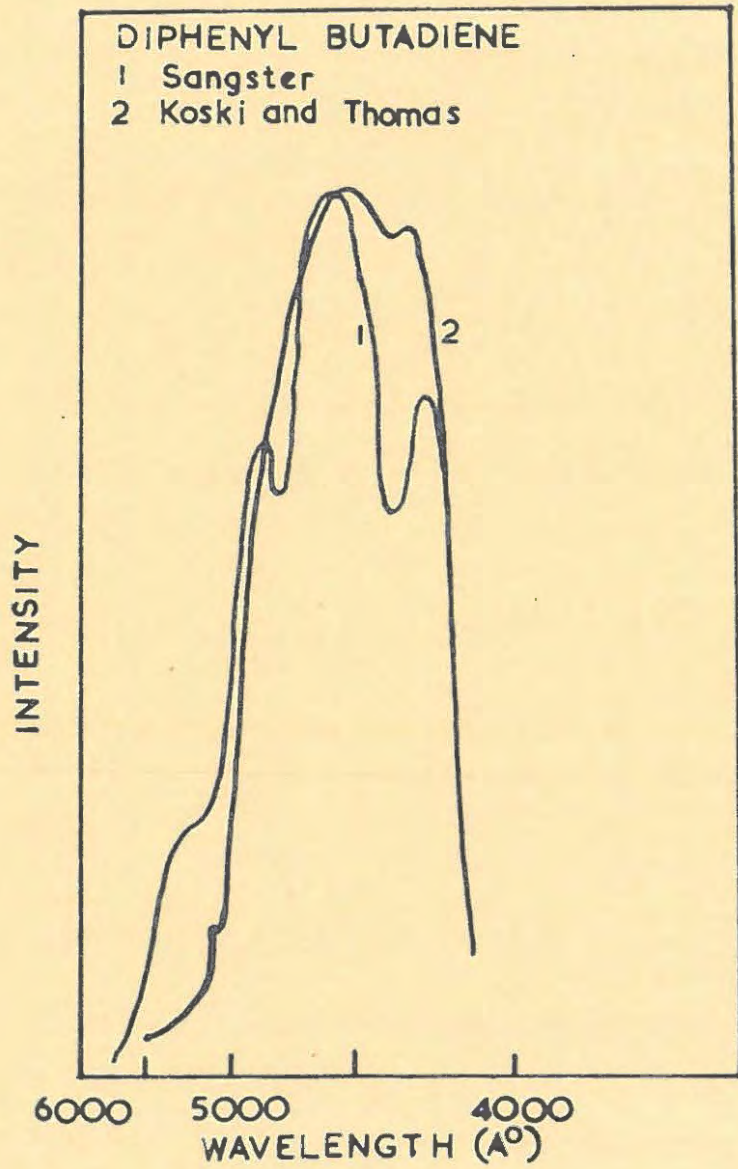


Figure 4

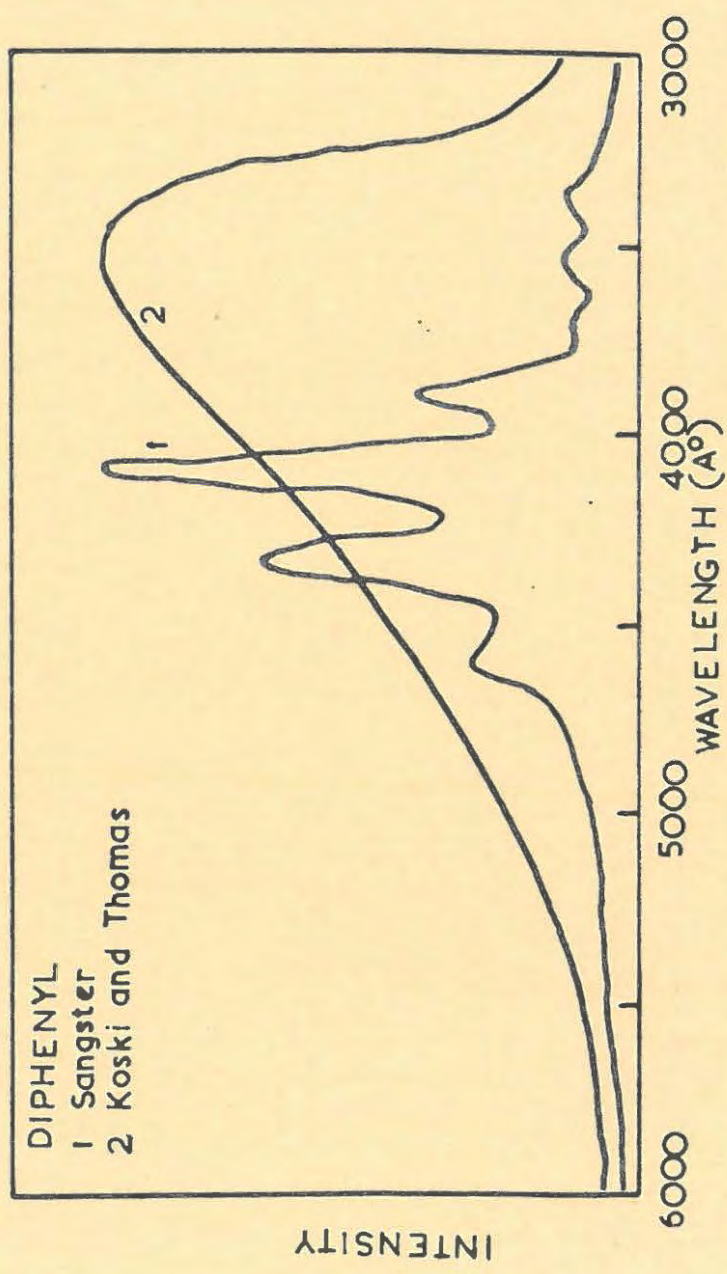


Figure 5

In support of the theory that it is possible for the absorption spectrum of the phosphor to affect the low-wavelength end of the emission spectrum, and hence the phosphor efficiency, Harrison and Reynolds (1950) have shown that when a phosphor is irradiated on the side opposite the one from which the fluorescence is emitted, the fluorescence spectrum does not extend as far into the ultra-violet as it does when the phosphor is irradiated on the same side as that from which the fluorescence is emitted. The long-wavelength end of the spectrum remains the same, however, in the two cases.

The recent experimental results of Chou (1952) indicate that 2% Terphenyl in polystyrene absorbs about 60% of its own fluorescent radiation in 20cms thickness, whereas the same concentration of anthracene in polystyrene absorbs only 15% in the same thickness.

W.A. Little of this laboratory has found that the emission spectrum of anthracene micro-crystals extends appreciably further into the ultra-violet than does the emission spectrum of powdered anthracene. Powdered anthracene, in turn, emits at shorter wavelengths than a 1 cm thick crystal, but the shift in the low-wavelength end of the spectrum is very much more pronounced as one changes from powdered crystals to micro-crystals, than from powdered anthracene to a thick crystal.

The effect of absorption within the crystal may

probably be used to explain many of the inconsistencies which exist even with carefully purified phosphors e.g. Koski and Thomas (1951) have found in agreement with Ravillious et al (1950), that diphenyl acetylene is a more efficient phosphor than stilbene, whereas Sangster has found it to be considerably less efficient.

A determination of the manner in which the relative efficiency of a phosphor changes with thickness could be carried out by comparing the efficiencies, preferably under ultra-violet irradiation, of two phosphors of different materials, but having the same thickness, and then reducing the thicknesses of both by equal amounts for further comparisons. Provided that one was an efficient phosphor such as anthracene where thickness, beyond a few mms. at any rate, had no appreciable effect, the relative variation of efficiency with thickness of the other one could be calculated.

The absorption spectra of organic crystals, which are at present not available in the literature, would enable this absorption effect to be studied quantitatively.

Temperature quenching of fluorescence.

Minarik and Drickamer (1951) and Liebson and Keller (1950) have investigated the effect of temperature on phosphor efficiency. The general conclusion is that there is a very noticeable increase in efficiency as the temperature of the crystal decreases from room temperature to temperatures of the order of -100°C . This increase in efficiency

with decreasing temperature is to be expected since heating would tend to increase lattice agitation within the phosphor and thus decrease the fluorescence.

Sangster (1952) has determined the efficiencies of various materials at 30°C and at -70°C, and has defined a mean temperature coefficient α given by

$$\alpha = \frac{(\text{efficiency at } -70^{\circ}\text{C}) - (\text{efficiency at } 30^{\circ}\text{C})}{(\text{efficiency at } 30^{\circ}\text{C})}$$

In table 2 values of α obtained by Sangster are shown. From these results figure 6 has been obtained, showing the variation of the average value of α for all the phosphors having efficiencies greater than a particular efficiency at 30°C, with the particular efficiency. It will be noticed that phosphors which have higher efficiencies at 30°C have, on the average, low values of α , while those which have lower efficiencies at 30°C have higher values of α . Phosphors which are less efficient at 30°C show greater relative increases in efficiency as the temperature is reduced than those which are more efficient at 30°C. This can only be interpreted as evidence that low efficiencies at 30°C are partly due to a quenching effect in the particular phosphor which increases with temperature. Phosphors which are more efficient at higher temperatures show less evidence of this quenching effect as this increase in efficiency with temperature reduction is, on the average,

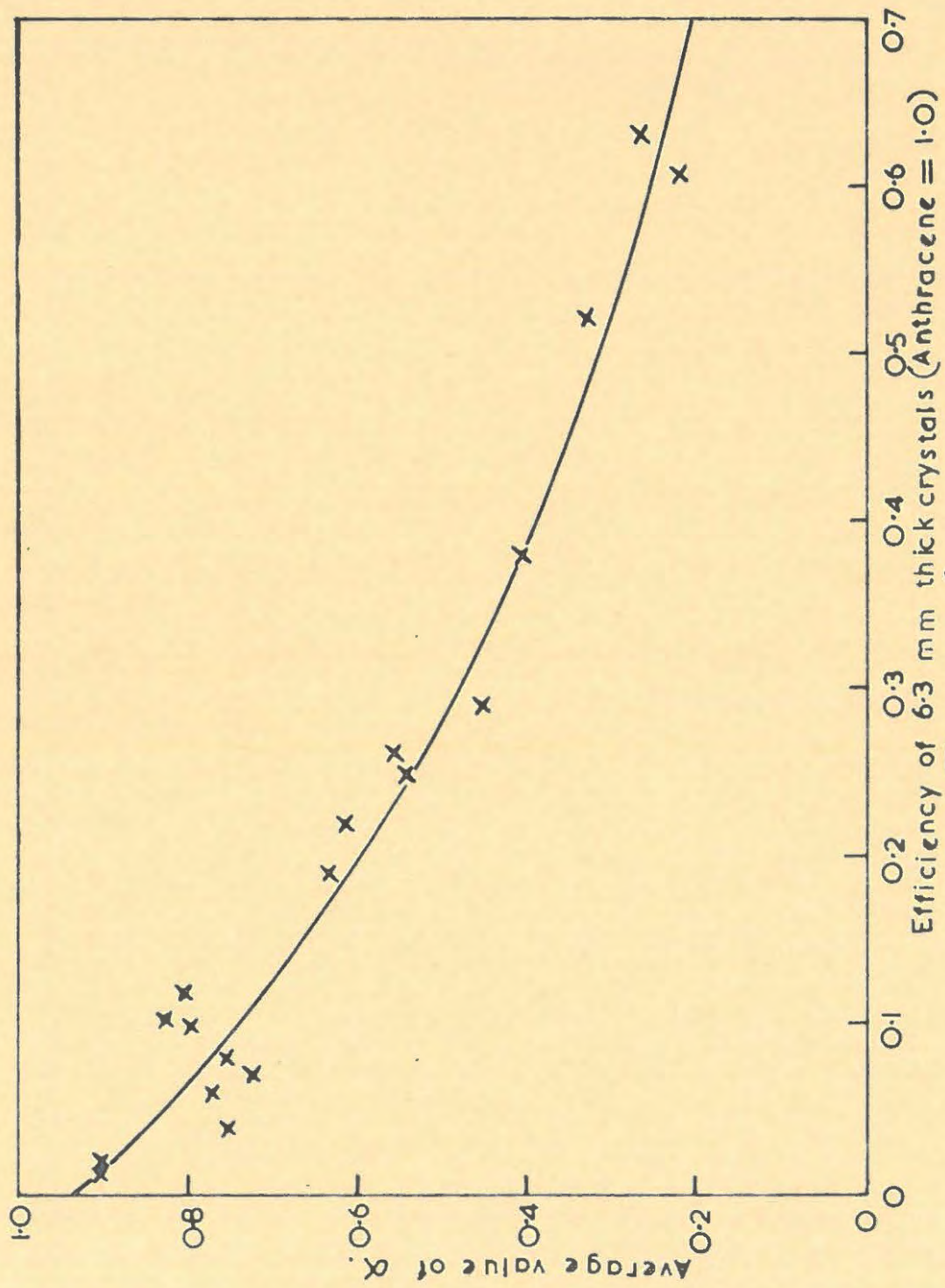


Figure 6

Table 2.

Efficiency data for organic phosphors.

Phosphor	Mean temperature coefficient, (Sangster(1952))	Efficiency relative to anthracene =1.00 (corrected for photo-multiplier response)(Sangster(1952))	Number of π -electrons per molecule.
Naphthalene	1.06	0.102	10
Anthracene	0.54	1.00	14
Phenanthrene	0.96	0.29	14
Chrysene	0.85	0.19	18
Pyrene	0.20	0.63	16
Acridine	4.20	0.02	14
Phenazine	0.17	0.003	14
Acenaphthene	0.10	0.08	10
Acenaphthylene	0.43	0.008	12
Fluoranthene	0.26	0.38	16
Fluorene	1.49	0.26	12
Carbazole	0.75	0.52	12
Diphenylene oxide	0.82	0.52	12
Bibenzothiophene	0.45	0.02	12
Diphenyl	1.00	0.04	12
O - diphenylbenzene	1.34	0.01	18
m - diphenylbenzene	0.40	0.10	18
p - diphenylbenzene (terphenyl)	0.40	0.25	18
quaterphenyl	0.06	0.77	24
dibenzyl (diphenyl ethane)	1.15	0.06	12
diphenylbutane	1.21	0.07	12
1,2 - diphenylethylene (stilbene)	0.40	0.38	14
diphenylbutadiene	0.08	0.61	16
diphenylhexatriene	0.18	0.07	18
diphenyloctatetraene	0.17	0.06	20
diphenylacetylene	1.41	0.22	16
tetraphenylethylene	3.10	0.12	26

relatively slight.

Phosphor efficiency and molecular structure.

Most of the investigations reported in the literature have been concerned with a study of factors such as phosphor efficiency, proportionality of pulse size and pulse decay constants. Only two investigators (Koski and Thomas (1951) and Sangster (1952) have carried out investigations specifically designed to make a systematic study of the influence of molecular structure on the efficiency of a phosphor. Before these investigations, however, Gittings et al (1949) had concluded from a comparison of eleven different phosphors that the scintillation property appeared to be connected with the molecular structure, since they found phenanthrene to be a very poor phosphor and its isomer anthracene a very good one. They suggested that it might be connected with the conjugate bonding occurring in stilbene. Koski and Thomas (1949) confirmed their suggestions, and pointed out that this was to be expected in view of the other correlations that had been observed between optical properties of organic molecules and the conjugation of double or triple bonds.

Bayliss (1948) has shown theoretically that the intensity of the most probable absorption of the conjugated polyenes is given in terms of oscillator strength by

$$f \approx 0.134 (2N + 1)$$

where $2N$ is the number of π -electrons in the molecule. The wave number of this most probable absorption is given by

$$\nu = (B/L^2)(2N + 1)$$

where $B = \frac{h}{8mc}$

and L is the effective length over which the π -electron can migrate.

The expression for f indicates that the absorption intensity increases with increasing number of π -electrons. A similar behaviour could be expected of the fluorescence if quenching processes were absent, since in such a case all of the energy used to excite the molecule could be re-emitted in the form of radiation.

Koski and Thomas (1951) investigated 1.5 mm thick crystals of two series of compounds. The first was composed of the symmetrical diphenyl derivatives of ethane, ethylene and acetylene. All these molecules have the same chain length. The results for this series are given in table 3, and show that, as predicted, the relative efficiency increases with increasing number of π -electrons.

The second series of compounds investigated by Koski and Thomas consisted of the diphenyl polyenes, from diphenyl to diphenyl octatetraene. The molecules of this series increase in length as one proceeds up the series. The

Table 3.

First series investigated by Koski and Thomas (1951)


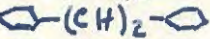






Phosphor	Structure	Number of π -electrons.	Efficiency (relative to anthracene =1.00, and corrected for photo-multiplier)
Diphenyl ethane		12	0.59
Diphenyl ethylene		14	0.84
Diphenyl acetylene		16	1.10

Table 4.

Second series investigated by Koski and Thomas (1951)




Phosphor	Structure	Number of π -electrons.	Efficiency (relative to anthracene =1.00, and corrected for photo-multiplier)
Diphenyl		12	0.32
Diphenyl ethylene		14	0.84
Diphenyl butadiene		16	0.78
Diphenyl hexatriene		18	0.13
Diphenyl octatetraene		20	0.11

results are shown in table 4, from which it will be seen that the diphenyl polyenes do not obey the predicted behaviour. Diphenyl hexatriene and diphenyl octatetraene exhibit very much lower efficiencies than expected. However, the fluorescence of both these compounds, since they have poor efficiencies at 30°C and for thick crystals, probably suffers relatively heavy temperature quenching as indicated in the last section and also relatively high absorption compared to diphenyl ethylene and diphenyl butadiene.

Koski and Thomas have suggested that the series departs from the expected behaviour owing to quenching processes which only become important with longer chain molecules. In the first series (table 3) any quenching which might be present would be the same for all three molecules owing to their equal chain lengths and thus not effect their relative efficiencies.

The exact significance, however, of molecular structure on scintillation efficiency is not as yet understood. In view of the fact that benzene rings are typical of all crystals which have exhibited the type of energy migration required for fluorescence, it would appear that these rings are necessary for energy migration. One generalisation may be made with certainty, and that is that strong fluorescence seems characteristic of resonating double bond systems, both the number and arrangements of the bonds being important.

Sangster (1952) has applied, with only moderate success, the concepts of steric hindrance and of quantum mechanical resonance to the interpretation of scintillation behaviour in terms of molecular structure. For a series of similar compounds it is predicted that the scintillation efficiency will be highest for the compounds in which exists the greatest amount of resonance between rings and other groups. Steric hindrance effects will interfere with the resonance linkage of the various groups and will also increase the vibrational freedom of the molecule by weakening the restoring forces. The greater the amount of vibrational excitation present in the molecule, the more the nuclear configuration corresponding to the maximum resonance conjugation will be perturbed, with a consequent decrease in the scintillation efficiency. However, a high proportion of phosphors are found to behave in an unpredicted and unaccountable manner.

The series diphenyl (, terphenyl () and quaterphenyl () possess efficiencies which increase as the number of electrons in each molecule increases, in agreement with the theory of Bayliss (1948). It may be predicted that quinquephenyl and sexiphenyl would probably be better than any other scintillator at present known.

Conclusions.

The fact emerges from a study of reliable data that

if any correlation is to be sought between experimentally determined phosphor efficiency values and molecular structure, allowance must be made for (i) absorption within the phosphor, by the extrapolation of measurements to very thin thicknesses, and (ii) for temperature quenching effects which are present, although not to the same extent, in each phosphor. This effect may be circumvented satisfactorily by using crystals at ~~very~~ low temperatures. Until these allowances are made a full understanding of molecular scintillation behaviour will not become possible. The efficiency values which should be used for comparison with theory would have no practical use, as phosphors are required in the laboratory to function at room temperatures and to have thicknesses consistent with stopping energetic radiations.

PART II

THE RESPONSE OF ORGANIC CRYSTALS TO DIFFERENT TYPES OF IONISING RADIATIONS.

Definitions.

The intensity of the scintillations produced in organic phosphors depends both on the energy, E (MeV), and on the nature of the incident ionising particle. The amplitude, S (Volts), of the voltage pulse from a photomultiplier operating under suitable constant conditions is

proportional to the number of fluorescent quanta produced, and hence S may be used as an arbitrary measure of the scintillation intensity.

The "fluorescent efficiency" of the phosphor is given by dS/dE (Volts/MeV) and is constant only when S increases linearly with E .

The "specific fluorescence" is the quantity dS/dr (Volts/cm. air equivalent) where r is the range of the ionising particle within the phosphor. The ranges are measured in standard cms. of air i.e. at 15°C and a pressure of 760 mms. of mercury. The specific fluorescence is proportional to the number of fluorescent quanta produced per unit path of the particle in the phosphor.

dE/dr (MeV/cm. air equivalent) is the "specific energy loss."

Speed of response.

The speed of response of a scintillation counter seems to be limited only by the length of time required for the phosphor to emit its light. It appears to be a good approximation to assume that each ionising event within the phosphor leads to a light pulse with an essentially instantaneous rise time, and a decay time which can be described essentially by a single exponential (Bittman, Furst and Kallman (1952)). This decay is characterised by a time constant τ , which is the time required for

the decay to fall to $1/e$ of its initial value. The most recent values of decay times are those given by Bittman et al (1952) and Liebson (1952), and are reproduced in tables 5 and 6. The average value of the decay constant γ is about 10^{-8} seconds for all the organic phosphors except naphthalene. This is a factor of a hundred smaller than the decay time of inorganic phosphors. Organic phosphors have in this respect a considerable practical advantage over inorganic phosphors.

Elliot et al (1950) have found that the decay time decreases by as much as 50% as the temperature of the phosphor falls from 30°C to -150°C , and that it approaches a constant value at low temperatures. The same tendency has been reported by Post and Shiren (1950) and Kelley and Goodrich (1950).

Detection of γ -radiation.

γ - rays interacting with matter undergo three principal types of interaction : Compton, photoelectric and pair production, the relative importance of each type depending upon the energy of the γ - ray and upon the atomic numbers Z of the nuclei present. For the organic phosphors the effective Z value is that of carbon, and γ - rays of energies less than a few MeV interact with these low Z value materials almost exclusively by Compton scattering, producing recoil electrons of all energies up to a maximum energy given by

Table 5.Decay times of organic phosphors (Bittman et al (1952)).

Phosphors	Time constant (secs x 10 ⁻⁸)	
	γ -ray irradiation	α -particle irradiation
Anthracene	2.28	1.65
Naphthalene	8.75	5.3
Stilbene	0.69	0.72

Table 6.

Decay times of organic phosphors (Liebson 1952)

Phosphors	Time constant (secs x 10 ⁻⁸)	
	ultra-violet irradiation	γ -ray irradiation
Anthracene	1.7	3.0
Carbazole	0.7	1.3
Chrysene	2.2	2.6
Diphenyl acetylene	0.25	0.4
Diphenyl butadiene	0.2	0.6
Fluoranthene	4.6	-
Fluorene	0.9	-
Phenanthrene	0.5	-
Quaterphenyl	0.4	0.7
Stilbene	0.3	0.8
Terphenyl	1.1	0.6

$$E_c = \frac{E_\gamma}{1 + \frac{E_0}{2E_\gamma}} \quad \text{----- (1)}$$

where E_0 = self-energy of the electron
 = 0.511 MeV.

The number of recoils reaches a maximum at greatest energy and then drops sharply to zero. By measuring the energy of the point of maximum slope in a pulse-height curve it is possible to calculate the γ -ray energy from equation (1).

The efficiency of an anthracene phosphor for detecting high energy γ -rays has been found by Lonsjo (1952) to be about 6% for 6 MeV γ -rays.

It is interesting to note that Hopstadter and McIntyre (1949) used a stilbene crystal to verify the simultaneity of the recoil electron and scattered quantum in the Compton process to within 10^{-8} seconds, and verified the angular distribution of the scattered quanta as given by the Klein - Nishina formula.

Detection of Neutrons.

Anthracene crystals have been used for the detection of fast neutrons by exposing the crystal to a neutron beam for a known time interval and then measuring the induced C^{14} activity. The more direct method first suggested by Bell (1948) is by counting the recoil protons produced in the

phosphor by the fast neutrons. He found that an anthracene crystal 1 cm. thick counted about 10% of fast neutrons.

A difficulty which arises when using this method, as pointed out by Harding (1951), is that it is usually necessary to detect the neutrons against a background of γ - rays, to which organic phosphors are very sensitive. It is not generally possible to distinguish between neutrons and γ - rays by means of a voltage discriminator because organic phosphors give larger pulses from electrons than from protons of the same energy. Greater sensitivity to neutrons relative to γ - rays may be obtained using inorganic phosphors.

Emission Spectra.

The spectra emitted by a particular organic crystal is independent of the mode of excitation. This has been shown by Koski and Thomas (1950) using α - particles, γ - rays and X- rays, and by Harrison and Reynolds (1950) for X- rays and ultra-violet light.

Proportionality between pulse height and incident energy.

One of the properties which an "ideal" phosphor might be expected to possess is that of exact proportionality between pulse height and incident particle energy. This property is in general possessed by inorganic phosphors.



(Franzen et al (1950); Taylor et al (1951)) except for α -particles of energy < 10 MeV, where the non-linearity has been investigated by Loveberg (1951). In organic phosphors, however, the number of fluorescent quanta produced by an ionising particle is in general not proportional to the energy given up by the particle, except in the case of electrons of energy > 125 keV (Hopkins (1951); Taylor et al (1951)). For all other types of particles, and for electrons of energy < 125 keV, the behaviour of organic crystals departs from the linear relation characteristic of inorganic crystals.

It has been known for several years (Broger et al (1949)) that, contrary to the measurements on inorganic phosphors, the resultant light scintillations from organic phosphors are smaller for heavy particles than for lighter particles of the same energy, e.g. Jordan and Bell (1949) observed that a 5 MeV α -particle produces a smaller pulse in anthracene than does a 1 MeV β -particle.

This phenomenon has been interpreted (Kellman (1949); Herforth and Kellman (1949)) as a saturation effect arising from the much higher specific ionisation produced by heavier particles of equal energies.

Bowen and Roser (1952) have found the fluorescent efficiency of anthracene to be the same for both electrons and high-energy μ -mesons at equal specific ionisations, which shows that the non-linearity of response of organic scintillators is caused by a saturation effect which is a

function only of the specific ionisation and which is independent of the type of particle.

In order to obtain the most fundamental significance of the response of an organic phosphor it is useful to compare the specific fluorescence with the specific energy loss, i.e. the number of fluorescent quanta produced per unit path length of the particle in the crystal with the energy lost by the particle per unit path.

This has been done by Taylor et al (1951) for their comprehensive results obtained using protons, deuterons, α -particles and electrons. The general features of their results had been predicted by Birks (1951) using the exciton theory.

On this theory the ionising particle will produce a certain number of excitons per unit path length, proportional to the specific energy loss, say $A \frac{dE}{dr}$. This excitation is potentially available for fluorescence. At the same time a quenching process, also proportional to the specific energy loss is assumed to occur. The exact nature given to this quenching process by Birks (1951) is as follows: a local concentration say $B \frac{dE}{dr}$ of "damaged" molecules is produced by the ionising particle and these damaged molecules tend to quench the fluorescence by capturing the excitons, thus preventing their capture and subsequent re-emission as fluorescence by fluorescent

molecules. A quantity k is defined as the exciton capture probability of a damaged molecule relative to an undamaged one, when the specific fluorescence will be given by

$$\frac{dS}{dr} = \frac{A \, dE/dr}{1 + kB \, dE/dr}.$$

This concept of damaged molecules has been used by Birks and Black (1951) to explain the deterioration of anthracene under intense α -particle irradiation. However, the exact nature of the quenching process within organic molecules cannot be ascribed with certainty to the production of damaged molecules, and the expression for the specific fluorescence is thus re-written

$$\frac{dS}{dr} = \frac{A \, dE/dr}{1 + B^1 \, dE/dr} \quad \text{--- (2)}$$

Where kB has been replaced by a constant B^1 , and $B^1 \, dE/dr$ represents the quenching process.

This expression for dS/dr gives at high values of dE/dr a constant maximum value, independent of the specific energy loss, for dS/dr . At low values of the specific energy loss the specific fluorescence is proportional to the specific energy loss, i.e. S is proportional to E . This is the region of linear response for organic crystals, occurring for electrons of energies > 125 keV.

Theoretical response curves.

Following the procedure of Birks (1951) the

author has calculated the response curves of anthracene for protons, deuterons, α -particles and electrons. Anthracene was chosen as it is the phosphor for which most experimental data is available for comparison with theoretical response values.

Using the data of Taylor et al, values of dS/dr and dE/dr have been obtained and substituted into equation (2), thus enabling values of A and B^1 to be calculated. A is found to be 71.9 and B^1 7.92 for anthracene. These values enable the curve of dS/dr vs dE/dr (figure 7) to be plotted using equation (2). In the same figure are shown the results of Taylor et al, and it is seen that the curve agrees very well with their results. There is, however, notable disagreement for low-energy electrons having equivalent air-ranges of < 9 or 10 mm. This disagreement will be discussed in part III.

The range-energy data of Curie (1935) for electrons, and Livingston and Bethe (1937) and Bethe (1950) for α -particles, protons and deuterons have been used to obtain the specific energy loss for particles of different ranges, r. The values of specific energy loss enabled the specific fluorescence to be calculated from equation (2) and curves plotted showing dS/dr as a function of r. These curves are shown in figures 8 - 11. Upon integration of the $(dS/dr, r)$ curves, which was done by planimeter, the

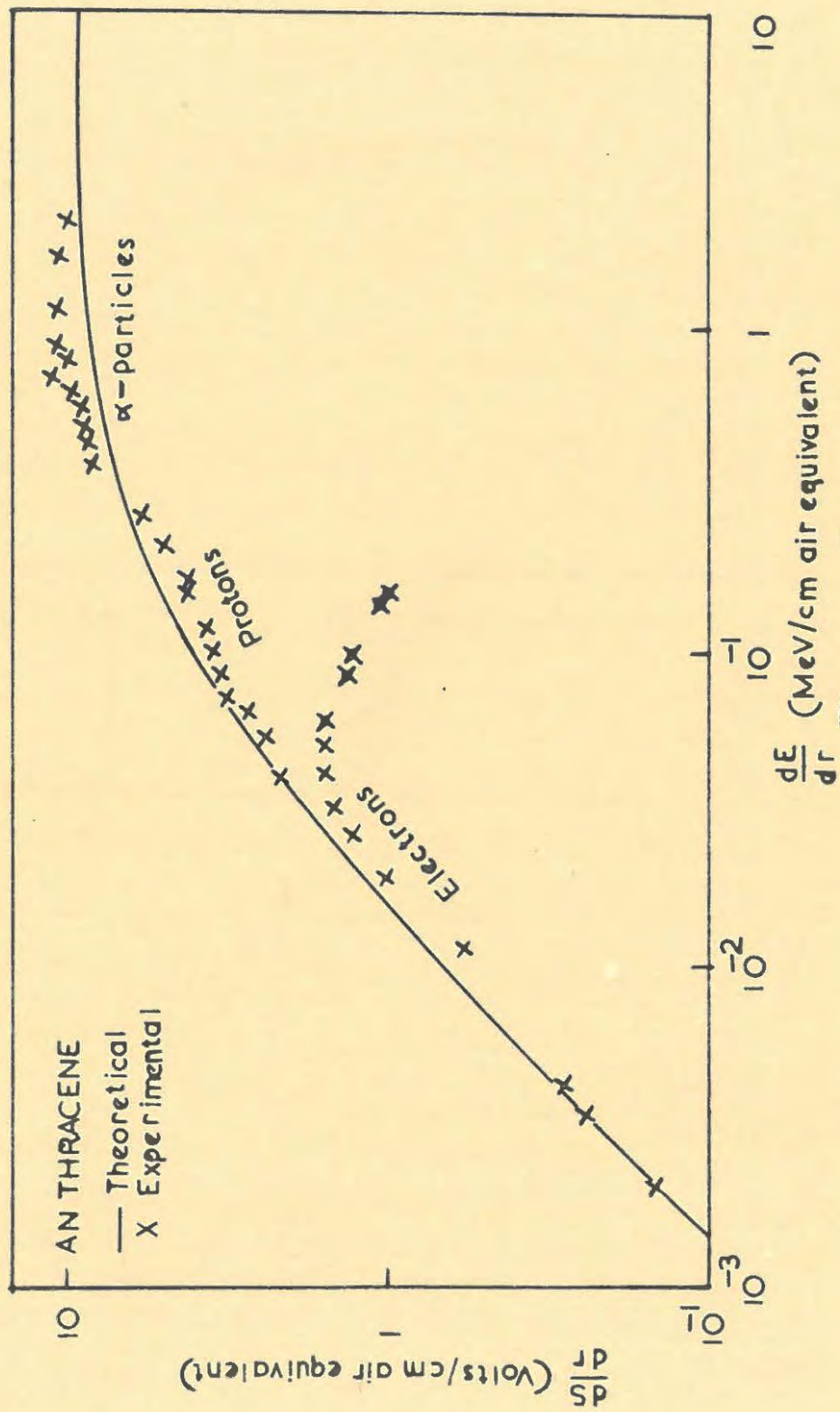


Figure 7

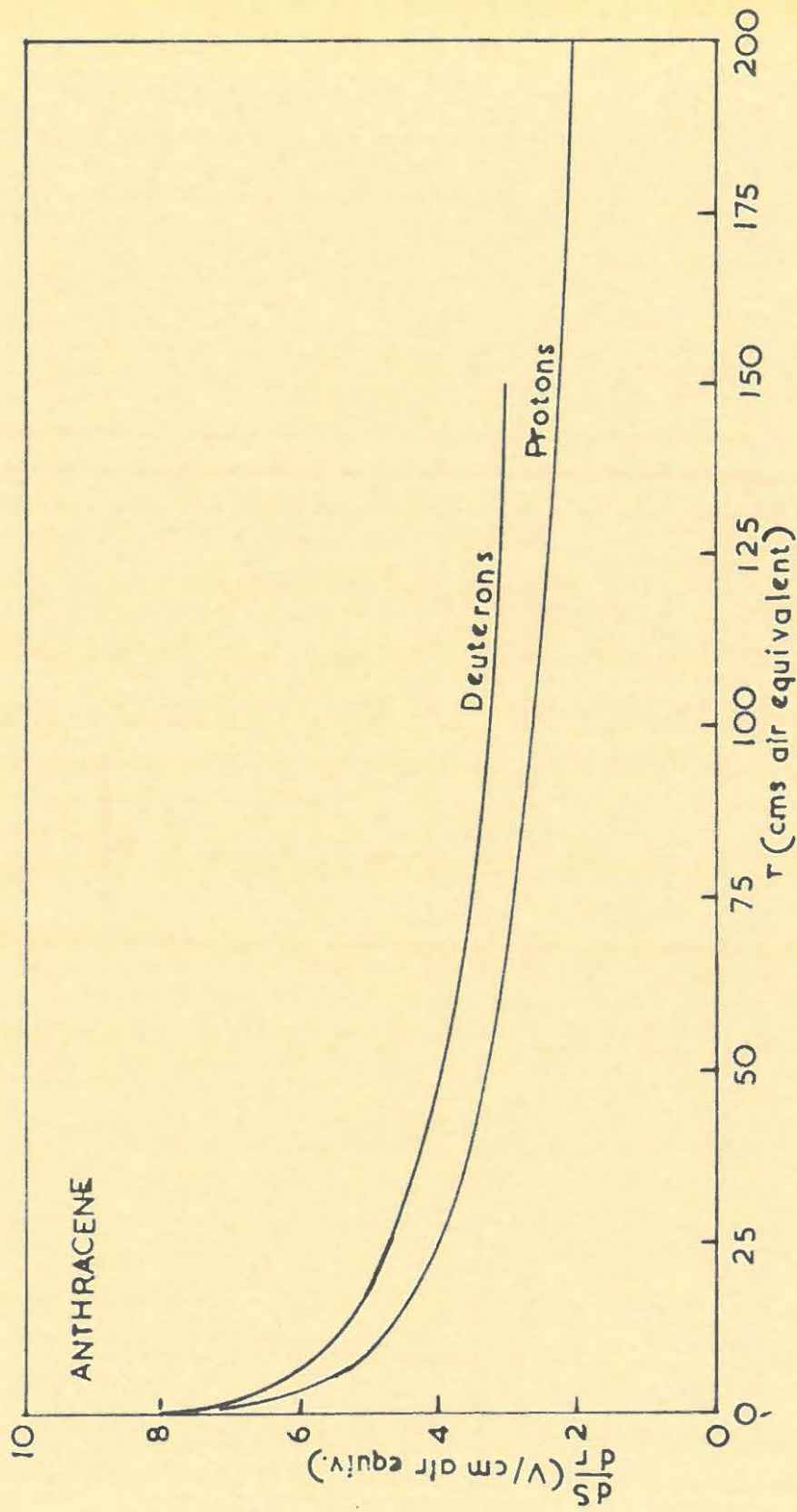


Figure 8

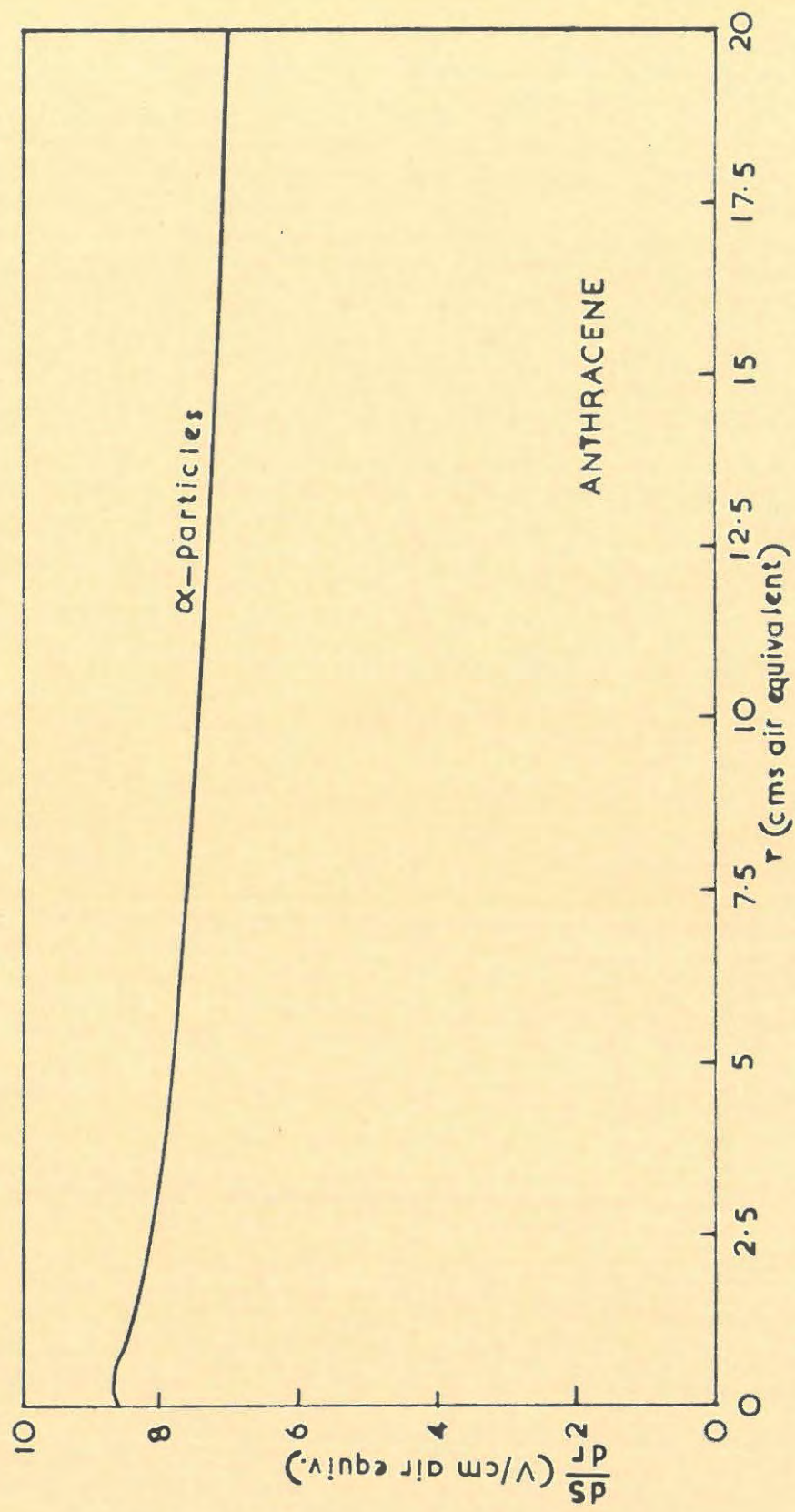


Figure 9

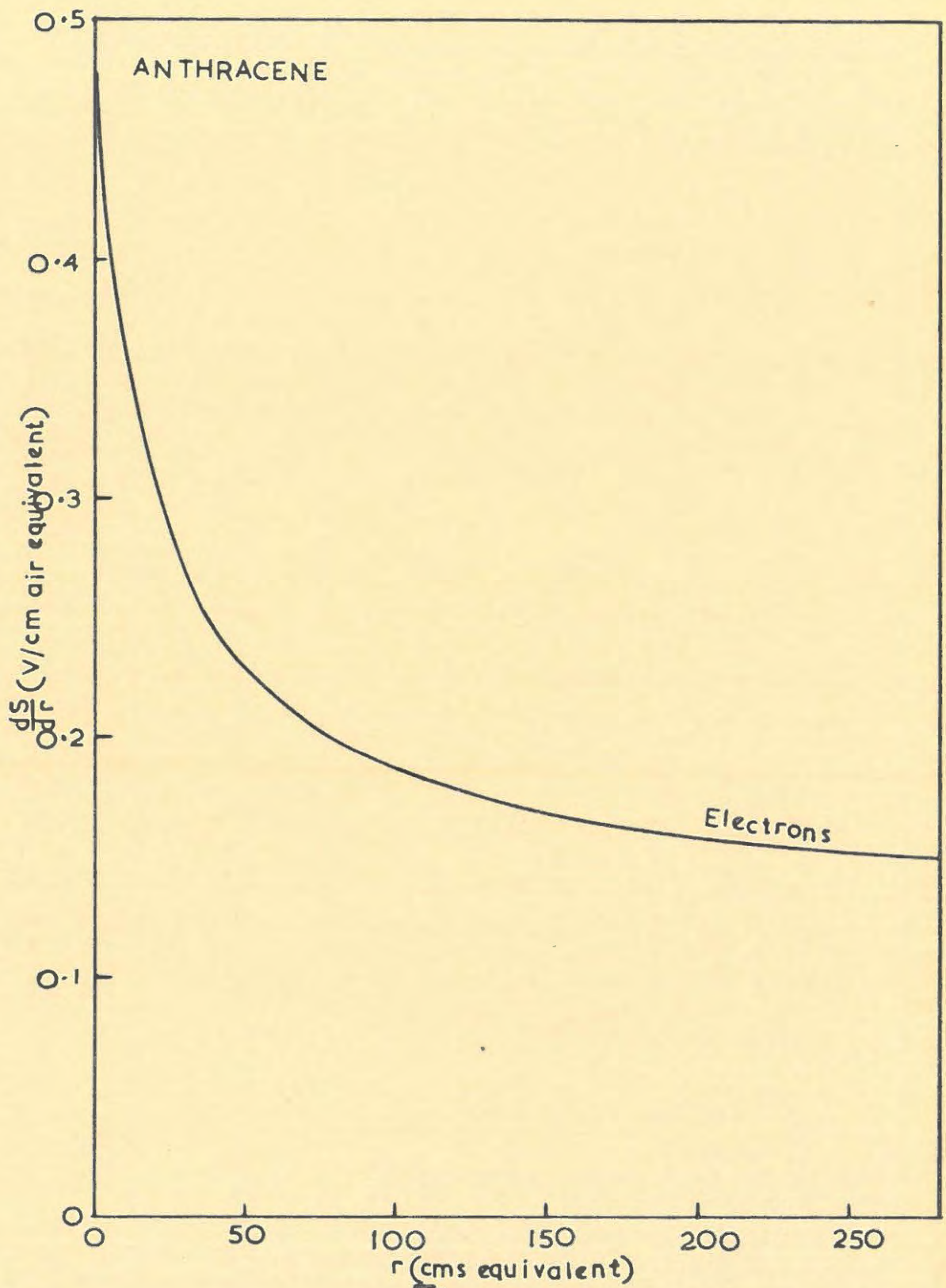


Figure 10

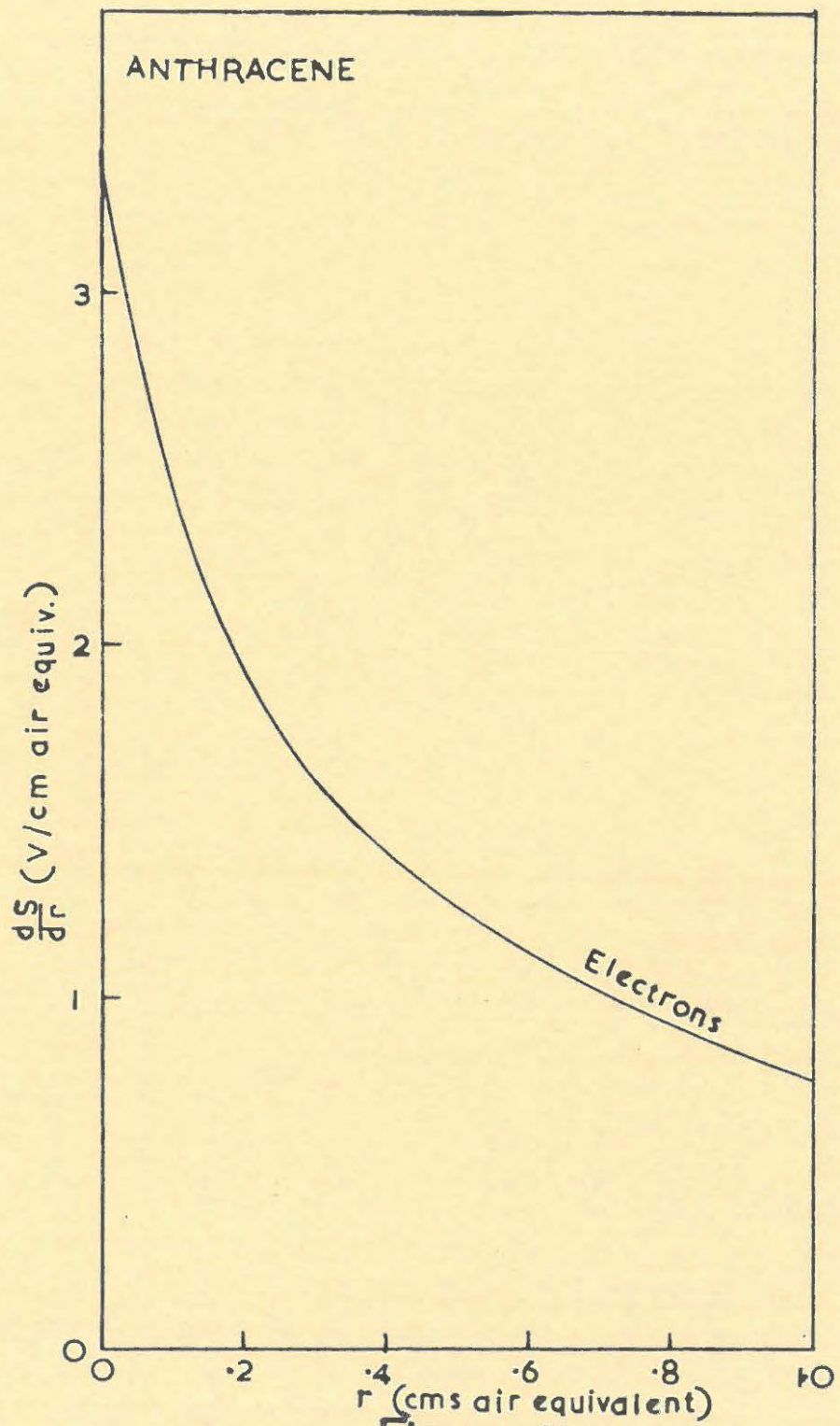


Figure 11

scintillation intensity S was obtained corresponding to particles of known range, and hence of known energy. The anthracene response curves are given in figures 12 and 13.

Comparison with experimental results.

The curves in figure 12 have been compared with normalised experimental response curves. The results used for comparison were those of Taylor *et al* (1951) for α - particles and deuterons, Hopkins (1951) for electrons and Frey *et al* (1951) for protons. The theoretical response curve agrees very well with the experimental curve in each case.

It should be noted that Raffle and Robbins (1952) have found that for E.M.I. photomultipliers the shape of the response curve becomes distorted if the charge density within the multiplier becomes too great, when the photomultiplier ceases to be a linear amplifier of the incident light pulses. This effect occurs when the output pulse becomes of the order of 0.1 volts.

In order to test further the accuracy of the calculated curves, and particularly the relative response ratios predicted for different particles, a comparison was made in table 7 of predicted pulse height ratios with the corresponding experimental ratios. The agreement is satisfactory.

The behaviour of other organic phosphors is similar

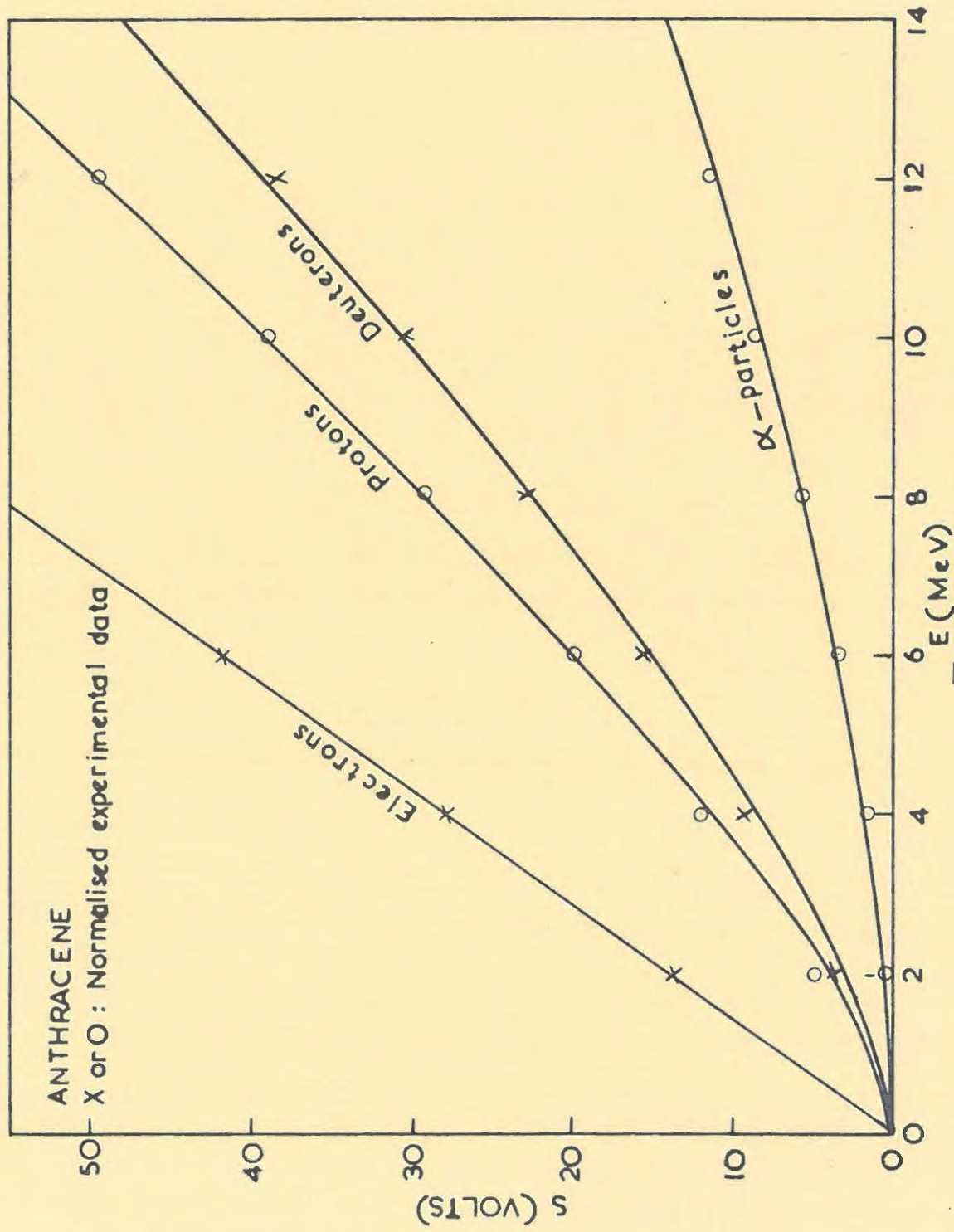


Figure 12

Table 7.

Comparison of theoretical and experimental pulse-height ratios.

Investigator	Radiations	Experimental pulse-height ratio.	Theoretical pulse-height ratio.
Birks (1950 b)	<u>186 MeV electrons</u> 162 MeV electrons	1.19	1.19
Collinge and Robbins (1950)	<u>8.78 MeV α- particles</u> 6.04 MeV α - particles	1.81	1.80
Jordan and Bell (1949)	<u>624 MeV electrons</u> 484 MeV electrons *	approx. 1.4	1.32
Jordan and Bell (1949)	<u>2.52 MeV electrons</u> *** 1.17 MeV electrons ***	2.30	2.60
Franzen et al (1950)	<u>5.3 MeV α- particles</u> 624 MeV electrons	.82	.76
Franzen et al (1950)	<u>16.4 MeV protons</u> 624 keV electrons	16.4	17.7
Hopkins (1951)	<u>5.3 MeV electrons</u> 5.3 MeV α - particles	11.6	12.1

* Produced by 668 MeV γ -rays which produce the 624 keV internal conversion electron.

~~**~~ Produced by 2.76 MeV γ -rays.

~~***~~ Produced by 1.39 MeV γ -rays.

to that of anthracene (Taylor et al (1951)) and the procedure followed above for calculating the response curves of anthracene could be followed for other phosphors.

Conclusions.

Calculations based on the exciton theory proposed by Birks (1951) are in good agreement with all available experimental data. The quenching effect responsible for the non-linearity of response in organic phosphors is an effect which depends on the specific energy loss.

PART III.THE RESPONSE OF ORGANIC CRYSTALS TO
LOW-ENERGY PARTICLES.The response to low-energy α -particles.

The scintillation responses of single flat crystals 1 cm. thick of anthracene, stilbene and terphenyl to low-energy α -particles have been studied using a collimated polonium source and a 14-stage E.M.I. 6262 photomultiplier. The circuit used is given in figure 14. The output of the tube was fed through an amplifier (figure 15) to a discriminator and scaling unit. The amplifier circuit was based on that of Jordan and Bell (1947) and was found to be linear up to 50 V for frequencies of \leq about 1.5 Mc/sec. The reasons for the inclusion of an amplifier in the circuit were twofold. It enabled the voltage applied to the photomultiplier to be reduced, thus increasing the signal/noise ratio which is important for measurements using low-energy particles and, secondly, a phase-inverter was required.

The mean pulse size S , derived from the integral pulse distribution (figure 16) was measured as a function of mean residual air range, r (at 15°C and standard pressure), of the α -particles incident on the crystal. Range corrections which amounted to \leq 0.15 mm. were applied to all

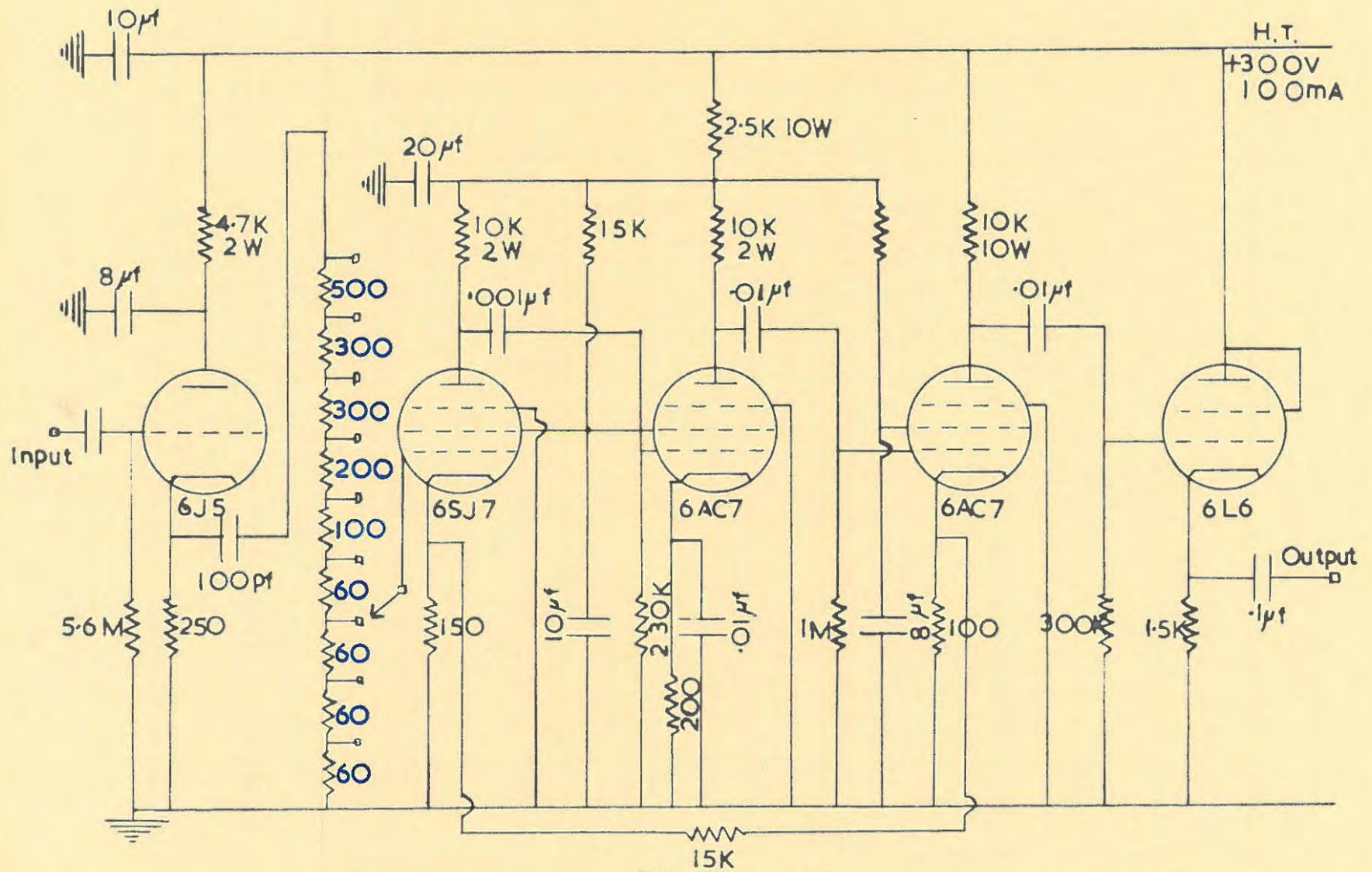


Figure 15

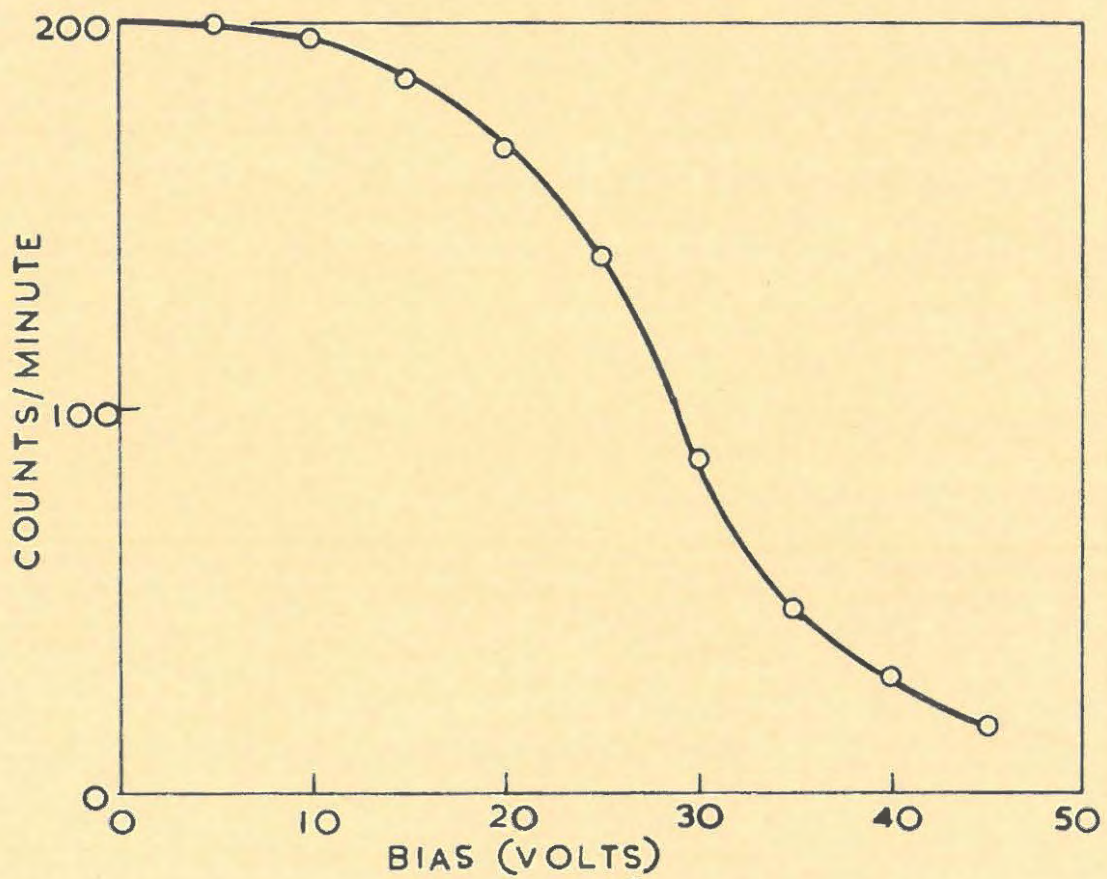


Figure 16

measurements for the slight divergence of the α -particle beam. The range correction for source thickness amounted to 2×10^{-4} mm. air equivalent and was neglected. The final corrected range is believed correct to 0.05 mm.

The experimental response curves S vs r for anthracene and stilbene are shown in figures 17 and 18. Similar results were obtained for terphenyl. It is found that for $r >$ about 10 mm., S increases linearly with r , but for $r <$ about 10 mm. the response S is a non-linear function of r . No pulse height determinations were made for particles of residual ranges. $<$ 3 mm. air equivalent owing to the high tube voltage necessary and consequent high noise count. It was observed, however, that the normal plateau of 200 counts/minute on an integral pulse height curve could still be obtained at 3 mm., indicating that the non-linearity of the response curve is not due to straggling effects.

These results are at variance with those predicted theoretically by equation (2) (page 28) which indicates in particular that dS/dr should be constant and independent of dE/dr for α -particles from Po^{210} . The experimental results show that for $r <$ about 10 mm. the value of ds/dr is less than that predicted. The ratio, ϕ , of the experimental values of dS/dr from figures 17 and 18 to the theoretical values from equation (2), i.e. to the values of

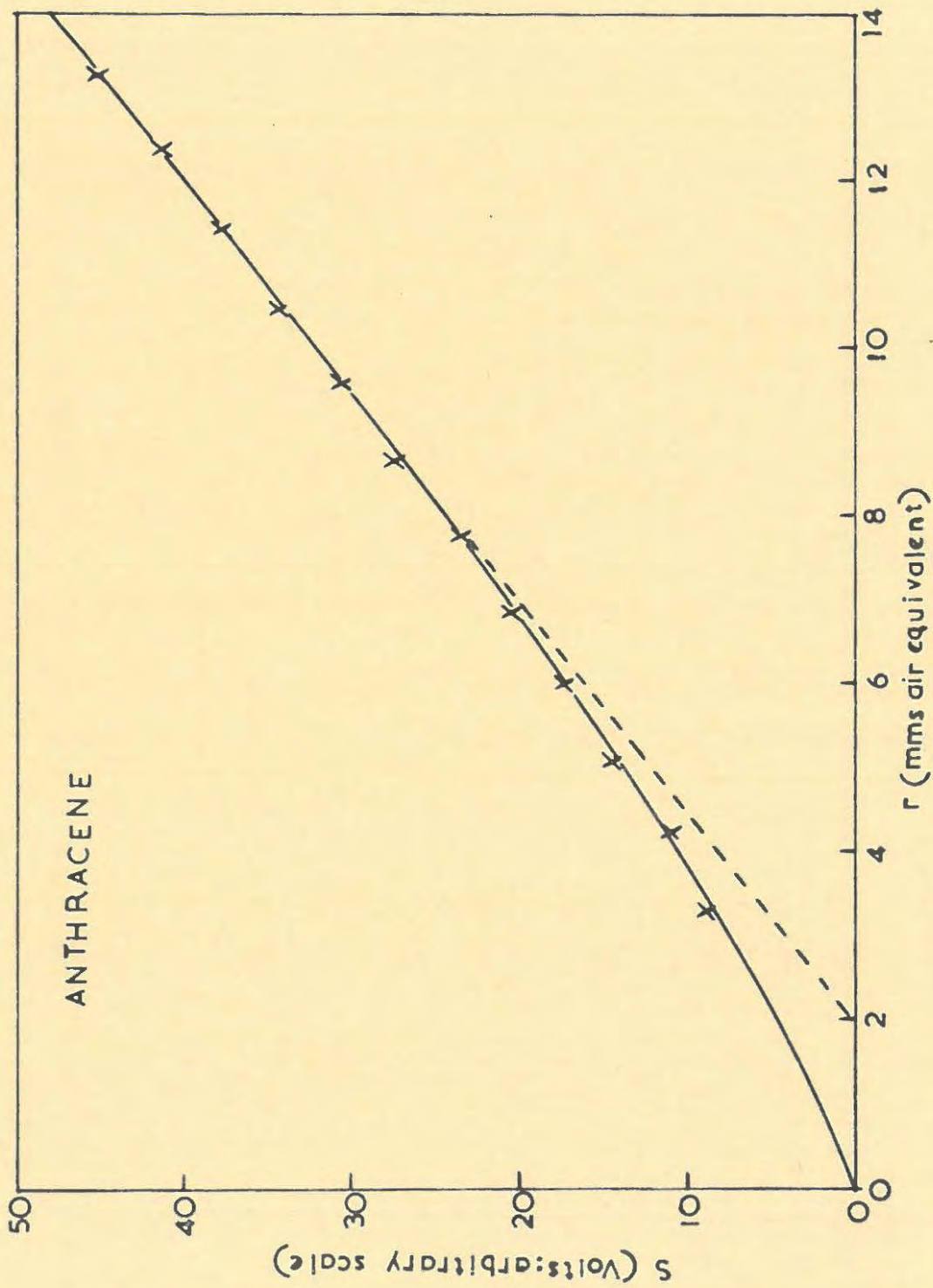


Figure 17

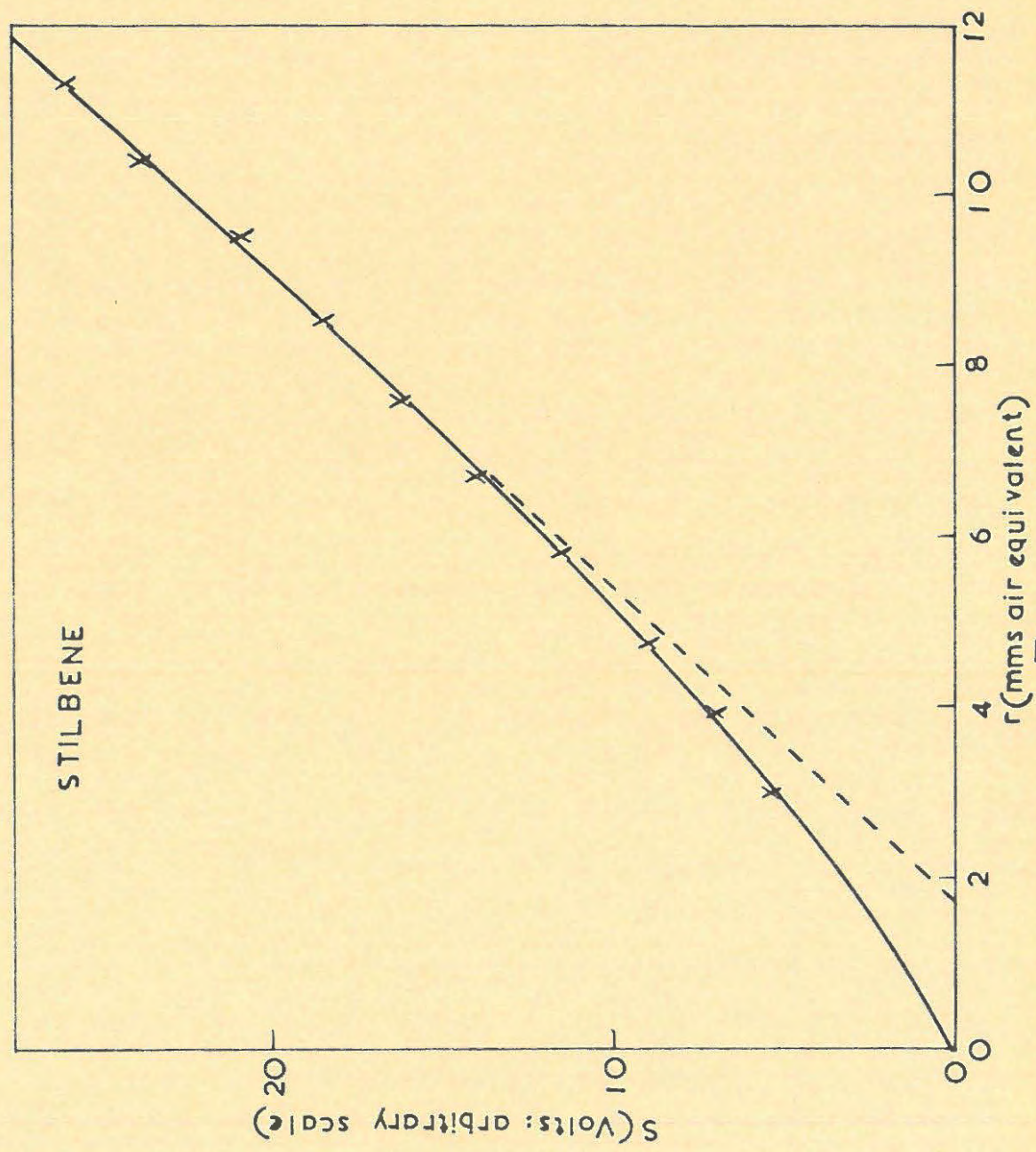


Figure 18

dS/dr from the linear portions of figures 17 and 18, are plotted for anthracene and stilbene against r in figure 19. As r decreases from about 10 mm. to zero, ϕ decreases from unity to 0.5. The value of r at which ϕ starts to decrease is also about 10 mm. in terphenyl. A brief account of this effect has been published (King and Birks (1952)).

Evidence for this effect is also given by Taylor et al who have found that the linear portions of the response curves obtained by bombarding anthracene and stilbene with α -particles do not extrapolate through the origin, but pass through approximately the 2 mm. mark in agreement with figures 17 and 18. A similar effect has been reported by Reynolds (1952) for liquid phosphors. His results are reproduced (figure 20) for a solution containing 5 g/l terphenyl in diphenyl oxide.

Theoretical extension of the exciton theory.

The exciton theory used in obtaining equation (2) has been extended (Birks (1952)) to account for this effect. It is assumed that equation (2) is valid for exciton production and capture within the bulk of an organic crystal, and that an additional factor ϕ , which depends on r , must be introduced into (2) to account for the possible "escape" of excitons produced near the surface.

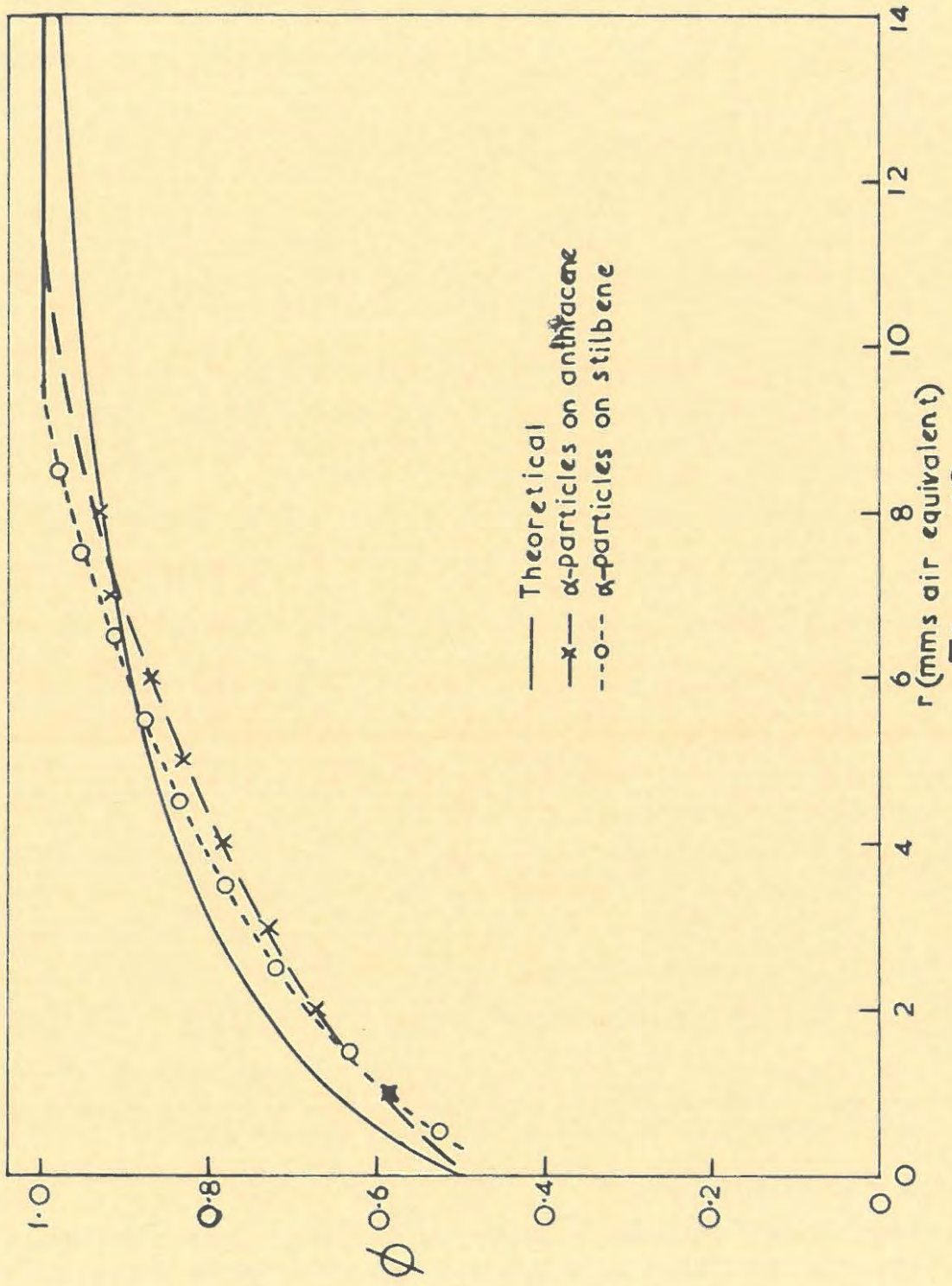


Figure 19

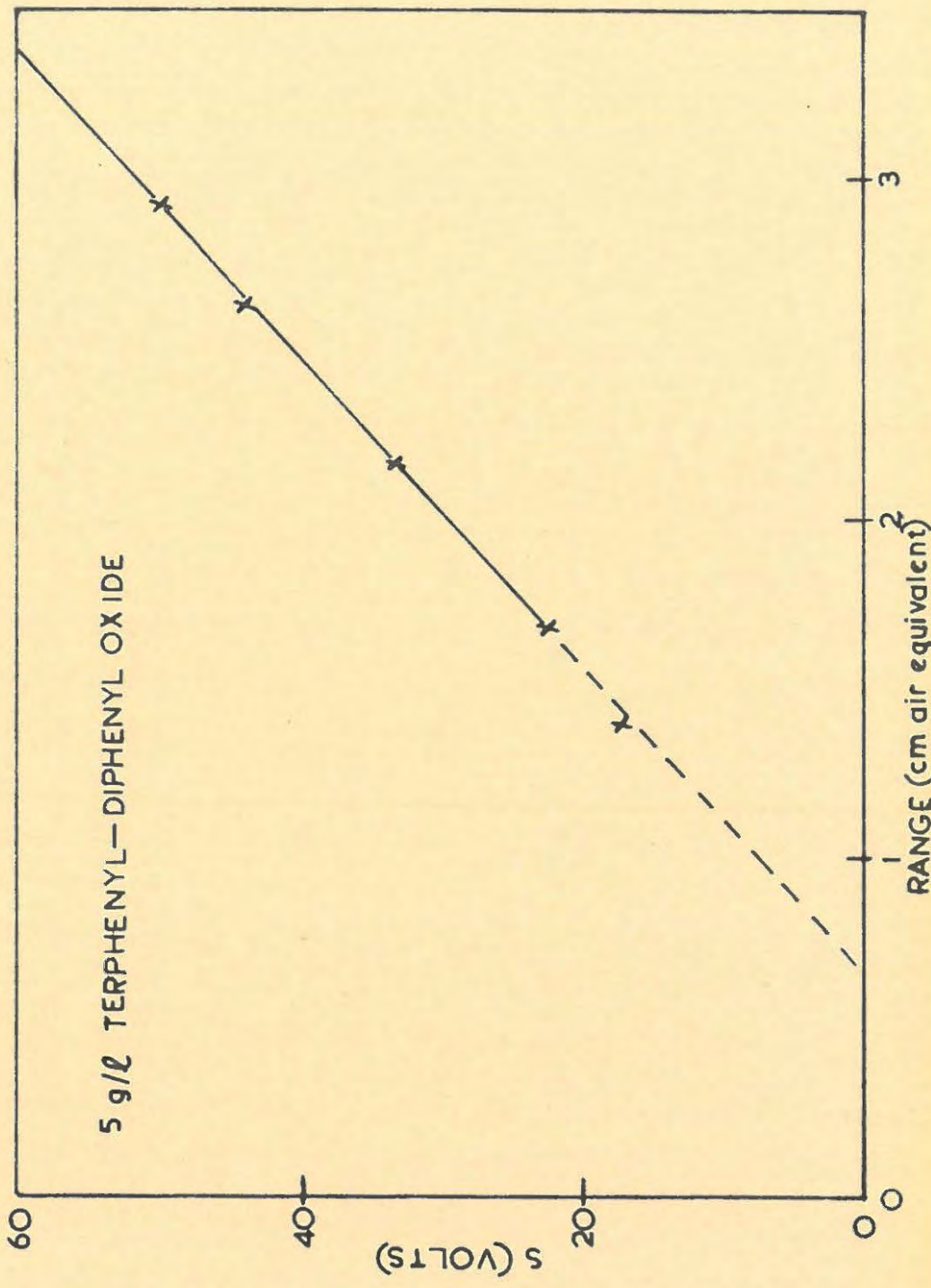


Figure 20

If a quantity a_0 be defined as the exciton mean free path, then the probability of the exciton travelling a distance a from the point of origin is proportional to $\exp(-a/a_0) / a^2$. If the point of origin of the exciton is at a distance r below the crystal surface (figure 21) then the probability of exciton capture within the crystal will be given by

$$\phi = \frac{I_r + I_\infty}{I_\infty}$$

$$\begin{aligned} \text{Where } I_r &= \int_0^r dy \int_0^\infty \frac{2\pi x dx e^{-\lambda a}}{a^2} \quad \text{where } \lambda = \frac{1}{a_0} \\ &= 2\pi \int_0^r dy \int_y^\infty \frac{e^{-\lambda a}}{a} da \\ &= 2\pi \left[\int_0^r \frac{e^{-\lambda a}}{a} da \int_0^a dy + \int_0^\infty \frac{e^{-\lambda a}}{a} da \int_0^r dy \right] \\ &= 2\pi \left[\int_0^r e^{-\lambda a} da + r \int_r^\infty \frac{e^{-\lambda a}}{a} da \right] \\ &= 2\pi \left[\frac{1}{\lambda} (1 - e^{-\lambda r}) + r \int_{\lambda r}^\infty \frac{e^{-t}}{t} dt \right] \\ &= \frac{2\pi}{\lambda} \left[1 - e^{-\lambda r} + \lambda r \int_{\lambda r}^\infty \frac{e^{-t}}{t} dt \right] \\ &= I_\infty [1 - e^{-\lambda r} + \lambda r \text{Ei}(\lambda r)] \end{aligned}$$

Where $\text{Ei}(\lambda r)$ is the exponential integral.

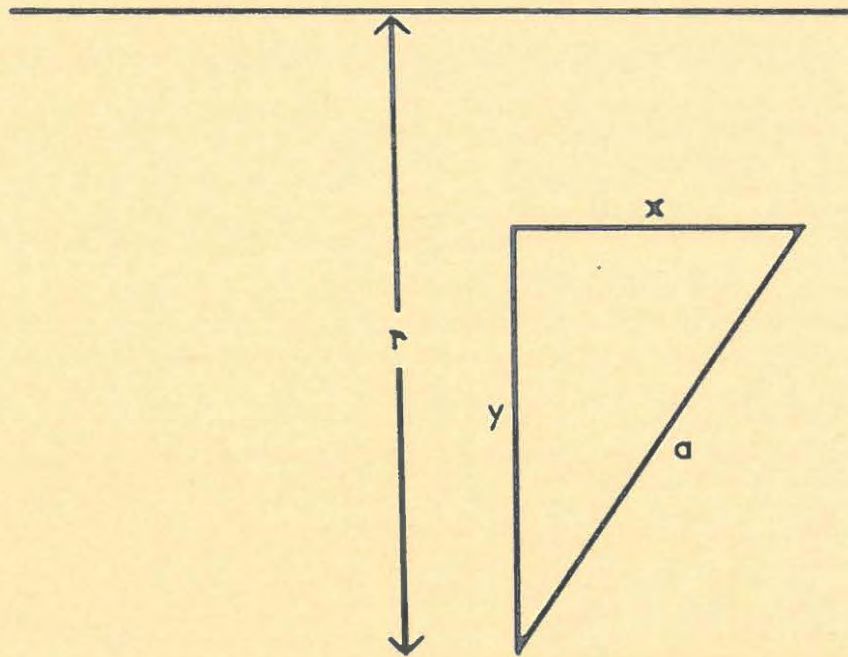


Figure 21

$$\begin{aligned}\phi &= \frac{1 + [1 - e^{-\lambda r} + \lambda r \operatorname{Ei}(\lambda r)]}{1 - \frac{1}{2} [e^{-\frac{r^2}{a_0^2}} - \frac{r}{a_0} \operatorname{Ei}(\frac{r}{a_0})]} \\ & \text{----- (3)}\end{aligned}$$

and the complete expression for dS/dr is given by

$$\frac{dS}{dr} = \frac{\phi A dE/dr}{1 + B^1 dE/dr.}$$

ϕ decreases from unity to 0.5 as r decreases from ∞ to zero. The curve of ϕ vs r from equation (3) taking $a_0 = 8$ mm. air equivalent is shown in figure 19, and gives qualitative agreement with the experimental curves.

The probable existence of preferred directions of exciton propagation has been neglected in the derivation of equation (3). Before further improvements in the theory are attempted it is considered that further experimental results are required, using crystals with top surfaces which have been carefully polished and are free from oxygen, as the possibility has not been excluded that reduced efficiency near the crystal surface could be an oxidation effect.

It may be remarked that it is difficult to imagine the escape of an exciton through a crystal surface. If, however, the primary process of energy migration was not exciton migration, but photon emission and re-absorption,

the escape of photons through the crystal surface would easily occur. It would then be expected that an aluminium foil placed on top of the crystal would produce a difference in the low-energy particle response curve, as photons which would normally escape would be internally reflected and reabsorbed. However, no change in the response curve could be detected when an aluminium foil of thickness 2.6 mm. air equivalent was used to cover the crystal. This fact yields no information on the mode of primary energy migration as the photons, if they existed, could possess wavelengths at which aluminium is a poor reflector. A critical experiment in deciding whether the primary mode of energy migration occurs by photon emission and re-absorption would be that of obtaining the response curve of a phosphor for low-energy particles striking the bottom of a deep hole of small cross-section in the crystal. Photons which escape into the cylindrical hole would travel through it and be re-absorbed, and the response curve would be linear down to zero range.

The response to low-energy electrons.

The results of Taylor et al (1951) for anthracene show that in the case of electrons of ranges \leq about 9 mm. air equivalent the specific fluorescence is, as for α -particles, less than theoretically predicted. Using the

theoretical curve of $d\bar{S}/dr$ vs r (figure 11) and experimental values calculated by multiplying $d\bar{S}/dE$, from the experimental response curve, by corresponding values of dE/dr , figure 22 has been obtained. This shows ϕ , the ratio $(d\bar{S}/dr)_{\text{expt.}} / (d\bar{S}/dr)_{\text{theor.}}$, as a function of r . This curve is found to be in qualitative agreement with the theoretical curve of ϕ vs r from equation (3), using $a_0 = 3$ mm. air equivalent.

It may be predicted in terms of the explanation given above that the response curve for low-energy electrons produced within the bulk of the crystal by X-rays would not show the effect noticed by Taylor *et al.* An attempt was made to verify this using 5.9 keV K X-rays from Fe⁵⁵ but it was found that only single-photon pulses were produced and no measure of the pulse height could be obtained with the electronic counting equipment available.

Conclusions.

The fact that the specific fluorescence produced by short-range particles in organic crystals falls below theoretical values seems to be a general effect, and may be explained by the non-capture of excitons produced near the surface of the crystal. An explanation in terms of particle momenta cannot be offered since the ratio of the momentum of an α -particle of range 10 mms. air equivalent to the momentum of a similar electron is about 10^4 . δ -rays

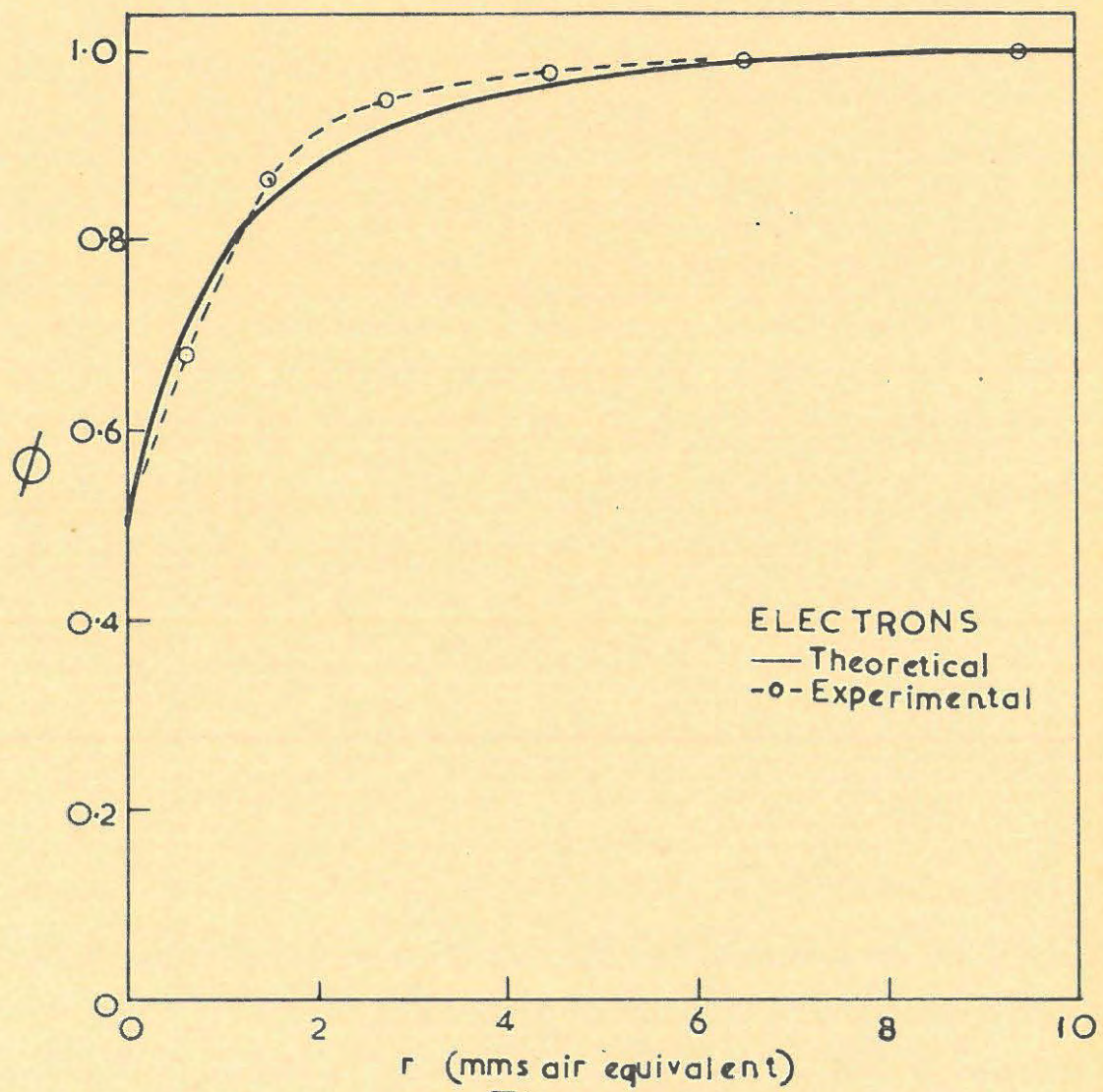


Figure 22

are not responsible for the effect in any way, since an α -particle of range 10 mms. produces δ -rays of energies < 0.8 keV, most of them having energies very much less than this value.

A value of about 3 - 8 mm. air equivalent is obtained for the exciton mean free path in anthracene, stilbene and terphenyl. The stopping power of each of these materials is about 1200 relative to air = 1 (Livingston and Bethe (1937)) which gives an exciton mean free path of about 3 - 7 microns, corresponding to about 10^4 molecular lengths.

In view of this mean free path value it is believed that the fluorescence observed in organic crystals by Ageno and Cortellessa (1952) at distances of several hundred times the α -particle range from the source must be due to a secondary process which is the absorption and re-emission of fluorescent light which has previously been emitted after exciton migration and capture.

To extend the data on the response of organic crystals to short-range particles the response to protons of energies < 1 MeV should be obtained.

PART IV.THE PRODUCTION OF LUMINESCENCE IN AIR,
GLASS AND QUARTZ BY α - PARTICLES.

During experiments using strong polonium α - particle sources and organic phosphors in a scintillation counter, an effect was noticed which could not be explained in terms of any normal response of the phosphor to the α - particle irradiation. It was believed that this effect might be partly due to the self-luminosity of the source.

The self-luminosity of polonium.

It is well known that Po^{210} and other α - particle sources are feebly self-luminous. The spectrum of this luminosity was first examined with a quartz spectrograph by Sir William and Lady Huggins (1903, 1905 and 1906) and found to coincide with that of nitrogen. They concluded that the effect was due to α - particle excitation of nitrogen occluded in the source, and in the air irradiated by the source. This has been confirmed by later observers, the most recent being Ortner and Salim (1952), who photographed the visible spectrum of the light emitted from a Po^{210} source, and from the air near the source. They identified the various lines and bands in the two spectra with those from nitrogen, and also oxygen. This luminosity, and other

possible effects, has been investigated as described below.

It is interesting to note that Albrecht and Mandeville (1951) have counted α - particles by the detection in a scintillation Geiger counter of radiation emitted from the glass walls themselves. They have reproduced the weak ultra-violet emission spectrum (excited by polonium α - particles) of Corning 9741 glass, which is used in the construction of scintillation Geiger tubes. The author has noticed that superimposed on this spectrum are four bands, each one of which has a wavelength which agrees, within the accuracy of measurements from the spectrogram, with one of the four main wavelengths of the nitrogen band spectrum.

Experimental method and results.

An E.M.I. type 5060 photomultiplier tube, which has a 1 cm. diameter semi-transparent photo-cathode deposited on the inside of a 1.5 mm. thick flat glass window, was used. A 2.4 mc Po^{210} source, deposited on the end of a 0.9 mm. platinum wire, was located at the end of a brass collimating tube, 15 mm. long and 1.5 mm. internal diameter, giving a narrow emergent beam of about 4×10^6 α - particles per minute. The source was mounted in air directly above the photo-cathode, on a micrometer screw, so that the

distance from the tube could be varied. The output from the photomultiplier was fed through a cathode follower, linear amplifier and discriminator to a fast scaling unit as described in part III. Throughout the experiments all surfaces were kept clean and free from materials not specifically mentioned.

With the source in position, a large number of small amplitude pulses, greatly exceeding the noise count, were observed. A typical pulse-height distribution curve is plotted in figure 23, and shows no indication of a plateau. Comparative measurements were therefore made of the counting rate at constant gain and a fixed discriminator bias. The number of counts observed at this bias was approximately eighteen per cent of the total number of counts, extrapolated to zero bias.

Figure 24 shows the observations on the variation of counting rate with the distance of the source from the photomultiplier. This distance is expressed in mm. of standard air equivalent, the range of the Po^{210} α - particles being 38.2 mm. The noise count of 370 c.p.m. has been subtracted throughout. The gross count curve (ABC) is attributed to counts arising from photons produced (A) at the source, (B) in the air between the source and the photomultiplier and (C) in the glass of the photomultiplier.

The counts (A) were eliminated by covering the source

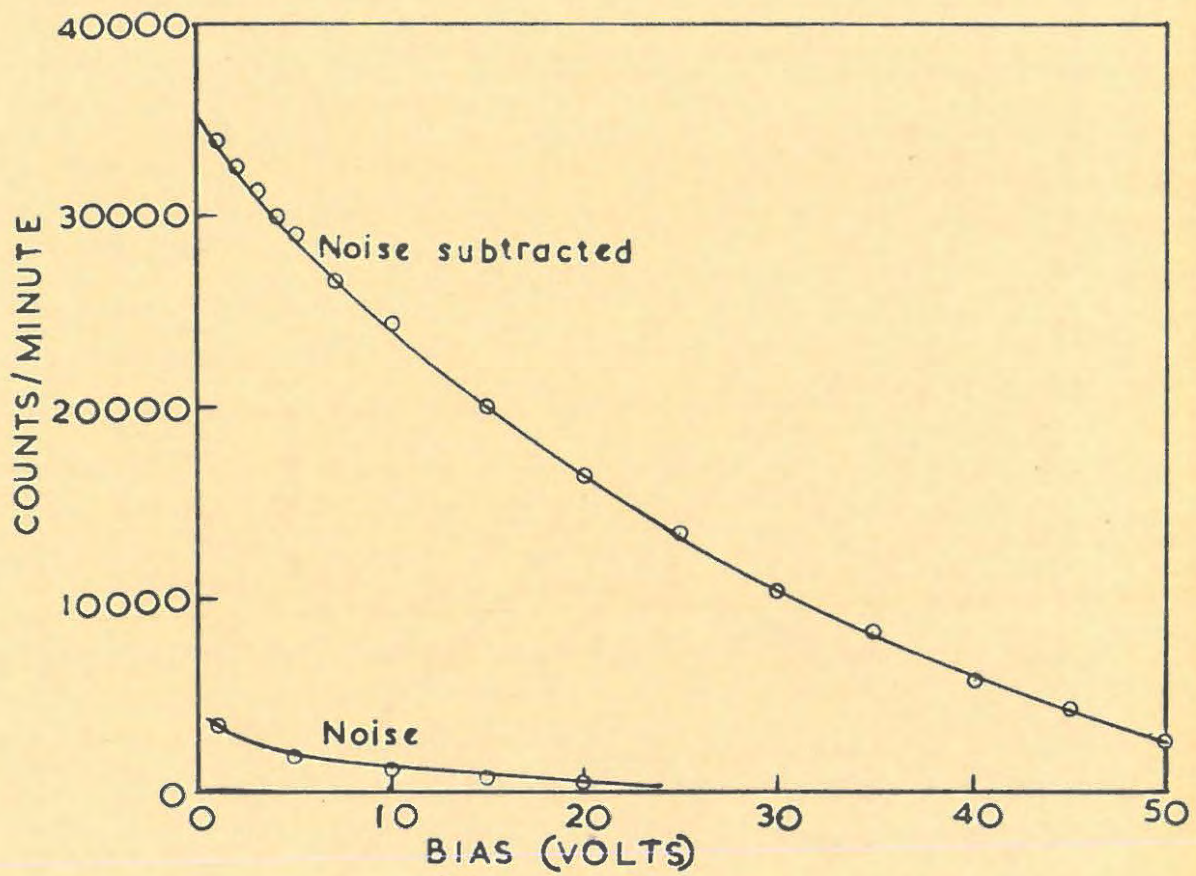


Figure 23

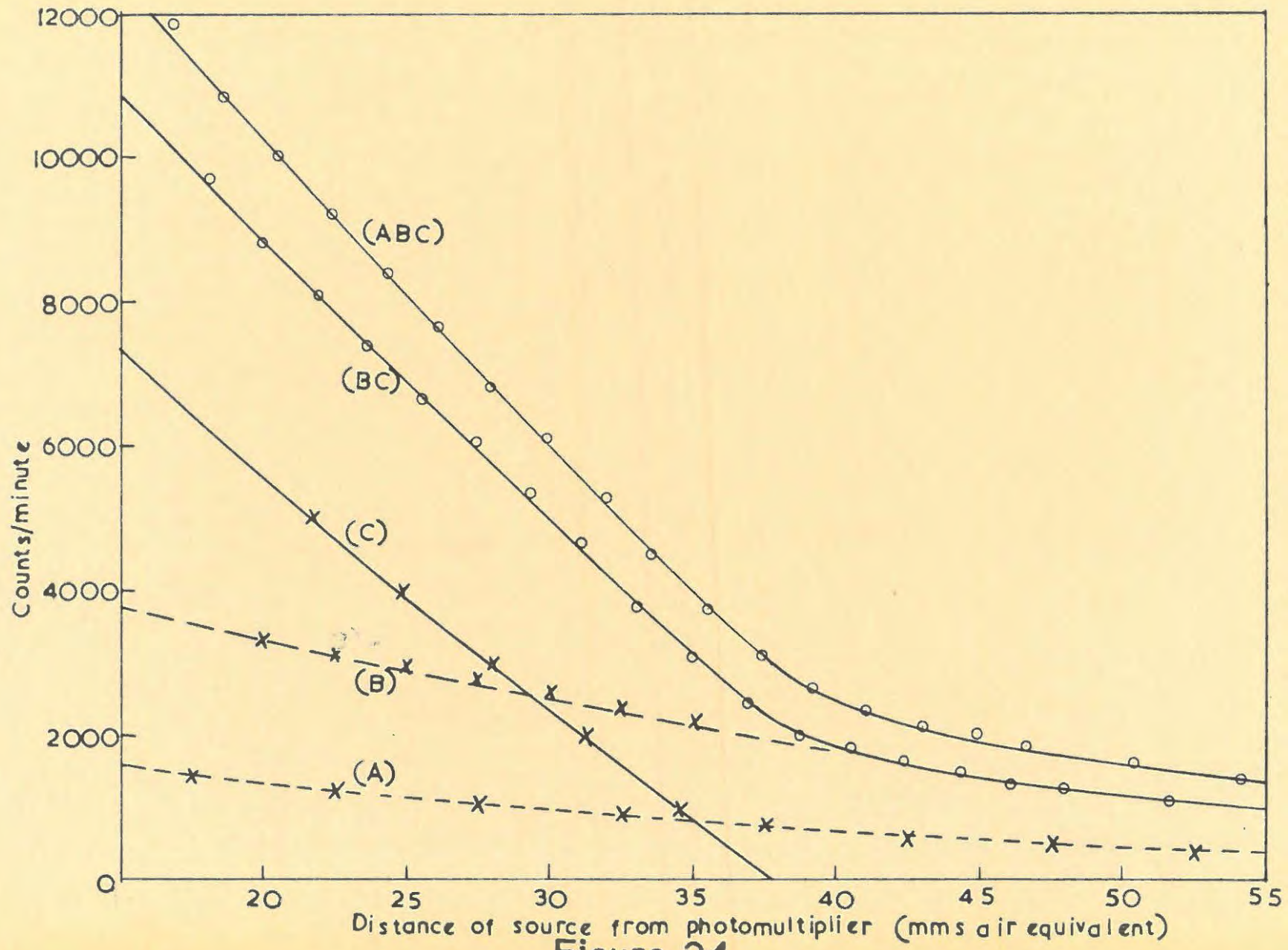


Figure 24

with an aluminium foil of 1.3 mm. equivalent air thickness, placed 0.1 mm. in front of the source. Curve (BC) was then obtained, the distance being corrected for the equivalent air thickness of the foil. This curve shows two separate effects, one of which occurs only when the source is less than about 38 mm. from the tube. Curve (A) gives the difference between curves (ABC) and (BC), and represents the counts due to photons produced at the source.

The counts (B), due to photons produced in the air, were next eliminated (figure 25) by placing two aluminium foils, each of 1.3 mm. air equivalent thickness, directly on the photomultiplier, and obtaining curve II. When the distance of the source from the photomultiplier was corrected by the addition of the equivalent air thickness of the foils, curve I was obtained. It is seen that the photons cease to be detected when the source distance is greater than 38.2 mm. air equivalent, i.e. greater than the α -particle range, and the number of photons detected increases with the residual range of the α -particles in the glass. Curve III was obtained when four more similar foils were placed on the photomultiplier. Curve III is parallel to curve II and coincides with it if the distance is corrected by the addition of the equivalent air thickness of the four extra foils. A series of parallel curves was obtained experimentally, the displacement of each curve from curve II depending on the

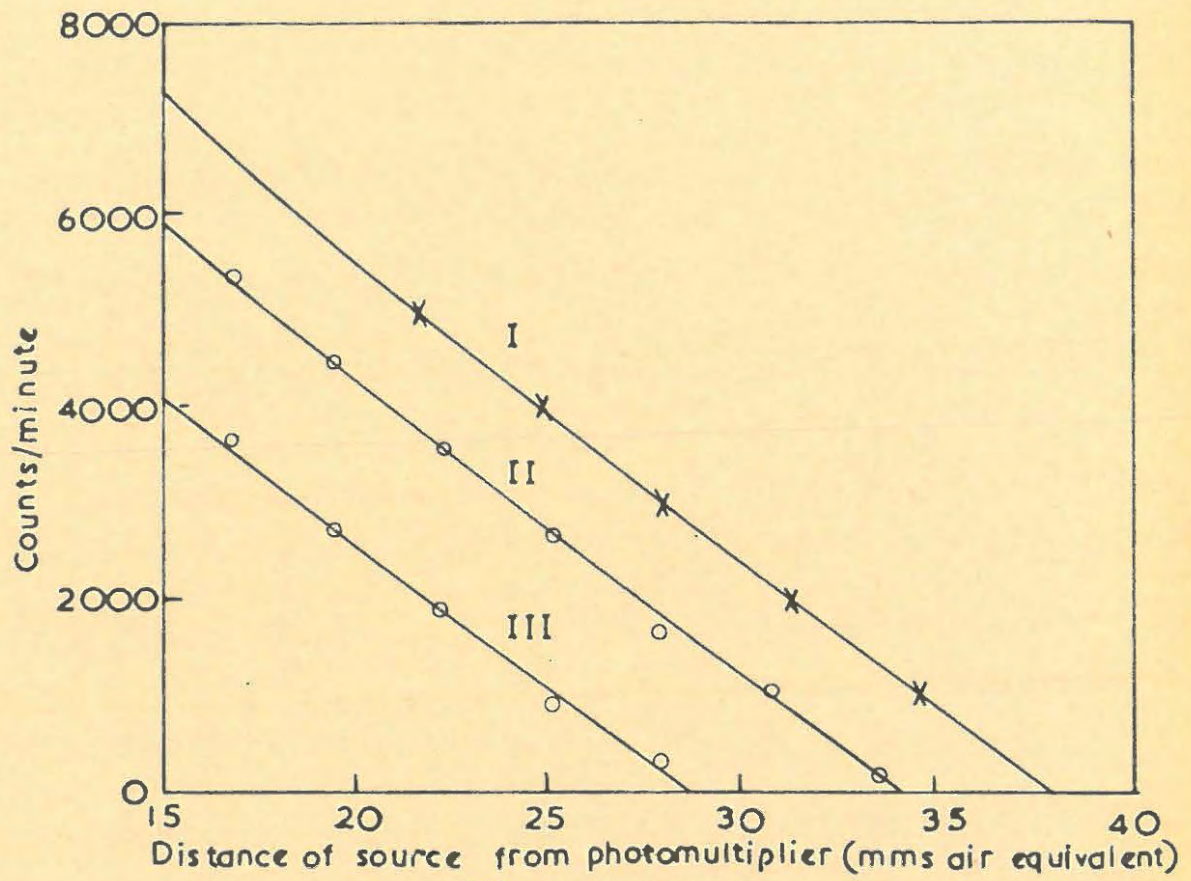


Figure 25

number of foils on the photomultiplier.

It is concluded that the photons are produced in the glass, and not in the aluminium foils, which have no effect on the number of counts observed. Curve (C), plotted in figure 24, shows the number of counts due to photons produced in the glass. Curve (B), obtained by subtracting (B) from (BC), represents the counts due to photons produced in the air.

These experiments were repeated with a fused quartz slab 1.45 mm. thick placed on the photomultiplier. Photons produced in the quartz were detected, and separated from those produced at the source and in air, by introducing aluminium foils above the quartz. When an aluminium foil was placed between the quartz and the photomultiplier, the effect was completely eliminated. The results were similar to those obtained with the glass (figures 24, 25), but the number of counts due to photons originating in the quartz was a factor of two less (figure 26), and those due to photons produced in the air and at the source a factor of three less. This is attributed to reduced solid angle of detection, and possible absorption in the quartz.

The effects described above were also obtained with a second polonium source of strength 1.7 mc deposited on a $\frac{1}{4}$ " diameter platinum plate.

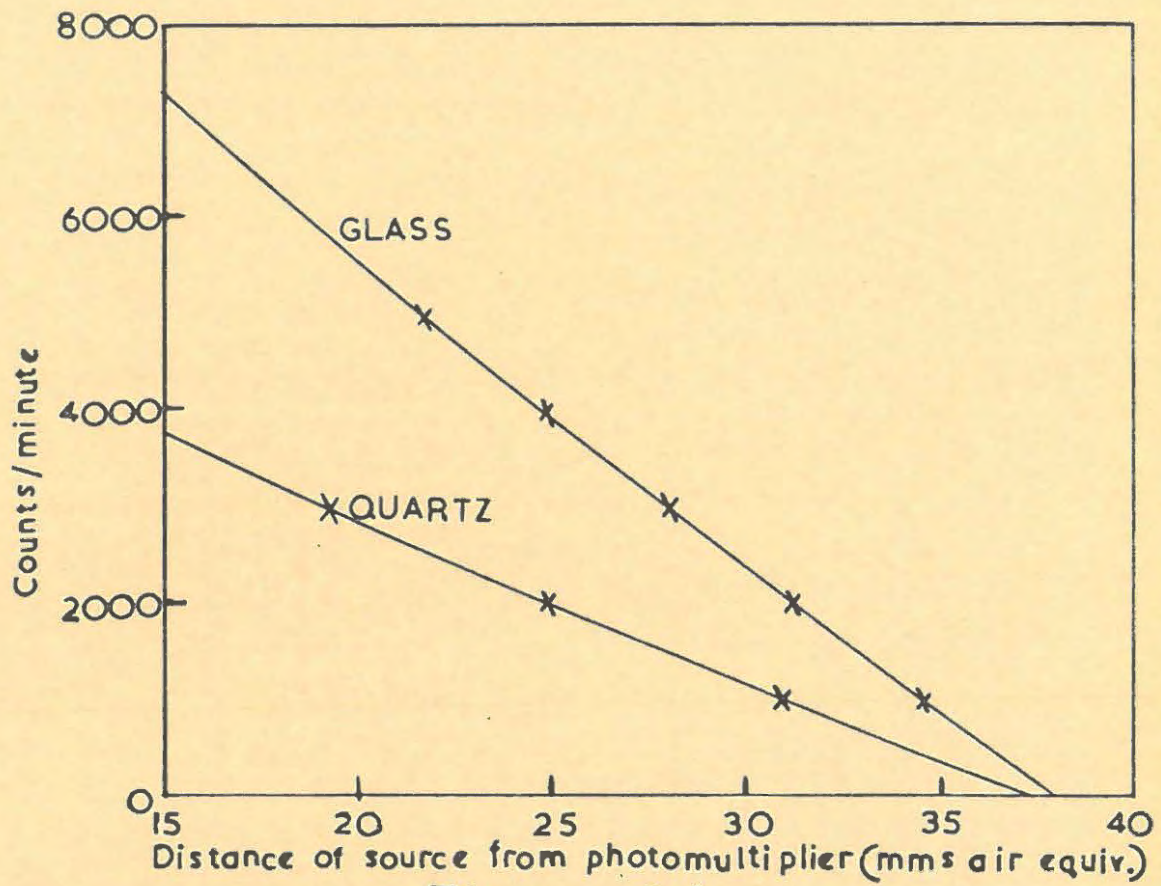


Figure 26

Discussion and conclusions.

It is concluded that the α - particles are responsible for the production of photons at the source, in air and also in glass and quartz. The effect cannot be ascribed to the weak γ - radiation from polonium (Grace et al (1951)), as the photons from the glass cease when the α - particles are unable to penetrate into it.

A rough estimate, made by extrapolating curve (C), figure 27, to zero source distance, indicates that the total number of counts due to photons that would be produced in the glass by 5.3 MeV α - particles is only 0.02 of the number of incident α - particles. Assuming a mean photo-cathode conversion efficiency of about 2%, this indicates that only about one photon per α - particle reaches the photo-cathode. The total number produced may, however, be several times higher, since the glass is opaque at wavelengths below $320m\mu$. The results with the quartz indicate that it is reasonably transparent to the photons, and it would be desirable to repeat the measurements using a photomultiplier with a quartz envelope.

Richards and Cole (1951) have described experiments in which they observed "scintillations" when thin films of many different materials were irradiated by α -particles

from a strong Po^{210} source. The "scintillations" were only observed when the thickness of the film was less than the range of the α - particles in the material. This has been interpreted as implying that the radiation is readily absorbed by the parent material. In the majority of cases, the films were deposited on a 1 mm. quartz slide, which was reversed to take noise readings.

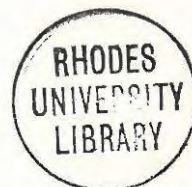
Richards and Cole referred to neither the self-luminosity of the source, nor to the photons produced by α - particles in air, quartz or the glass of the photomultiplier envelope. It appears from their reports that in each case the residual range of the α - particles, after emerging from the film, was in one or more of these three materials. It is evident that some, and probably all, of the "scintillations" ascribed to the thin films, originated in the quartz, glass or air. This would explain their statement that "the efficiencies of all these films are approximately the same", since the efficiency in question would be that of the quartz slide below the film. Finally, it is difficult to imagine how the "scintillations" emitted by glass (which was one of the materials included by Richards and Cole in their list) were able, if they are highly absorbed by glass, to be detected through the wall of the photomultiplier used.

The results and conclusions of the investigation described in part IV are to be published (Birks and King (1952)).

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