PROPORTIONAL COUNTER STUDIES OF ELECTRON CAPTURE TO POSITRON EMISSION RATIOS IN LIGHT ISOTOPES

by

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CONTENTS

PREFACE

This thesis contains an account of research conducted in the Natural Philosophy Department of the University of Glasgow between October 1963 and December 1966.

Chapter One consists of a brief review of the theory of beta decay with emphasis on orbital electron capture and on K capture to positron emission ratios. Techniques which have been used for the measurement of K/β^+ ratios are discussed and a review is given of the experimental results obtained by various workers for isotopes of low atomic number. The various predicted and observed deviations from current theory are noted and the sensitivity of the K/β^+ ratio to small energy-dependent deviations is discussed.

Proportional counter techniques for the measurement of K/β^+ ratios are discussed in some detail in Chapter Two, and different types of continuous flow counter designed to minimise the wall effect are described. These include the plastic counter developed by Drever and Moljk, and the foil counter, a variation on the above, built by the author and Dr. K. W. D. Ledingham. A new technique suggested by Dr. R. W. P. Drever for the investigation of very short-lived activities by the direct activation of the gas contained in a static proportional counter is also described. Finally, the difficulties involved in the interpretation of experimental pulse height spectra are discussed, and attention is drawn to the lack of information on the spectrum shapes produced by the soft radiation emitted following electron capture in isotopes of low atomic number.

The remaining chapters describe the author's experimental research, which falls into two distinct sections; two chapters are devoted to a study of the response of proportional counters at very low energy and two to work on K/β^+ ratios. The results obtained in the first project are applied in the interpretation of the experimental data obtained in the second.

In Chapter Three, an account is given of an investigation of the pulse height distributions produced when mono-energetic x rays of energies below 3 keV are absorbed in a proportional counter. The distributions were found to approximate to the Poisson function, the relative standard deviation having the value $0.169/E^{\frac{1}{2}}$, where E is the x-ray energy in keV. As a corollary to this work, an experimental study was made of the fluctuations in the gas multiplication process, existing results being in apparent conflict. This investigation, described in Chapter 4, determined the contribution to the width of a peak from the statistical variations in the avalanche size, and the results are shown to be in quantitative agreement with those of Chapter Three.

The experimental work described in these two chapters was carried out by Dr. K. W. D. Ledingham and the author, the analysis of the results being the responsibility of the latter. The paper published on this work was written by the author.

Chapter Five contains an account of the measurement of the $K/\beta K/\beta^+$ ratio in Carbon 11 using the plastic counter. The result

of $(2.30 \pm^{0.14}_{0.11}) \times 10^{-3}$ is in reasonable agreement with current theory and is considered to be more reliable than the earlier measurement on this isotope. The experiment was carried out by Dr. Ledingham, Mr. W. Leiper and the author, computer programmes for the analysis of results and the calculation of the theoretical ratio being written by the latter two persons. The report presented to the Glasgow Conference on Nuclear and Particle Physics (1966) was the work of the author, and a paper based on this report is in the process of publication.

The development of the counter irradiation technique and its first application, to the isotope neon 19, in which electron capture had not been observed previously, is described in Chapter Six. This work was undertaken mainly in order to evaluate the applicability of the technique to a proposed investigation of very short-lived sources produced by the Glasgow linear accelerator. Various minor experiments and a measurement of the K/β^+ ratio in neon 19 were carried out by the author and Mr. Leiper. The author was responsible for the analysis and interpretation of the experimental results.

The results of the work described are discussed briefly in Chapter Seven and an indication of possible future developments is given.

PUBLICATIONS

Pulse height distributions from proportional counters

J. L. Campbell and K. W. D. Ledingham,

Brit. J. App. Phys. Vol. 17, No.6, June 1966.

The ratio of K-capture to positron emission in the decay of carbon 11

J. L. Campbell, W. Leiper, K. W. D. Ledingham and R. W. P. Drever,

Accepted for publication (February, 1967) in Nuclear Physics.

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Latterly the author worked in collaboration with Mr. W. Leiper; this association was both stimulating and instructive.

The author made full use, during his research, of the various technical facilities available in the Department of Natural Philosophy; his thanks are due to many members of the technical staff for their help.

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CHAPTER ONE

INTRODUCTION

1. THE GENERAL THEORY OF BETA DECAY

The complete theoretical description of the radioactive disintegration of nuclei by the well known beta decay processes

$$N(Z,A) \longrightarrow N(Z+1,A) + e^- + \bar{\nu} \text{ (negatron emission)} \quad (1)$$

$$N(Z,A) \longrightarrow N(Z-1,A) + e^+ + \nu \text{ (positron emission)} \quad (2)$$

$$N(Z,A) + e^- \longrightarrow N(Z-1,A) + \nu \text{ (orbital electron capture)} \quad (3)$$

has been a problem of major interest for the past thirty years since the publication of the first, partially successful, theory by Fermi (1934). The many radical changes of opinion which have occurred during the development of the subject necessitate a careful and critical approach to all work which has a bearing on the currently accepted theory.

Since the lifetimes involved in beta decay indicate that the force responsible for the process is weak compared with the strong nuclear binding forces, the problem lends itself to treatment by perturbation theory, which is the method used to treat the analogous process of emission and absorption of photons in the electromagnetic interaction. Fermi invoked both this analogy and the neutrino hypothesis of Pauli (1933) to construct an interaction Hamiltonian which was invariant under the Lorentz transformation.

In the electromagnetic case, the transition rate is proportional

to the four-vector current $j_\mu = \psi_f \bar{\gamma}_\mu \psi_i$ where the γ_μ are the Dirac matrices and ψ_i, ψ_f the initial and final atomic wave functions. Fermi assumed that the weak interaction involved the product of two four-currents, a nucleon current $\bar{\psi}_f \gamma_\mu \psi_i$, and a lepton current $\bar{\psi}_\nu \gamma_\mu \psi_e$, where ψ_i, ψ_f denote the initial and final nuclear wave functions and ψ_e, ψ_ν the electron and neutrino wave functions. The Hamiltonian is then

$$H_\beta = g \langle \bar{\psi}_f \gamma_\mu \psi_i \rangle \langle \bar{\psi}_\nu \gamma_\mu \psi_e \rangle \qquad (4)$$

where g is the weak interaction coupling constant, analogous to electric charge. This describes process (1) only, but if the Hermitian conjugate term is added to ensure that H_β is real, then the latter describes processes (2) and (3). In negatron emission ψ_e describes a negative energy positron whose absorption is equivalent to the creation of a negatron, while the converse is the case in positron emission. In electron capture ψ_e is simply the wave function of a bound atomic electron. The emission of different types of neutrino in (1), (2) and (3) is based on the principle of lepton conservation, which will be discussed later. Realising that the properties of the leptons involved are described by other covariant forms constructed from the Dirac matrices as well as the vector form, later workers extended the Hamiltonian to include five interactions in all, behaving respectively as scalar, vector, tensor, axial vector and pseudo-scalar under Lorentz transformation.

Until 1956 each of these interactions was associated with a coupling constant $C_{S,V,T,A,P}$ whose square was a measure of the relative strength of the interaction. In 1956, however, the suggestion of Lee and Yang that parity was not necessarily conserved in beta decay, and the subsequent experimental demonstrations by Wu et al and other workers that indeed the process is invariant under neither the parity transformation nor under charge conjugation, led to radical alterations of the theory. Five parity non-conserving interactions were introduced, together with their associated coupling constants $C^1_{S,V,T,A,P}$. The resulting Hamiltonian is

$$H_\beta = g\sum_K \langle \bar{\psi}_f O_K \psi_i \rangle \left[\langle \bar{\psi}_\nu O_K (C_K + C_K' \gamma_5) \psi_e \right] + \text{h.c.} \quad (5)$$

where the O_K represent the five interaction terms

and $$\sum_{K=1}^{5} \left(|C_K|^2 + |C_K'|^2 \right) = 1 \quad (6)$$

Combination of the above experimental results with the well known CPT theorem (Schwinger, 1953; Pauli, 1955) indicates that the process is invariant under time reversal, which in turn demands that the coupling constants be real. This result will be assumed throughout this thesis.

The Plane Wave Approximation

If the Coulomb force between the daughter nucleus and the charged leptons is neglected, then the leptons can be represented by plane

waves of the form

$$\psi_e = \exp(i\,\underline{p}.\underline{r}) \quad \text{and} \quad \psi_\nu = \exp(i\,\underline{q}.\underline{r}) \qquad (7)$$

where $\underline{p}$ and $\underline{q}$ are momenta and $\underline{r}$ is the position vector of the lepton with respect to the nucleus. (The unit system is that where $\hbar = m = c = 1$.) H_β then contains the factor

$$\exp(-i(\underline{p}+\underline{q}).\underline{r})$$

and, since the nuclear radius R is of the order of 10^{-2} and $|\underline{p}+\underline{q}| \leq 1$, this factor can be expanded in a rapidly diminishing exponential series, each term of which imposes certain selection rules on the nuclear transition. An allowed transition is one where the nuclear wave functions are such that the first term of H_β does not vanish, the other terms then being negligible, and the transition proceeding at maximum rate; an n-forbidden transition is one where ψ_i and ψ_f are such that the first non-zero term is that containing the n^{th} power of $(\underline{p}+\underline{q}).\underline{r}$. The selection rules for allowed transitions are listed in Table 1.

TABLE 1.

INTERACTION	SPIN CHANGE	PARITY CHANGE
S,V	0	NO
T,A	0,1 not $0 \rightarrow 0$	NO
P	0	YES

A nucleus N(Z,A) energetically able to decay to N($Z\pm1$,A) will do so by the lowest order transition compatible with the necessary changes in spin and parity. Since the P interaction cannot co-exist with any of the four others, allowed transitions can only contain some or all of the latter, which can be further sub-divided into Fermi (S,V) and Gamow-Teller (T,A) types.

The purpose of most experimental work in the field of beta decay has been to discover the relative magnitudes of the various interactions by evaluating the coupling constants C_K and $C_K{}^1$.

The Allowed Beta Decay Probability

Substitution of the relevant Dirac operators and lepton wave functions in equation (5) yields the following expression for the energy spectrum of the emitted electrons in an allowed decay

$$N_{\pm}(W) = \frac{g^2}{2\pi^3} F(\mp Z,W)\, pW(W_0-W)^2 |M|^2 (1 \mp b/W) \qquad (8)$$

where p and W are the momentum and energy of the emitted electron, and W_0 its maximum energy; (these energies include rest mass). $F(\mp Z,W)$ is the Coulomb correction factor, or Fermi function, (the negative sign holds for positrons and vice versa);

$$|M|^2 = (C_S{}^2 + C_S{}'^2 + C_V{}^2 + C_V{}'^2)|M_F|^2 + (C_T{}^2 + C_T{}'^2 + C_A{}^2 + C_A{}'^2)|M_{GT}|^2;$$

M_F and M_{GT} are the Fermi and Gamow-Teller matrix elements for the nuclear transition;

b is the Fierz interference term, to be discussed in detail later.

A more convenient form of equation (8) is

$$\sqrt{\frac{N(W)}{pWF(Z,W)}} = \text{const.}\ (W_o - W)(1 \mp b/W); \qquad (9)$$

this is called a Fermi-Kurie plot and, if b = 0, should be a straight line intersecting the energy axis at $W = W_o$.

The total decay rate λ is given by

$$\lambda = \int_1^{W_o} N(W)\,dW = g^2 |M|^2 f / 2\pi^3$$

where $$f = \int_1^{W_o} F(Z,W)\,pW\,(W_o - W)^2\,(1 \mp b/W)\,dW$$

The comparative half-life or ft value is defined by

$$ft = f\ \ln 2\ /\lambda = 2\pi^3 \ln 2\ /g^2 |M|^2 \qquad (10)$$

This is a useful quantity for the classification of allowed decays, which fall into two groups - the normal transitions where log ft is about 5, and the super-allowed cases, where log ft is about 3.5.

Fierz Interference

The Fierz term b has the form

$$b = \frac{2\gamma\left(\left[C_S C_V + C_S' C_V'\right]|M_F|^2 + \left[C_T C_A + C_T' C_A'\right]|M_{GT}|^2\right)}{|M|^2} \qquad (11)$$

where $\gamma = (1-\alpha^2 Z^2)^{1/2}$ and $\alpha = 1/137$ (the fine structure constant). It is obviously zero if each of the Fermi and Gamow-Teller interactions

contains only one of the four basic interactions S,V,T and A.

In Fermi transitions the nuclear angular momentum does not change and the spins of the emitted leptons must therefore be anti-parallel; if both the S and V interactions are present, interference will occur between them, giving rise to cross terms of the type $C_S C_V$. Similar terms will occur in the Gamow-Teller case. In the latter, the emitted leptons have parallel spins and thus form a triplet state in contrast to the Fermi singlet state. Since these final spin states are orthogonal, there can be no interference between Fermi and Gamow-Teller radiations.

Before the revolution in beta decay theory in 1956, one could draw specific inferences about the relative magnitudes of the coupling constants from the very small measured values of b (discussed later) without recourse to any other information. For a pure Fermi decay

$$b = 2\, C_S C_V/(C_S^2 + C_V^2) \qquad (12)$$

and thus a value for the ratio C_S/C_V (and similarly C_T/C_A) could be deduced from a measurement of b.

If time reversal invariance were not assumed in the present theory, a zero value for b would not necessarily imply that either C_S or C_V were zero, but might simply reflect the fact that they were equal in magnitude but opposite in phase. Under the simplifying assumption of T invariance, b reduces to the form (11), which is still only of limited usefulness, since a very small value of b can

only yield the information that one interaction (S or V in the Fermi case) is dominant without indicating which.

Recourse is necessary therefore to other experimental information if any use of an experimental measurement of b is to be made. The nuclear recoil experiments performed by Hermannsfeld et al (1958) on various inert gases have shown that

$$C_A^2 + C_A{}^{1\,2} \gg C_T^2 + C_T{}^{1\,2}$$

$$\text{and} \quad C_V^2 + C_V{}^{1\,2} \gg C_S^2 + C_S{}^{1\,2} \qquad (13)$$

thereby establishing the predominance of the VA combination while not ruling out the presence of a small admixture of S and T. Another source of data lies in the results of the work on longitudinal polarisation of the emitted electrons carried out by Fraunfelder et al (1957), and by Deutsch et al (1957); these experiments yield the results

$$C_A = + C_A{}^1 \text{ and } C_V = +C_V{}^1 \qquad (14)$$

The two component neutrino theory of Lee and Yang (1956), which demands a zero neutrino rest mass and a polarisation either parallel or anti-parallel to its momentum, predicts the relationship

$$C_K = \pm C_K{}^1 \qquad (15)$$

where the sign is the same for all four interactions. The above experimental results indicate that the positive sign should be taken. In this case, use of the information presented here leads

to the expression

$$b = 2\left[\frac{C_S}{C_V} + \frac{C_T}{C_A}\,\frac{C_{GT}^{\,2} M_{GT}^{\,2}}{C_F^{\,2} M_F^{\,2}}\right] \Bigg/ \left[1 + \frac{C_{GT}^{\,2} M_{GT}^{\,2}}{C_F^{\,2} M_F^{\,2}}\right] \qquad (16)$$

which can be used to determine upper limits for the ratios C_S/C_V and C_T/C_A.

If the negative sign is in fact correct for S and T, then equation (11) automatically yields a zero value for b.

The restriction (15) imposed by the two component theory is linked to the assignment of helicity to the neutrino and to the law of lepton conservation. The latter assigns a leptonic number +1 to the negatron and neutrino, and -1 to the positron and the anti-neutrino, and demands conservation of total lepton number in any process. The assignment is justified by the experiments of Davis (1955, 1958) and of Reines et al (1960), who have demonstrated the existence of two different types of neutrino. The electron polarisation experiments mentioned previously show that in negatron decay, where lepton conservation requires the emission of an anti-neutrino, the latter has positive helicity, and similarly that neutrinos from positron decay have negative helicity. These assignments of helicity are supported by the investigation of Goldhaber et al (1958) into the electron capture decay of europium 152, which showed that the neutrino emitted in a Gamow-Teller transition has in fact negative helicity. The two component theory only predicts the observed

helicities if the positive sign is used in equation (15).

If the two component theory is not correct and the S and T coupling constants are arranged so that $C_S = -C_S^1$ and $C_T = -C_T^1$, then b is automatically zero. This result follows physically from the argument that in the V and A interactions, neutrinos of a certain helicity are generated, and if the negative sign is used for S and T, then neutrinos of the opposite helicity are generated; there can be no interference between neutrinos of opposite helicity due to the orthogonality of the final spin states and hence b must be zero.

2. ORBITAL ELECTRON CAPTURE AND K-CAPTURE TO POSITRON EMISSION RATIOS IN ALLOWED TRANSITIONS

The process

$$N(Z,A) + e^- \rightarrow N(Z-1,A) + \nu \qquad (17)$$

is called orbital electron capture, and has been observed in over 150 isotopes. Capture from the K shell predominates in most decays, except in some cases of high atomic number where K capture is energetically impossible and the L shell is the lowest level from which electrons may be captured.

The theoretical description of the electron capture process is similar to that of electron emission apart from some simplifying differences. The electron wave functions are now those for bound states of unique energy and therefore the neutrino energy is also discrete. Since the angular momentum of the bound electron is fixed, no summation over angular momenta occurs in the derivation of the expression for the transition probability.

Energy balance in β^+ emission and electron capture.

a) β^+ emission: If the principle of conservation of energy is applied to the process

$$N(Z,A) \rightarrow N(Z-1,A) + e^+ + \nu \qquad (18)$$

then, neglecting the nuclear recoil energy and the change in atomic binding energy,

$$\Delta M_a = M(Z,A) - M(Z-1,A) = W + mc^2 + W_\nu$$

where the M's represent the initial and final atomic masses, and mc^2 the electron rest mass. Positron emission is only possible therefore if the condition

$$\Delta M_a \geqslant 2mc^2 \qquad (19)$$

is fulfilled.

The positron has its maximum energy Wo when the neutrino carries off no energy; in this case

$$Wo = \Delta M_a - mc^2$$

The transition energy ΔM_a may then be found by a measurement of the end point energy of the positron spectrum, or by the other methods which are discussed below.

b) Electron capture: In the case of the transition (17) use of the same approximations leads to the energy balance

$$\Delta M_a = W_\nu + |En|$$

where En is the binding energy of the captured electron. Capture is therefore energetically possible if

$$\Delta M_a \geqslant En \qquad (20)$$

Thus, if positron emission is detected in a particular isotope, electron capture can also be expected, although the converse is not true.

The transition energy ΔM_a

Obviously ΔM_a cannot be directly measured in an electron capture decay, and if $\Delta M_a < 2mc^2$, so that there is no positron emission, then it has to be evaluated by another method; it can be obtained from the maximum energy of the internal bremsstrahlung spectrum accompanying electron capture, but a more convenient and very accurate method is by measurement of the Q value of the (p,n) reaction whereby the daughter nucleus of the decay is transmuted to the parent. In this reaction,

$$\Delta M_a = -Q - (\text{neutron-proton mass difference}) \qquad (21)$$

In most cases, including those of ^{11}C and ^{19}Ne, (p,n) reaction measurements yield much more precise values for Wo than do direct measurements of the positron end point (see e.g. Mattauch et al, 1965).

K/β^+ Ratios

The probability of allowed capture from the K shell is given by the expression

$$\lambda_K = \frac{g^2}{4\pi^2} (Wo + W_K)^2 |M|^2 g_K^2 (1 + b) \qquad (22)$$

where W_K = total energy of K-shell electron;

and g_K = K electron wave function evaluated at the nuclear radius.

The ratio of K capture to positron emission in an allowed

decay is therefore

$$R = \frac{\frac{\pi}{2}(W_0+W_K)^2 g_K^2 (1+b)}{\int_1^{W_0} F(-Z,W)\, pW (W_0-W)^2 (1-b/W)\, dW} \qquad (23)$$

The electron radial functions g_K were first given by Bethe (1933) for the case of a point charge and without considering screening. More accurate calculations were performed by Brysk and Rose (1955, 1958), who made allowance for screening and for the finite volume of the nucleus; these results were published in the form of a curve giving g_K (averaged over the nuclear volume) as a function of atomic number, but the numerical values of g_K^2 for various positron emitters have been given by Depommier, Nguyen-Khac and Bouchez (1960). At values of $Z<15$, the screening correction is rather badly known (Depommier, 1967) and values of g_K^2 in this region may be in error by several percent.

Theoretical values of K/β^+ ratios have been calculated by Zweifel (1957), Perlman et al (1958) and Depommier et al (1960). In calculating the positron spectrum, the latter authors used the screening corrections given by Feister (1952) and the second order terms in the Fermi function expansion given by Dzelepov and Zyrjanova (1952); they then calculated K/β^+ ratios for various allowed transitions, the values being given later in this chapter.

Unfortunately, there are few calculations of theoretical K/β^+ ratios in the very low atomic number region where the present

interest lies, and is not possible to use the graphs of Brysk and Rose with any accuracy here. However, corrections to the positron spectrum for screening and for finite nuclear size are very small at low Z, and it is possible to obtain the positron intensity by graphical integration over the positron spectrum. Computer programmes have therefore been written in K.D.F.9 Algol by the author and by Mr. W. Leiper to evaluate the necessary Fermi functions and to perform this integration accurately; these are described in Chapter Five.

The Fierz Interference Term b

The term b may be evaluated by comparing an experimentally measured K/β^+ ratio R with the theoretical ratio Ro evaluated for zero b: from equation (23)

$$R = \frac{Ro(1+b)}{1-b\langle W^{-1}\rangle}$$

$$\text{where } \langle W^{-1}\rangle = \frac{\int_1^{W_0} F(-Z,W)pW(W_0-W)^2\frac{1}{W}dW}{\int_1^{W_0} F(-Z,W)pW(W_0-W)^2 dW} \qquad (24)$$

and b is then given by

$$b = \frac{R-Ro}{Ro+R\langle W^{-1}\rangle} \qquad (25)$$

This relationship yields an extremely sensitive test for Fierz interference, and, accordingly, measurement of K/β^+ ratios

has frequently been used as a test of the validity of beta decay theory. An alternative method for the measurement of b is the analysis of Fermi-Kurie plots for deviations from linearity. Experimental results from both these methods of attack on the theory of beta decay will be discussed later.

Overlap and Exchange Effects

Most previous measurements of K/β^+ ratios have been interpreted solely in terms of the estimation of an upper limit to the Fierz parameter. However, account must now be taken of the work of Bahcall (1963), who has predicted small deviations from conventional theory due to overlap and exchange effects in beta-decay processes.

The basic departure of Bahcall's work from the theory outlined earlier in the explicit inclusion of atomic states in the description of a nucleus undergoing beta decay. This idea was first suggested by Benoist-Gueutal (1950, 1953) and was developed by Odiot and Daudel (1956) for specific nuclei. Bahcall has succeeded in overcoming the mathematical difficulties involved and has made detailed calculations over a range of atomic number from Z = 14 to 38.

The change of nuclear charge by one unit in beta decay means that the electrons in the initial and final states are in different potentials, so that the initial and final atomic states do not overlap perfectly. Secondly, since electrons are indistinguishable,

the observation of a K vacancy need not necessarily imply that a K electron has been captured; capture may have occurred from a higher shell and the resulting vacancy then exchanged with the K shell. Both these points are ignored in the expression (23) for the K/β^+ ratio, the necessary corrections to the conventionally calculated ratio being called "overlap and exchange" corrections.

Using the analytic wave functions of Watson and Freeman (1961), Bahcall has made extensive calculations of the magnitudes of these effects. He finds that K/β^+ ratios should be slightly smaller than expected from conventional theory, the correction factor B_K being a function of the atomic number Z of the parent nucleus, and varying from 0.924 at Z = 14 to 0.984 at Z = 38. Similar predictions concerning the magnitudes of L/K branching ratios are in good agreement with experimental results (Bahcall, 1965); however, the precision required to detect the effects discussed on K/β^+ ratios in the quoted range of atomic number has not yet been attained. Bahcall has not extended his calculations to lower atomic number, the validity of the assumptions involved in the theory being uncertain at small values of Z (Bahcall, 1966).

3. EXPERIMENTAL TECHNIQUES AND DATA ON K/β^+ RATIOS

When a K electron is captured by the nucleus, the daughter atom has a vacancy in its K shell and an extra outer electron; during the

subsequent de-excitation, Auger electron and x rays are emitted. At low Z, the fluorescence yield is very small (e.g. Fink, 1965) so that K capture manifests itself predominantly by emission of Auger electrons of energy approximately equal to the K binding energy of the daughter less twice the L binding energy.

The experiments to be described in this thesis may be classified as direct measurements, where the intensity of these soft radiations is measured directly, along with the intensity of the accompanying positron spectrum. In the alternative approach, applicable only when the decay takes place to an excited state of the daughter nucleus, coincidences are taken between the de-excitation gamma rays and the positrons, giving the positron intensity as a fraction of the total decay rate; the ratio of electron capture from all shells to positron emission can then be deduced.

1) Direct Spectrometry:-

Direct detection of the Auger electrons and x rays can be carried out using sources either external to the detector or dispersed within it. In the external source method, difficulties are encountered due to self-absorption and self-scattering of these radiations; at low atomic number, where the fluorescence yield is small, very few of the Auger electrons emerge from a solid source; if only the x rays are detected it is then necessary to know the

fluorescence yield to obtain the total capture rate from the K shell, and such yields are not at present accurately known.

None of these difficulties is encountered if the source is uniformly dispersed throughout the sensitive volume of the detector. This is easily done in the case of a proportional counter by using a gaseous source; this detector is particularly suitable at low values of Z. At higher Z where escape of x rays from a gas counter becomes a problem, a scintillation counter may be suitable, since often a crystal can be grown with the source distributed uniformly throughout its volume. The only major limitation on the precision attainable with these techniques is the accuracy with which the pulses produced by positrons can be separated from those resulting from K capture. The different methods of effecting this separation in the case of the proportional counter are discussed in detail in Chapter Two.

2) Coincidence Spectrometry:-

When the decay takes place to an excited state of the daughter nucleus, the ratio of total electron capture to positron emission may be determined by taking coincidences between the de-excitation gamma rays and the positrons. The gamma rays are detected by a scintillation counter and the positrons by either a proportional counter or a lens spectrometer; the detector efficiences have to

be calibrated accurately, and the use of solid sources, however thin, necessitates corrections for back-scattering of positrons.

The K/β^+ ratio can be deduced from the total capture to positron ratio if the ratio $(L + M + \ldots)/K$ is known. Any uncertainty in the latter then leads to an uncertainty in the K/β^+ ratio over and above the experimental error.

Experimental Results

The most recent reviews of experimental measurements of K/β^+ ratios are those of Depommier et al (1960) and Berényi (1963). In Table 2, the more accurate of the available measurements on light nuclei are listed and compared with theory. Most of these are indirect measurements (denoted by I), giving the total capture to positron ratio, and in these cases, the present author has deduced the K/β^+ ratio using the $(L + M)/K$ ratio. The L/K ratios employed are those of Brysk and Rose (1955) with overlap and exchange corrections by Bahcall (1963a), and are known to agree well with measured values; the M/L ratios used are those most recently published (Bahcall, 1963b) and have yet to be compared extensively with experiment. (No correction was applied to the L/K ratio of ^{22}Na, there being no information available for Z<15).

The only case where an author claims specifically to have detected a finite Fierz term is the $4\pi\beta\gamma$ coincidence experiment on ^{22}Na by Williams (1964), who finds $b = -(5 \pm 1.2)\%$. According

TABLE 2

COLLECTED RESULTS ON K/β^+ RATIOS IN LIGHT ISOTOPES

Nucleus	Measured K/β^+ ratio	Ref.		Theoretical ratio (Depommier, 1960)	B_K	% Fermi
^{11}C	$(1.9\pm.3)\times10^{-3}$	Scobie and Lewis (1957)	D	2.18×10^{-3} *	-	63
^{13}N	$(1.68\pm.12)\times10^{-3}$	Ledingham et al (1963)	D	1.78×10^{-3} **	-	78
^{18}F	$.03\pm.002$	Drever et al (1956)	D	.029	-	0
^{22}Na	$.103\pm.006$	Sherr and Miller (1954)	I	.107	-	0
	$.115\pm.010$	Allen et al (1955)	I			
	$.103\pm.008$	Konijn et al (1958)	I			
	$.105\pm.004$	Ramaswamy (1959)	I			
	$.098\pm.001$	Williams (1964)	I			
^{44}Sc	$.064\pm.016$	Blue and Bleuler (1955)	I	.042	.969	0
	$.020\pm.019$	Konijn et al (1958)	I			
^{48}V	$.59\pm.10$	Sterk et al (1953)	I	.66	.973	0
	$.66\pm.08$	Bock (1955)	I			
	$.65\pm.02$	Konijn et al (1958)	I			
	$.66\pm.02$	" " " "	I			

TABLE 2 (continued)

Nucleus	Measured K/β^+ ratio	Ref.		Theoretical ratio (Depommier, 1960)	B_K	% Fermi
^{52}Mn	1.67±.15	Good et al (1946)	I	1.77	.976	0
	1.80±.20	Sehr (1954)	I			
	1.77±.07	Konijn et al (1958)	I			
	1.82±.08	" " " "				
^{58}Co	5.19±.18	Cook et al (1956)	I	4.87	.978	0
	5.19±.18	Good et al (1946)	I			
	5.07±.15	Konijn et al (1958)	I			
	4.91±.17	Ramaswamy (1961)	I			
	4.92±.09	Joshi and Lewis (1961)	D			
	4.83±.1	Kramer et al (1962)	D			

(* calculated by present author; ** calculated by Ledingham)

Notes

1) D denotes a direct measurement; I an indirect one.

2) Results for heavier isotopes are summarised by Berényi (1963).

to Murthy and Ramaswamy (1964) overlap and exchange effects do not alter the K/β^+ ratio for this decay significantly, but it appears that these authors have applied to K capture a correction which is applicable only to the total capture rate. The deviation could therefore be due to these effects, or to an error in the K wave function or L/K ratio. Further, the ^{22}Na decay has several unusual features for an allowed transition, and the deviation might be related to these.

An alternative method for the measurement of the Fierz term is the analysis of beta spectrum shapes; if b is non-zero, then the Fermi-Kurie plot will be non-linear. This method has been developed by several workers to a high degree of sophistication, and Langer et al, among others, have observed deviations from linearity for various isotopes, including ^{22}Na and ^{31}P; these deviations correspond to the presence in the expression for the allowed beta decay probability of a shape factor $(1 + b/W)$, where $0.2 < b < 0.4$. However, it is found that b has the same sign for negatrons as for positrons, and therefore cannot be due to Fierz interference. No adequate theoretical explanation of these observations has been proposed.

4. OBJECTIVES OF THE PRESENT WORK

It will be clear from the foregoing sections that there is a need for accurate theoretical calculations of K wave functions and

and of overlap and exchange effects for both K/β^+ and L/K ratios, in the range $Z < 15$. On the experimental side, Table 2 shows that, while there have been various K/β^+ measurements in Gamow-Teller decays, there have been few in Fermi decays, and few in the region $(14 < Z < 20)$ where the deviations predicted by Bahcall might be large enough to observe.

It would therefore be of interest to augment the experimental data at values of Z up to about 20, especially for Fermi decays; the properties of various such decays are summarised in Table 3. Several of the parent isotopes can be obtained in the form of gases suitable for use in proportional counter measurements. The work described in this thesis constitutes part of a series of measurements, which, together with the development of the requisite experimental techniques, is being carried out at Glasgow. It might be hoped that the results obtained from this work would stimulate some advance in the corresponding theory.

TABLE 3

PROPERTIES OF VARIOUS PURE OR 'PARTIALLY' FERMI TRANSITIONS

Nucleus	Half-life	Fermi Fraction (%)*	K binding energy of daughter (keV)	B_K
^{11}C	21 min.	63	.19	NOT CALCULATED
^{13}N	10 min.	78	.28	
^{14}O	72 sec.	100	.40	
^{15}O	2 min.	73	.40	
^{17}F	70 sec.	38	.53	
^{19}Ne	18 sec.	31	.63	
^{26}Al	7 sec.	100	1.30	
^{27}Si	4 sec.	74	1.56	.924
^{34}Cl	1.6 sec.	100	2.47	.954
^{35}A	1.8 sec.	93	2.82	.959

*Fermi fractions for mirror transitions calculated from ft values given by Konopinski (1966)

CHAPTER TWO

EXPERIMENTAL TECHNIQUES FOR MEASUREMENT OF K/β^+ RATIOS

1. INTRODUCTION

The direct measurement of K/β^+ ratios in isotopes of low atomic number displays a combination of problems which effectively delimits the choice of techniques suitable for such a project. Firstly, the low energy of the Auger electrons and x rays emitted following electron capture requires that the source be in the form of a gas in order to avoid self-absorption, and therefore dictates the use of a gas proportional counter. Secondly the short half-lives of most of the isotopes of interest necessitate that sources either be produced continuously in the vicinity of the counter and flowed through it at a rate determined by the half-life, or in the case of isotopes whose half-life is only a few seconds, produced within the counter itself and detected immediately after the production process has ceased. These two methods will be considered in outline in this chapter, and in detail in the later chapters dealing with the measurements performed on carbon 11 and neon 19.

A proportional counter is normally filled with a mixture of an inert gas such as argon or neon, plus a smaller amount of an organic quenching agent such as methane or propane. It is therefore immediately obvious that ^{11}C, ^{13}N, ^{19}Ne and ^{35}A would be

extremely convenient isotopes to study, provided they could be manufactured in quantity. As stated in Table 2, the K/β^+ ratio in ^{13}N has been measured by Ledingham et al; in this thesis the measurements made by the author on ^{11}C and ^{19}Ne are described. Most other common gases, especially electro-negative ones such as oxygen and chlorine ruin the operation of a proportional counter due to the process of electron attachment to gaseous atoms; however ^{15}O might be studied using CO_2 and ^{34}Cl using methyl chloride.

2. THE CONTINUOUS FLOW TECHNIQUE

Continuous flow counters have often been used and their operation presents few difficulties provided certain precautions are taken. If the counter gases are derived from different sources, they must be well mixed before entering the counter, otherwise the energy resolution will be poorer than in the static case. It is also necessary that the flow rates be very accurately stabilised so that the partial pressures of the different gases remain constant; if these pressures vary even slightly, the resulting change in the gas gain of the counter will give rise to considerable drifts in the mean pulse height of the peak being observed. Considerable care was taken over the design of the flow system employed in the carbon 11 experiment and this is discussed in detail in Chapter Five.

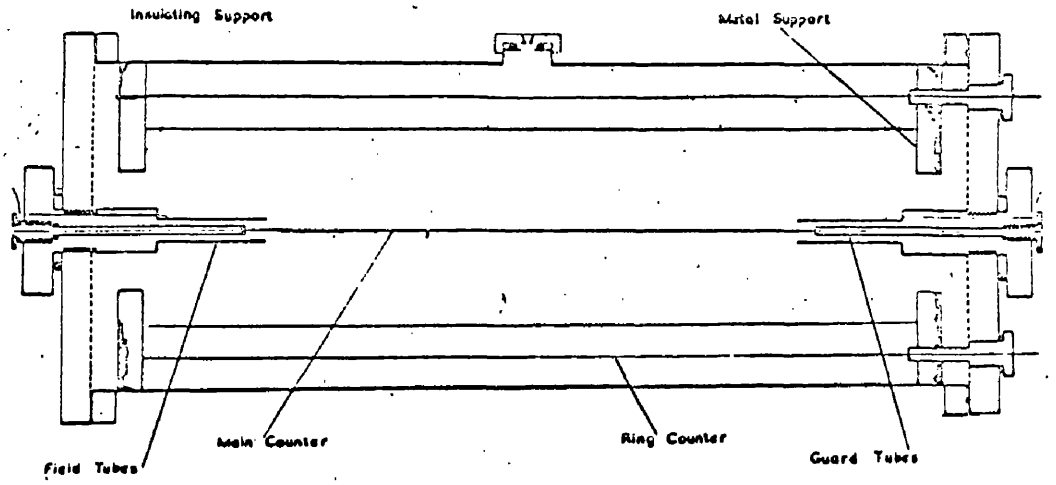
The continuous production of a gaseous isotope for measurement in a proportional counter can be effected conveniently by flowing

the gas through a beam of x rays or charged particles from an accelerator or through a nuclear reactor, depending on the type of isotope required. In the present case, where positron emitters, neutron-deficient nuclei, were required, the gas was exposed to the bremsstrahlung x-ray beam from the 300 MeV Glasgow electron synchrotron, and the isotope of interest produced by the (γ,n) reaction on the most abundant stable isotope present.

Wall-less counters

The third of the basic problems mentioned at the start of this chapter arises when the counter itself is considered; at low values of atomic number, K/β^+ ratios are very small, of the order of 1 in 1000. The result of this is that in a conventional proportional counter, the K capture peak tends to be obscured by a continuous background of pulses, arising from the "wall effect"; since the positrons have ranges much greater than the counter dimensions, they lose only a small fraction of their energy inside the counter, giving rise to a continuous pulse height spectrum, which rises steeply at low energies.

One solution to this problem is to confine the radioactivity to the centre of the counter, so that all the positrons produce large pulses, which are clearly distinguishable from the small K capture pulses, before escaping from the counter (a technique



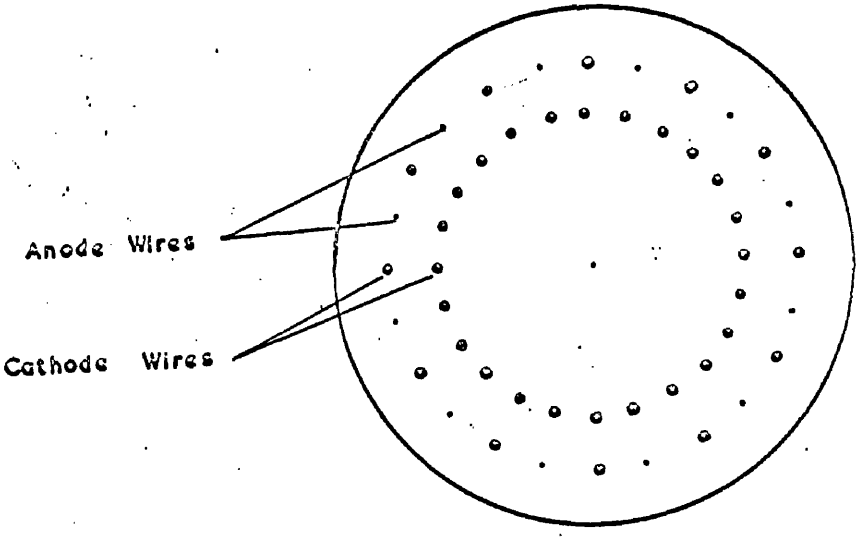


Fig. 1. Schematic diagrams of multiwire counter

described later). The solution most easily allied with the flow technique is the use of a "wall-less" counter of the type introduced by Drever, Moljk and Curran (1957). This consists of a central proportional counter completely surrounded by a ring of counters operated in anticoincidence with it, so that a particle escaping from the central counter identifies itself by producing a pulse in the ring counter. It is then possible to distinguish between short-ranged particles born in the central counter without sufficient energy to reach the ring, and higher energy particles, which are detected by both counters.

The basic form of wall-less counter is the multiwire counter shown diagramatically in fig. 1; this consists of a central proportional counter whose cathode is defined by an array of wires placed cylindrically round the central anode wire and joined externally to the outer case. Between this array and the case is a second array of alternate anode and cathode wires. Each anode in this array is surrounded by a cathode consisting of several wires together with part of the case, and the ring therefore contains a set of proportional counters. Although this type of counter was originally designed for the measurement of L/K branching ratios, it may be applied to the measurement of K/β^+ ratios of half-life at least a few hours. However, it is not totally suited to the investigation of short-lived sources, for the following reason:- although the detection of K events in the central counter is straightforward, measurement

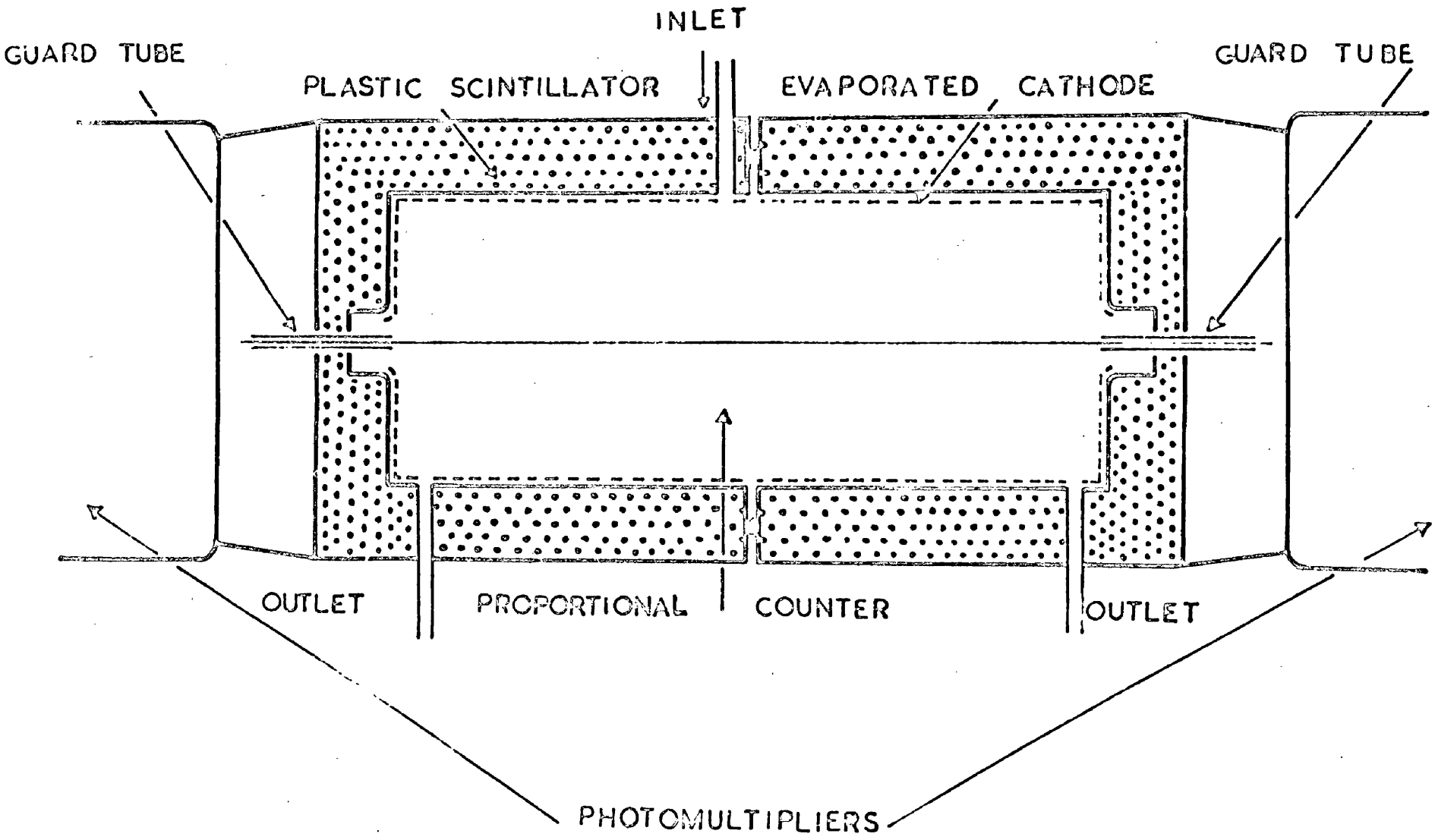


Fig. 2. Schematic diagram of plastic counter

of the positron counting rate in this region demands that the source be evenly distributed throughout the counter. The positron rate in both counters together is measured and multiplied by the ratio of the volumes to give the counting rate in the centre; when the gas is flowing through the counter, it is extremely unlikely that the activity will be evenly distributed and therefore the positron count rate is uncertain.

To solve this problem it is necessary to maintain physical separation between the central and ring counters, confining the radioactive gas to the former. It must be ensured, however, that the barrier between the counters is designed to minimise absorption of escaping positrons, otherwise the anticoincidence efficiency will be decreased. Two types of counter meet these requirements.

a) The Plastic Counter

The counter employed in the measurement of K/β^+ in carbon 11 consisted of a central proportional counter surrounded by a shield of plastic scintillator to act as the anticoincidence detector. The radioactive gas flowed through the central counter, in which the Auger electrons and x rays resulting from K capture were detected, while the positrons had sufficient ranges to be detected in both counters. The spectrum obtained when anticoincidences were taken then contained the K peak.

This counter, originally designed by Moljk and Drever (1957)

for low background applications, is shown in Fig. 2. The basic component is a hollow cylinder of plastic scintillator (divided in two sections) having walls 1.9 cm thick along its length and 3.2 cm thick at the ends. An aluminium coating, a few hundred angstroms thick, evaporated onto the inner surface, serves as the cathode for the proportional counter, several high voltage connections being made to it. The central counter, 29.2 cm in length and 8.9 cm in diameter is fitted with earthed guard tubes but not with field-correcting tubes. The central wire, 0.01 cm in diameter, is of spectroscopically pure nickel.

The light output from the scintillator is analysed by a five inch E.M.I.6099F photomultiplier at each end, coupled to the scintillator by perspex light guides. All junctions are optically coupled with silicone oil. The outer walls of the scintillator are covered with 0.002 cm thick aluminium foil to prevent the escape of light, which would decrease the light collection efficiency. Calibration of the central counter can be carried out by allowing x rays from an external source to pass through two windows drilled in the plastic. The whole counting system is operated within an optically sealed brass can.

Although well suited to the measurement of K/β^+ ratios, the plastic counter was found during the work on carbon 11 to have several shortcomings. The choice of flow rate was restricted by the necessity to ensure that the radioactivity had decayed to

a low level before reaching the ends of the counter, since the electric field in this region was distorted owing to the lack of field tubes; the fitting of field tubes to the counter was not possible. Secondly the constant outgassing from the large surface area of plastic led to the presence of impurities in the counter filling with the result that the energy resolution was rather poorer than that attainable in a conventional counter. In order to detect as many of the escaping positrons as possible, it was necessary to operate the photo-multipliers at a high gain, with a resultant high counting rate (about 200,000 per minute) in the anticoincidence circuitry due to multiplier noise. This led to problems which will be discussed in the section dealing with the electronic circuitry. Finally extreme care had to be exercised when handling this counter in order to avoid causing cracks in the plastic scintillator. With these, albeit minor, defects in mind, an alternative type of anticoincidence counter - a foil counter - was proposed by Dr. K. W. D. Ledingham, and designed and put into operation by the author.

b) The Foil Counter

This is simply a development of the multiwire counter in which the ring of cathode wires which defines the boundary between the centre and the ring counters is replaced by a very thin cylindrical foil; to minimise absorption of positrons, this foil

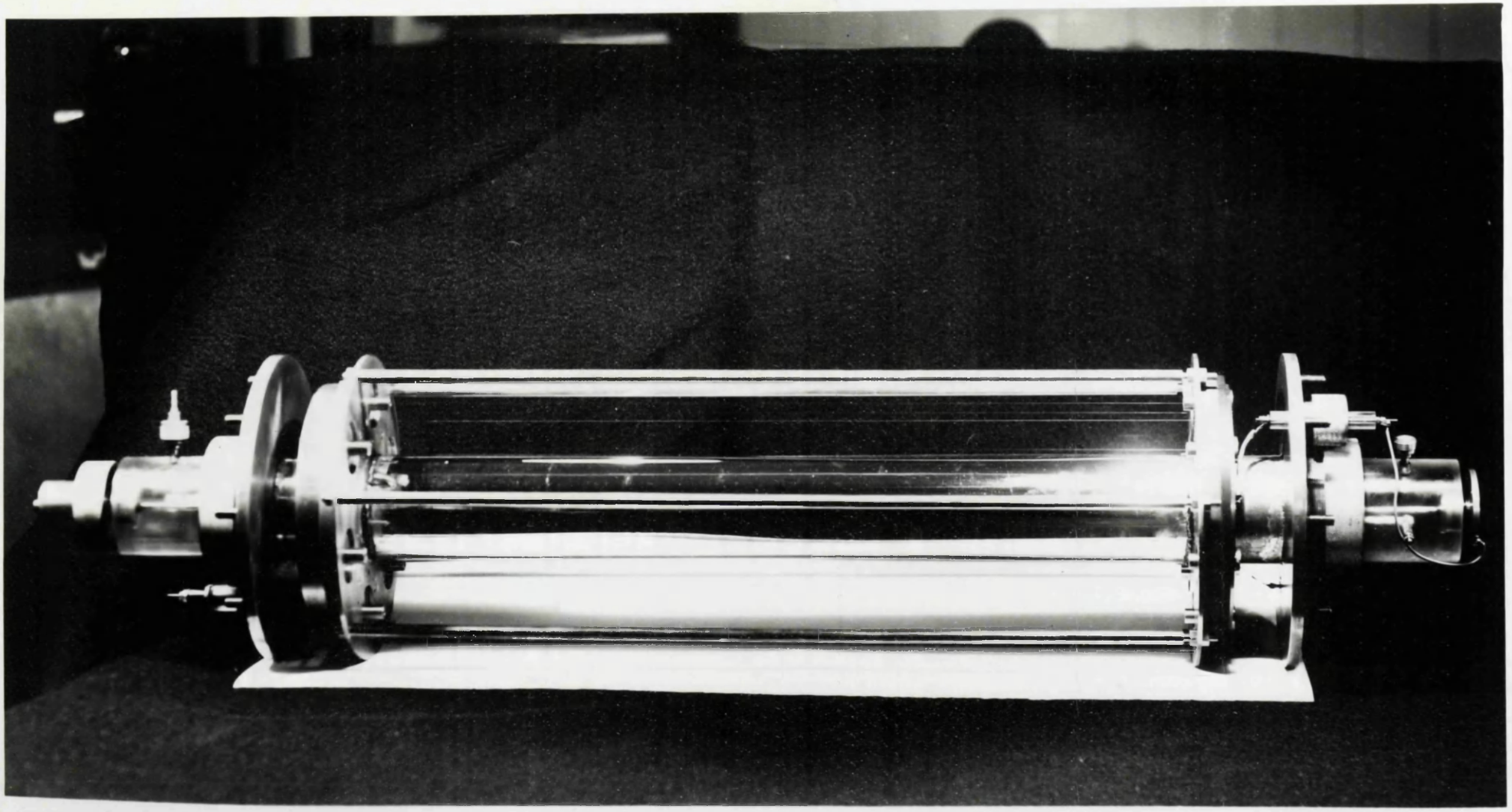


Fig. 3. Foil Counter

should be of a material of low atomic number, and should be as thin as possible compatible with the strength required to maintain its shape along the length of the counter. It must be made of, or coated with, a conducting material, so that the requisite voltage may be applied to it. The counter built by the author is shown in fig. 3; the foil is of mylar, coated on both sides with aluminium, the total thickness being 25 microns. The radioactive gas flows through the inner cylinder, while the ring is supplied with gas from a separate source. Positrons of energies greater than about 30 KeV should therefore penetrate the foil and enter the ring counter. The latter must be thick enough for the positrons to produce a useful amount of ionisation; if this were not the case, the statistical fluctuations in ionisation (first discussed by Landau, 1944) could result in a decrease in the anticoincidence efficiency.

The main difficulty in the construction of this counter lay in the mounting of the foil in such a way that it remained perfectly cylindrical along the length of the counter. The counter was built in three separate sections: the two end assemblies could be connected by six tie rods lying along the circumference, and then slid into the third section, the outer casing. Each end assembly includes a hollow cylindrical section of diameter 7.5 cm and length 2.5 cm which serves as a support for the end of the foil. The foil was initially held tightly round a long smooth cylinder of brass and the two edges joined with a layer of very thin araldite; it was then

slid onto its mountings and made secure at each end by a thin ring of araldite. Any relaxation of the foil occurring during the hardening of this araldite was compensated for by adjusting the nuts on the tie rods to move the end assemblies very slightly further apart.

The length of the central counter is 61 cm and its diameter 7.5 cm; it is fitted with earthed guard tubes and with field tubes. The six outer counters, fitted with earth tubes only, are several centimetres longer but have the same diameter, so that the same high voltage supply may be used for both the centre and the ring. All the wires are made of tungsten, the anodes being 0.075 mm in diameter and the cathodes 0.125 mm.

One point of interest lies in the introduction of the radioactive gas into the central counter. To avoid end effects, it must enter at the centre; this is done by flowing it through a very thin stainless steel hypodermic needle (internal diameter 0.2 mm) lying along the inside of the foil. The very small volume of this tubing ensures that at typical flow rates, the time spent by the gas inside it is only a minute fraction of a half-life; if this were not the case, so that appreciable decay occurred within the tube, positrons passing through its walls would be detected by the counter, giving a value for the K/β^+ ratio which would be smaller than the true one by an uncertain amount. The gas leaves the central counter through holes drilled in the perspex insulators.

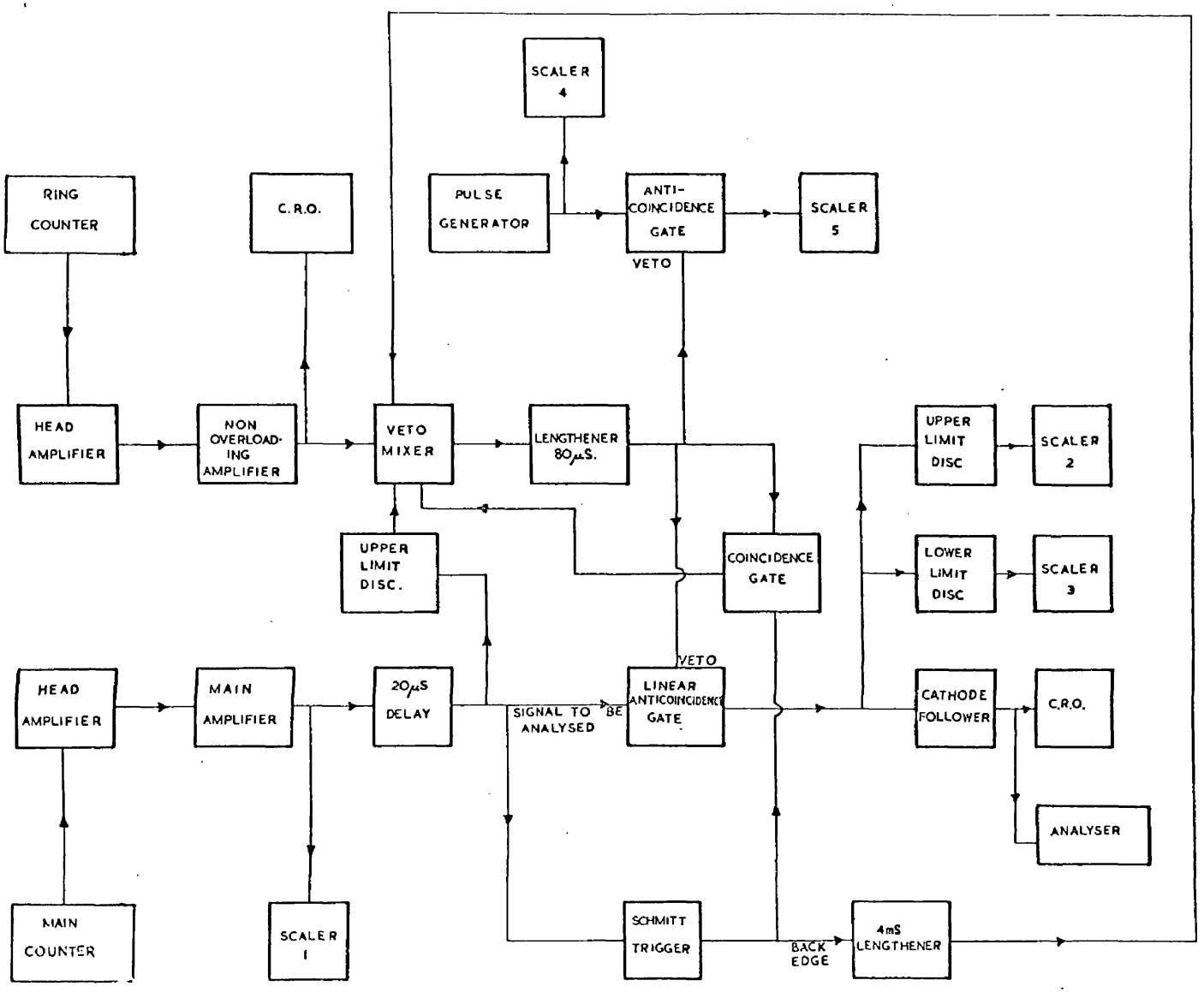


Fig. 4. Electronic arrangement employed in conjunction with wall-less counter.

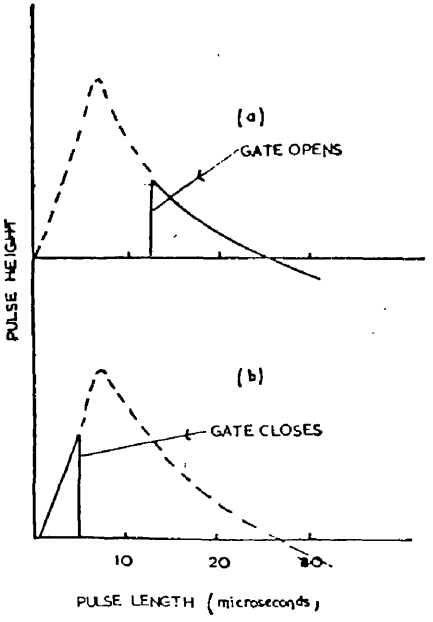


Fig. 5. Pulse degradation due to operation of gate.

This counter has not yet been employed in a measurement but is ready for use at the present time. It avoids the basic disadvantages of the plastic counter and is in fact the only counter which has been designed specifically for the measurement of K/β^+ ratios in short-lived sources; the author expects that it will be used when the Glasgow linear accelerator becomes available for the production of high source intensities.

A final point of considerable importance regarding both the counters described here is that the anticoincidence operation removes pulses due to natural background with very high efficiency.

Electronic circuitry

The electronic circuits used in conjunction with wall-less counters have been described in detail by Drever (1958) and Dougan (1961). Certain additions were made, dictated by the requirements of the experiments on ^{11}C and ^{19}Ne, but the basic circuitry (shown in fig. 4) was retained in full.

The basic circuit is an anticoincidence gate which transmits the pulse from the central counter only if it is unaccompanied by a pulse from the ring counter. The various units within the gate are D.C. coupled where possible to minimise distortion of pulses which arrive close behind large pulses which would overload an A.C. coupled system. Another source of pulse distortion, namely movement of the base-line, is minimised by using double differentiation

in the amplifier. The circuitry includes a facility which immediately re-closes the gate should it happen to open while a signal pulse is decaying, as in fig. 5(a); this prevents a pulse from being registered with a height which is smaller than the true height. It was found necessary in the ^{11}C experiment to introduce additional circuitry to prevent such degradation occurring on the rising edge of a pulse, as in fig. 5(b).

An important feature of this system is the paralysis unit, which injects a pre-selected dead time after every signal pulse, in order to reduce the number of after-pulses; the latter are discussed on page 39. This paralysis causes the gate to be closed for a certain fraction of the actual running time, depending on the counting rate; the total and open times are recorded by an auxiliary multivibrator circuit, whose pulses are counted by two scalers, one coupled to its output directly and the other via an anticoincidence gate operated in parallel with the main gate.

These scalers, together with that which counts all pulses from the central counter were transistorised Harwell 2000 series models. Sensitive discriminator units for the scalers, having bias levels which were insensitive to temperature changes, were designed by the author.

3. THE DIRECT IRRADIATION TECHNIQUE

It was pointed out earlier that an alternative method of overcoming the wall effect is to confine the radioactivity to the centre of the counter; this is possible if the half-life of the isotope concerned is a few seconds or less. In this technique a static counter is used and the activating beam is passed through it. Provided that the beam is kept well clear of the walls, the activity will be confined to the centre, and the decay will be over before many radioactive atoms have diffused to the walls. All the positron pulses should be large, with few small wall-effect pulses to obscure the peak.

This new method was suggested by Dr. R. W. P. Drever for the examination of the isotopes ^{35}A and ^{34}Cl, which will be available from the intense x-ray beam of the linear accelerator. It would appear that, if certain precautions are taken, one could dispense with the complicated anticoincidence systems used in the previous work on longer-lived isotopes.

If the beam is passed axially along the counter, then the counter diameter should be as large as is commensurate with keeping cosmic ray background to an acceptable level. If it is passed diametrically across, however, and if the counter is long enough, end corrections will be much smaller. The beam must not strike metallic parts of the counter in case undesired activities are produced, and the

entrance and exit windows should be made of a material which will not give rise to activities of short half-life. A background of ^{11}C will, of course, build up due to the carbon in the quenching gas, but this can be estimated accurately by analysing the induced activity as a function of time as well as of pulse height.

Such an analysis requires the use of a multi-channel pulse height analyser whose memory can be split up to provide several independent analysers. The counter is irradiated for two to three half-lives of the isotope of interest, and, after the resulting ionisation has cleared, the pulse height spectrum is recorded in successive sub-groups, each of which counts for a pre-determined time; this time is chosen so that nearly all of the activity of interest is recorded in the first few sub-groups, and the later sub-groups register mainly ^{11}C and background pulses. It is then relatively simple to subtract off the latter contribution, leaving the desired spectrum.

When this technique was suggested, the linear accelerator was over a year from completion and it was therefore decided to carry out a preliminary experiment on the synchrotron during this period. This work is discussed in detail in Chapter Six.

4. THE INTERPRETATION OF PULSE HEIGHT SPECTRA

Whichever technique is employed to separate the positron pulses from the K-capture events, the pulse height distribution due to the

latter is always obscured to some extent by a continuous spectrum of pulses which rises steeply at low energies.

Part of the steeply rising section of this spectrum is thought to be due to negative ion after-pulses. Negative ions, formed by capture of electrons by gaseous atoms or molecules, drift slowly to the central high field region; here the electron is detached from the ion and, as it accelerates towards the wire, produces an avalanche of electrons, giving a small pulse. The intensity of this avalanche spectrum can be decreased by using a paralysis time longer than the ion drift time, (typically several msec.) but this paralysis cannot be increased indefinitely due to the resultant decrease in counting rate. Another approach to this problem is purification of the filling gas, discussed in the relevant chapters.

A second source of pulses below the K peak is electron capture from other shells; if the binding energy of the shell in question is less than about 30 eV a single electron avalanche spectrum (to be discussed in Chapter Four) will result, while at higher energies a peak will be produced.

Another contribution to the continuous spectrum arises from the wall effect. Low energy positrons, together with higher energy positrons which have eluded the precautions taken to minimise the wall effect, give rise to a spectrum of pulses which extends fairly linearly above a K peak; it seems reasonable to assume that it

extends similarly below.

The precision with which a K/β^+ ratio can be determined using a proportional counter depends almost entirely upon the accuracy with which the K peak can be separated from the continuous spectrum produced by the above processes. Ideally this separation would be carried out by recording a positron spectrum similar to the one under investigation, and subtracting it, suitably normalised, from the experimental spectrum; this is impossible, however, since positron emitters also produce electron capture peaks in the energy region of interest. It is not possible to use a negatron spectrum, since those β^- spectra similar to the positron spectra would have to be in the form of gases, such as ^{19}O or ^{16}N which would alter the conditions in the counter.

An alternative approach to this problem is the use of a computer, which can be programmed to subtract various backgrounds from the experimental spectrum, and to compare the remaining K peak with the theoretical pulse height distribution at the energy considered. This was the approach adopted by the author in the K/β^+ measurements described in this thesis.

CHAPTER THREE

PULSE HEIGHT DISTRIBUTIONS FROM PROPORTIONAL COUNTERS

1. INTRODUCTION

As mentioned in Chapter Two, the major drawback of the proportional counter method for the measurement of electron capture decay rates lies in the fact that it is difficult to assess the areas under low energy peaks, since these are always superimposed on a continuous distribution of unwanted, but to some extent unavoidable, pulses. This measurement of area would be considerably simplified if the analytical form of the peak were known; the experiment described in this chapter was undertaken primarily to determine the shapes of pulse height distributions at low energies, although various other conclusions of some relevance to the operation of proportional counters were eventually drawn from it.

As is made clear in the review article of Curran and Wilson, (1965), most previous work in this field had been directed towards elucidating the relationship between energy resolution and energy, while experimental results on actual line shapes are scanty.

The spectrum shape due to the absorption of mono-energetic electrons in the filling gas of a proportional counter is determined basically by two factors; these are the variation in the number of primary ion pairs produced by each electron and the fluctuation

in the number of ion pairs formed in the avalanche which occurs as the primary electrons are drawn towards the anode wire by the high electric field across the counter. Additional variations are introduced by instrumental defects such as non-uniformity of the wire and the presence of gaseous impurities. It will be assumed that, if due attention is paid to the elimination of such defects, variations resulting from them can be ignored. If it is also assumed that there is no interaction between the ionisation produced by the incident electrons and that resulting from the avalanche, it can be shown that

$$\left(\frac{\sigma_P}{\bar{P}}\right)^2 = \left(\frac{\sigma_N}{\bar{N}}\right)^2 + \frac{1}{\bar{N}}\left(\frac{\sigma_A}{\bar{A}}\right)^2 \qquad (27)$$

where $\bar{N}$ is the mean number of initial ion pairs produced, $\bar{A}$ is the mean gas multiplication factor, $\bar{P}$ is the mean value of the total number of ion pairs, and σ_N, σ_A, σ_P are the standard deviations in these quantities. (The relative variance is defined as the square of the relative standard deviation).

Fano (1947) and Herring and Merzbacher (1957) have discussed the theory of the initial ionisation process, showing that the relative variance $(\sigma_N/\bar{N})^2$ should be $1/k\bar{N}$ where, according to Fano, the constant k should lie between the values 2 and 3; the latter authors estimate the value of k as 2.5. This variance is less than that obtained from Poisson's distribution, for which k has the value 1.

Various workers have investigated the statistical properties of the multiplication process, but the results, both experimental and theoretical, appear to be in conflict. For this reason, an independent investigation of the process was carried out by the author and Dr. K. W. D. Ledingham; this work, together with the previous results, is presented in the next chapter.

If $(\sigma_A/\bar{A})^2$ is taken as 0.681 (Curran, Cockroft and Angus, 1949), and k as 2.5, then equation (27) reduces to

$$\frac{\sigma_P}{\bar{P}} = \frac{1.04\,w^{1/2}}{E^{1/2}} \qquad (28)$$

where E is the radiation energy (in keV) and w ($=E/\bar{N}$) is the energy required to form an ion pair in the gas mixture used. Several workers have investigated the energy-dependence of $\sigma_P/\bar{P}$ and from a survey of experimental data, Mulvey and Campbell (1958) suggested that the relation

$$\sigma_P/\bar{P} = 0.15/E^{\frac{1}{2}} \qquad (29)$$

represented the best attainable energy resolution. Further evidence for a square root law is provided by the more recent experiments of Lukirskii, Ershov and Brytov (1963). Using an argon-methane filling (3:2) filling, these authors measured $\sigma_P/\bar{P}$ for pulse height spectra produced by x rays in the energy range between 114 eV and 8 keV; a square root relationship was obtained with a coefficient of 0.171, and this fell to 0.158 for a methane filling at very

low energies (75 to 282 eV). However, the measurements of Bisi and Zappa (1955) in the energy range 2 to 66 keV for argon-methane mixtures in the ratios 9:1 and 7:3 do not fit an $E^{\frac{1}{2}}$ law, obeying instead the relationship

$$\frac{\sigma_P}{\bar{P}} = \frac{0.138}{E^{0.395}} \qquad (30)$$

As regards actual line shapes, Bisi and Zappa have shown that these approximate to Gaussian distributions at energies above 2 keV, while various authors, including Lukirskii et al, comment on the increasing asymmetry of pulse height distributions as the energy is lowered. The main aim of the work described below was to obtain more quantitative information concerning the analytical form taken by such distributions.

2. EXPERIMENTAL TECHNIQUE

Various sources of electrons and x rays of low energy were considered, gaseous electron capture sources being excluded since the object of the experiment was to provide data for application to exactly this type of peak.

The first approach employed involved an electron gun arrangement in which electrons were accelerated by a variable voltage towards a copper foil anode (6 μm thick) which formed part of the wall of a proportional counter; a very small aperture in the foil allowed the electrons to enter the counter. Since the gun had to

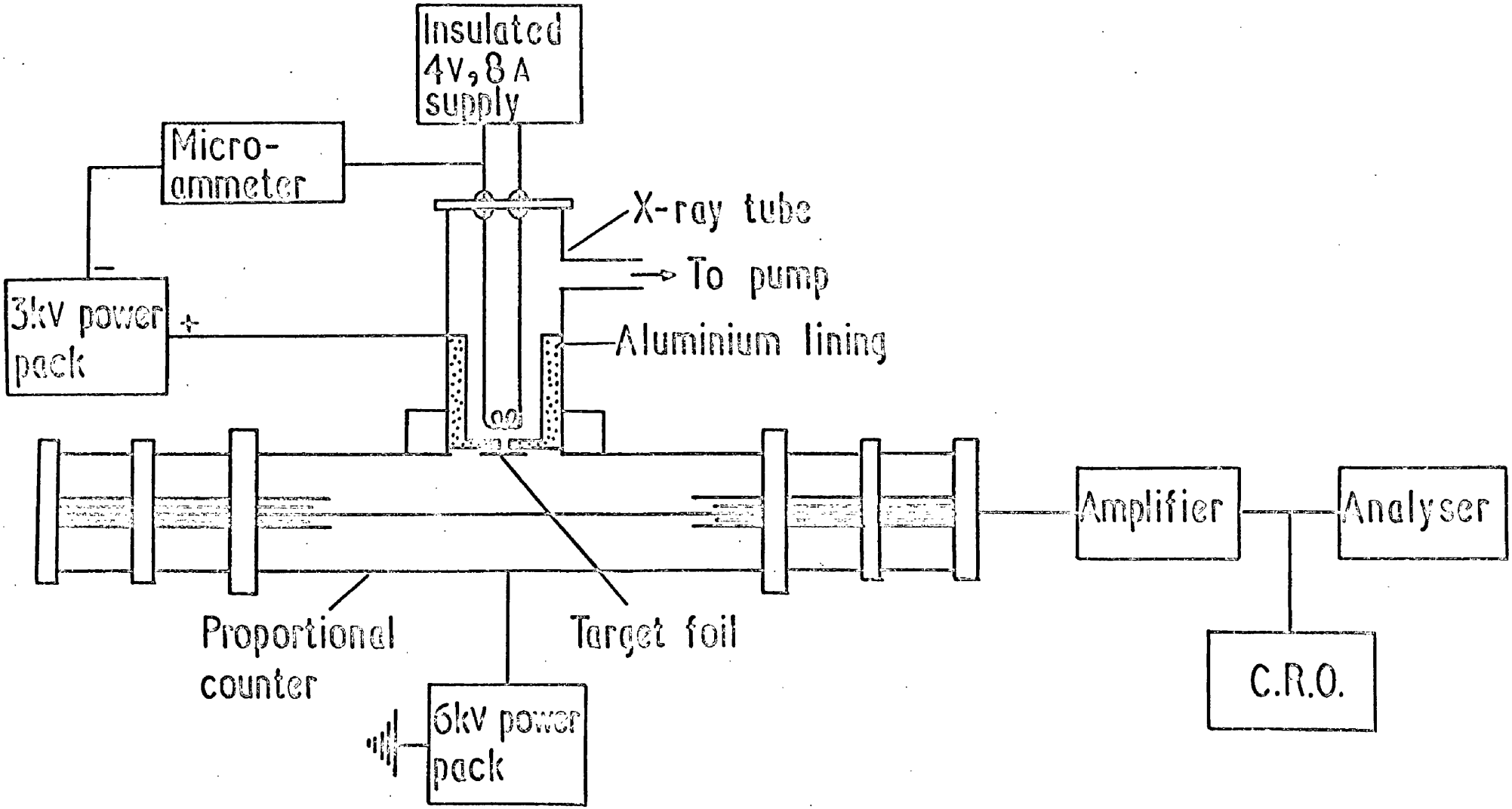


Fig. 6. Experimental arrangement for the study of x-ray peaks.

be pumped continuously to remove gas leaking through the aperture from the counter, it was necessary that the aperture be very small, and, to this end the spark erosion technique described by Stanier and Mee (1964) was adapted to drill holes of a few microns diameter in the copper foils.

The peaks obtained with this arrangement invariably had a mean pulse height corresponding to an energy slightly below that expected from the accelerating voltage and were accompanied by a continuous tail on the low energy side. This degradation appeared to depend on the shape of the hole, and although some excellent peaks were obtained, the results were not reproducible, and this approach was therefore abandoned.

It was decided instead to detect photoelectrons liberated in the counter gas by characteristic x rays from light elements. This approach has the obvious disadvantage that the characteristic spectrum from any element contains more than one x ray energy, but in the energy region of interest the error caused by this is negligible since both the energy difference between the K_α and K_β x rays and the fraction of K_β present are small. Distortion of the line shapes due to escape of x rays from the counter and to Compton scattering should also be negligible at these energies.

The experimental arrangement is shown in fig. 6. A small proportional counter having a sensitive length of 10 cm and a diameter of 2.54 cm was used. This was fitted with earthed guard tubes and

with field-correcting tubes in order to obtain a uniform field in the active volume of the counter; fhe field tubes were 1.3 cm in length and 0.63 cm in diameter. The counter was lined with aluminium sheet of thickness 0.015 cm and a stainless steel wire served as anode. According to the manufacturers, this wire had a uniform cross-section along its length, while at any one point the diameter varied between the limits of 0.0752 and 0.0762 mm round the wire. The effects on the results of this slight departure from circularity will be discussed later. The counter gas was a mixture of argon and propane at partial pressures of 50 cm and 20 cm Hg respectively, and before each filling this was purified by exposure to sodium-lead alloy for about 12 hours.

A miniature x-ray tube was constructed and fitted to the counter. Electrons from a heated cathode, accelerated by a voltage variable from 0 to 3 kV, were incident on a very thin foil of the target material attached by adhesive to the end of the tube. The end was curved to fit flush with the counter wall in order to avoid distortion of the electric field. Non-conducting foils were coated on the counter side with a layer of colloidal graphite to prevent field distortion due to electrostatic charging of the foil. A high vacuum was maintained in the tube by continuous pumping with an oil diffusion pump.

Electrons impinging on the foil excited characteristic K x rays, some of which were transmitted through the foil into the

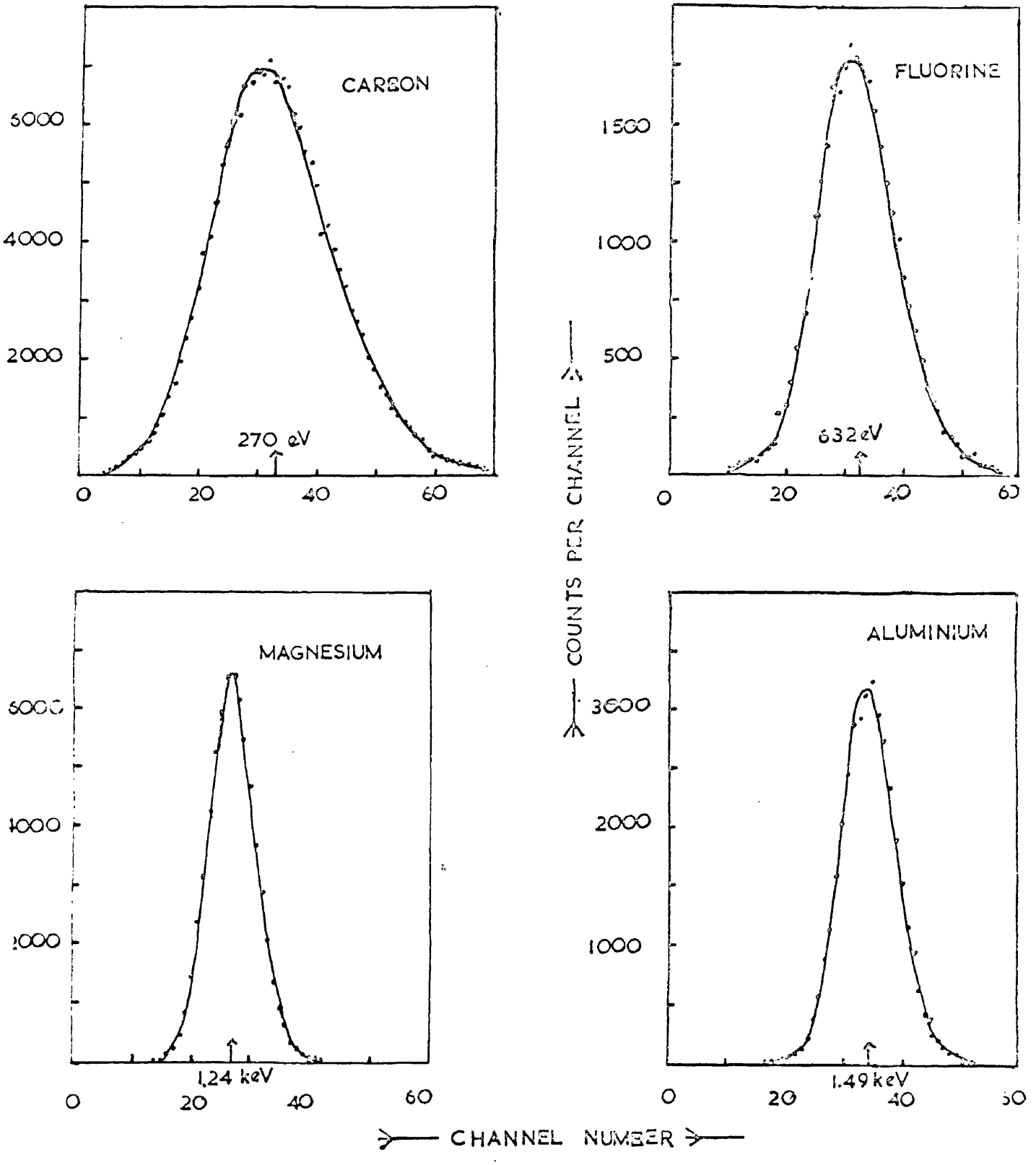


Fig. 7. Pulse height distributions from low energy x rays.

counting volume. The self-absorption in the foils was sufficiently small to allow counting rates of several thousand per minute to be attained. With foils of aluminium, magnesium, Teflon and Mylar, about 10 μm thick (30 μm in the case of Teflon), the peaks due to K x-ray peaks of aluminium, magnesium, fluorine and carbon were obtained. For each element, the x-ray tube was run at two voltages; the first spectrum was taken at a voltage slightly above the absorption edge and the second slightly below, the difference giving the K x-ray peak, natural background and bremsstrahlung being removed by the subtraction. When higher voltages were used, no change in the mean pulse height was observed, indicating that the peaks were entirely due to transmitted x rays and not to electrons passing through small holes in the foils. A set of typical peaks is displayed in fig. 7.

Energy calibration of the peaks was performed by adding a trace of argon 37 to the counter gas on fitting and noting the position of the K capture peak at 2.82 keV. Amplifier gains were measured accurately by feeding pulses from a mercury pulse generator into the counter capacity, using an aerial which could be attached to the counter wall.

3. RESULTS AND DISCUSSION

i) the values of the relative standard deviation $\sigma_p/\bar{p}$ for the four x-ray peaks are given in Table 4, along with the x-ray energy E

(in keV) taken from the tables of Bearden (1964) and the energy Em corresponding to the centroid of the peak. The centroid of the argon 37 K peak was taken as 2.82 keV, the contribution of 2.62 keV chlorine K_α radiation resulting from the absorption of 200 eV L x rays in the insensitive end regions of the counter being negligible. From the fourth column of the table it is seen that the results obey the relationship

$$\frac{\sigma_p}{\bar{P}} = \frac{c}{E^{1/2}} \qquad (31)$$

where the constant c has the value 0.169 ± 0.004.

The difference between this value and the value of 0.15 in equation (29) can possibly be explained partly in terms of the departure from roundness of the wire. From the variation in radius, the variation of the point where multiplication commences can be found approximately, and the resulting fractional variation in the distance over which multiplication occurs estimated. Such a calculation suggests a possible broadening of the peaks by several percent. Lukirskii et al also feel that imperfections in the wire contributed to the departure of their results from equation (29);since their counter had a small length-to-diameter ratio (2.5:1) and was not fitted with field tubes, imperfect field geometry may also have been a contributory factor in their case.

It would seem that there is now considerable evidence for

TABLE 4

RESULTS OF EXPERIMENT ON X-RAY PULSE HEIGHT DISTRIBUTIONS

Element	E(keV)	E_m(keV)	$\sigma_P/\bar{P}$	$E^{\frac{1}{2}}(\sigma_P/\bar{P})$	m	E/m (eV)
Carbon	0.277	0.275 ± 0.010	0.320 ± 0.010	0.168 ± 0.005	10.1 ± .3	27.4 ± 0.8
Fluorine	0.677	0.632 ± 0.020	0.210 ± 0.015	0.173 ± 0.012	22.7 ± 1.4	29.8 ± 1.8
Magnesium	1.259	1.246 ± 0.025	0.149 ± 0.004	0.167 ± 0.004	45.7 ± 1.1	27.5 ± 0.6
Aluminium	1.494	1.475 ± 0.025	0.136 ± 0.004	0.166 ± 0.005	53.7 ± 1.6	27.8 ± 0.8

Notes:

1. The quoted errors are estimated from statistical errors, uncertainties in calibrations and observed spreads in the results. The presence of systematic errors is not discounted, especially in the case of fluorine where considerable experimental difficulties were encountered.

2. The values of E for magnesium and aluminium are the weighted means of the K_α and K_β energies calculated by extrapolation of the β/α ratios tabulated by Wapstra, Nijgh and van Lieshout (1959).

the $E^{\frac{1}{2}}$ relationship, the coefficient c being dependent on instrumental factors. It was noted that the values of Em and $\sigma_P/\overline{P}$ for various carbon x-ray peaks were dependent to some extent on the gas purity. When freshly purified gas was used, the peaks were at their narrowest and the values of E and Em were in good agreement. Only measurements performed with such gas are included in Table 4. If the gas was not purified or if it was left in the counter for several days, the width of the peaks was noticeably greater, and Em was less than E by several percent (10% in the worst cases). These observations may go some way towards explaining the non-linearities observed at low energies by various workers, and they emphasise the need for adequate purification of the counter gas when a linear response is required.

ii) The Glasgow University K.D.F.9 computer was programmed to evaluate various statistical distributions for comparison with the experimental peaks. These were

a) Gaussian distributions;

b) curves of the form $P(x) = x^n \exp(-x)$;

and

c) a form of Poisson distribution

$$P(x) = m^x \exp(-m) / \Gamma(x+1), \qquad (32)$$

where the variable x is allowed to assume both integral and non-integral values, and m is the mean value of x. The gamma function $\Gamma(x)$ replaces the usual factorial $x!$ in order to deal with non-

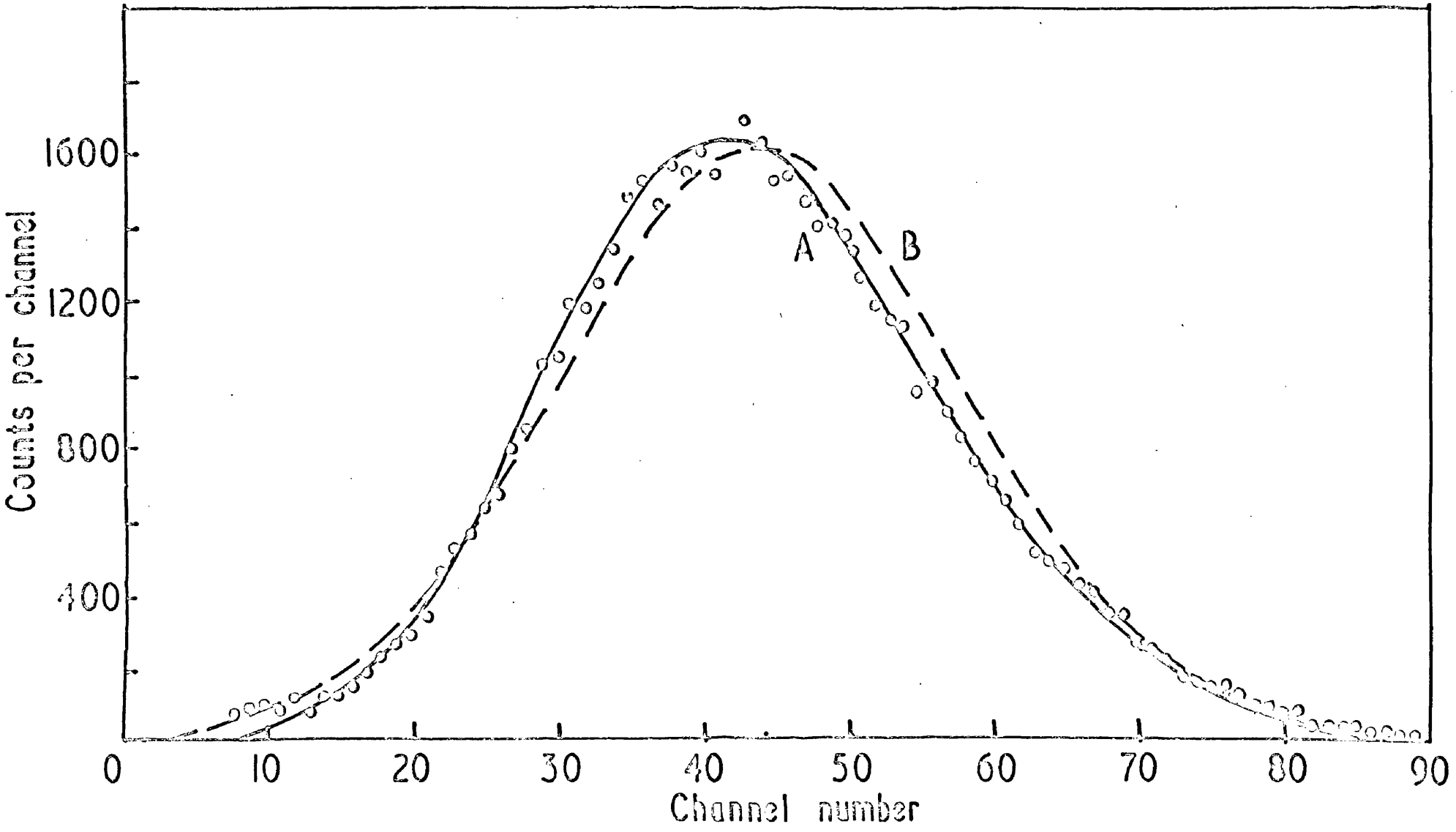


Fig. 8. Carbon x-ray peak (full circles) compared with A, Poisson and B, Gaussian distributions having the same centroid.

integral x-values.

The two lower energy peaks were evidently not Gaussian due to their distinct asymmetry, but the magnesium, aluminium and argon 37 K peaks agreed well with this distribution. The Poisson distribution, however, proved to be the best fit of the three to the experimental spectra, the value of the parameter m which gave the closest fit being evaluated independently for each peak. In fig. 8 an experimental carbon peak is compared with the Gaussian and Poisson distributions, the close fit with the latter being apparent.

It is of interest to link the correlation between increased width and decreased mean pulse height in impure gas with the properties of the Poisson distribution. Since the mean of this distribution is equal to m and the relative variance to $1/m$, then these two effects must occur together.

The values of m are presented in Table 4, and it would appear that within the limits of error m is proportional to the energy; as required for a Poisson distribution, $(\sigma_p/\bar{p})^2$ is equal to $1/m$. Obviously the proportionality constant (E/m) will vary from one counter to another, as does the related constant c in equation (31), depending on the instrumental factors involved. An accurate measurement of one peak at a definite energy should suffice to determine the constant required for any particular counter. It is fairly obvious that if the counter is to be used for the measurement of an internal gaseous source, then the calibration source should be

of the same type, while the same applies in the case of external sources.

The information obtained in this experiment is not only useful in the case of electron capture work. In the analysis of low energy beta spectra it is necessary to know the counter response in order to make resolution corrections which allow for distortion of the theoretical spectrum by the finite energy resolution of the counter. Another application is in the field of x-ray microanalysis where specimens are analysed for their content of different elements by detection of the characteristic x rays emitted due to electron bombardment; when more than one element of low atomic number is present, overlapping peaks must then be accurately separated.

CHAPTER FOUR

AN EXPERIMENTAL STUDY OF FLUCTUATIONS IN THE GAS MULTIPLICATION PROCESS

1. INTRODUCTION

The multiplication process can be investigated by employing the photo-electric effect to release single electrons of essentially zero energy from the inside walls of a proportional counter. The variance $(\sigma_A/\bar{A})^2$ may be calculated from the resulting pulse height spectrum P(x), where P(x)dx is proportional to the number of pulses whose height lies in the range (x,x+dx); P(x) obviously represents the probability distribution for the total number of electrons in the avalanche.

The first attempts to formulate a theoretical description of the multiplication process were those of Snyder (1947) and Frisch (1947); these authors made the assumption that the number, α, of ion pairs created by an electron per unit path length is constant, i.e. that the probability of ionisation is dependent only on the position of the electron and not upon its previous history. With this assumption, a simple calculation yields a pulse height distribution for single electrons of the form

$$P(x) = \exp(-x) \qquad (33)$$

with a relative variance $(\sigma_A/\bar{A})^2$ of unity.

Shortly after this work, experiments carried out by Curran, Cockroft and Angus (1949) using single photo-electrons released in a counter containing a 1:1 argon-methane mixture, suggested that the distribution could be fitted by the expression

$$P(x) = x^{\frac{1}{2}} \exp(-x) \; ; \qquad (34)$$

this distribution has a relative variance of 0.666 while the variance of the measured distribution was 0.696. However, a similar experiment performed by Birkhoff et al (1960) gave an exponential distribution in agreement with the theory proposed by Snyder and Frisch.

Byrne (1962) has recently reconsidered the problem theoretically, discarding the assumption of constant ionisation per unit path length and taking the ionisation probability to be a function of the number of electrons already present in the avalanche. A similar model has been proposed independently by Lansiart and Morucci (1962). For the same special form of α, viz.

$$\alpha(s,n) = (1 + \frac{k}{n})\alpha_o(s) \qquad (35)$$

where the term $(1 + \frac{k}{n})$ gives the dependence on avalanche size, both Byrne and Lansiart and Morucci obtain a distribution of the form

$$P(x) = x^{c_1} \exp(-x/c_2) \; , (c_1, c_2 \text{ constants}) \qquad (36)$$

in agreement with the result of Curran and his co-workers.

It would be of interest to consider the measurements required in the present study in the light of the earlier experiments.

To investigate a possible dependence of the single electron

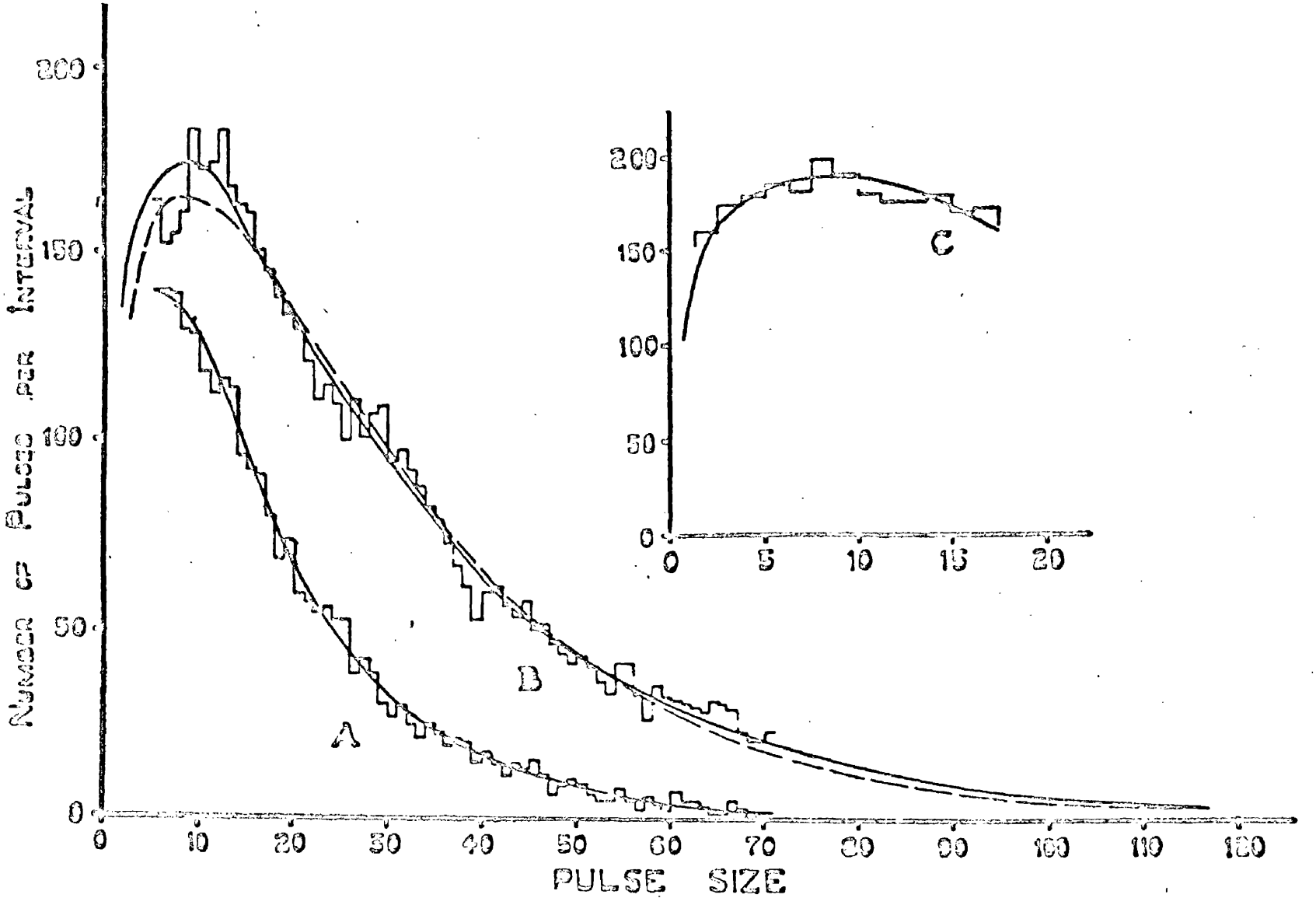


Fig. 9. Single electron spectra recorded by Curran et al. (Curve A measured at 3280 volts, curve B at 3550 volts; curve C shows part of curve B as analysed at higher amplifier gain).

spectrum shape on avalanche size, the spectrum should be recorded over a wide range of values of $\bar{A}$. This range is obviously limited at the upper end where the counter goes into the continuous discharge associated with Geiger operation. At the lower limit of the range, the very high electronic amplification required to examine the spectrum down to a sufficiently small pulse height results in a high noise level which in turn obscures the detail of the lower end.

It would also be useful to calibrate the counter with known low energy radiations at each gas gain in order to correlate its response with the observed single electron spectrum.

Curran's work was carried out at values of $\bar{A}$ up to greater than 10^6. The means were not then available to perform energy calibrations, but the counter appeared to be well-behaved over this range, and no variation in spectrum shape was reported. The distributions for counter voltages of 3280V (A) and 3550V (B) are shown in fig. 9; the latter curve corresponds to a gas gain of 1.5×10^5, and the inset (C) shows the lower end of B recorded at higher amplifier gain. Although the function $x^{\frac{1}{2}}e^{-x}$ is a good fit to the decreasing part of the spectrum, the author feels that the extrapolation to zero in the lower channels is perhaps rather difficult to justify. The efficiency of the electronic system for very small pulses was therefore noted carefully in the present work.

Birkhoff et al used a gas gain of about 4×10^4 and employed electron capture peaks at 5 and 2 keV to demonstrate the proportionality

of their counter; no details of amplifier gain or relative pulse heights for the single electron spectrum are given. It would appear possible that the amplification may not have been high enough to analyse the lower end of the spectrum in sufficient detail.

It is obvious that care has to be taken in attempting to fit the single electron spectrum with expressions such as e^{-ax} or $x^{c_1}e^{-(x/c_2)}$ due to the similarity of the upper portions of such curves. Precise knowledge of the centroid, standard deviation, and lower limit (set by the electronics) of the experimental curve would be of use in such attempts.

2. EXPERIMENTAL PROCEDURE

The procedure employed to detect the pulse height distribution was similar to that used by the previous workers. A small torch bulb was focussed on the aluminium cathode of the proportional counter described in Chapter Three; the photoelectrons liberated by the ultra-violet component of the light had insufficient energy to cause ionisation and therefore the recorded spectrum represented the pulse height distribution due to single electron avalanches alone. The high values of multiplication required were obtained by using an anode wire of diameter 0.025 mm, in preference to the original wire three times this thickness; the danger of spurious pulses resulting from high voltage sparking was thus reduced.

The experiment was carried out using two counter gas mixtures,

first the 3:1 argon-propane mixture used earlier and then a 9:1 mixture of argon and methane. The counter gas was flowed through the counter at a constant rate of 10 ccs per minute. The drift in gas gain was measured with a ^{71}Ge external x-ray source (9.2 keV) and found to be negligible over the two-hour periods in which single electron spectra were recorded.

The curve of counting rate versus voltage for single electrons is displayed in fig. 10 for the range 2200 to 3000 volts over which the spectrum shape was examined. It is well-behaved and shows no sudden increase at the top end. Curran et al took a similar curve as an indication that proportional response was maintained, but in the present work this was not assumed and a trace of argon 37 was added to the counter gas for calibration purposes. At each value of gas gain, a variable attenuator in the amplifier was employed to record the ^{37}A K and L electron capture peaks at 2.82 keV and about 250 eV respectively. The spectra were recorded at the highest amplifier gain compatible with an acceptable noise level.

The response of the pulse height analyser to very small pulses was checked and the first two channels found to be unreliable. No data were subsequently recorded from these channels.

Gas multiplication was measured by two different methods. First the counter was operated as an ionisation chamber for 5.3 MeV polonium alpha particles and the counting rate plateau plotted. With a working voltage of 300 V (in the centre of the plateau) and

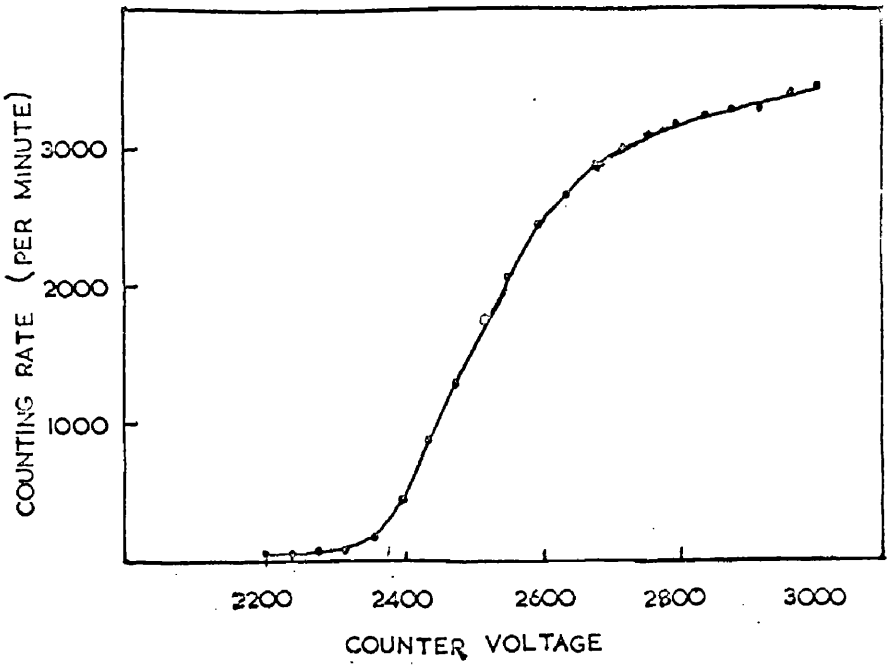


Fig. 10. Counting rate curve for single electrons.

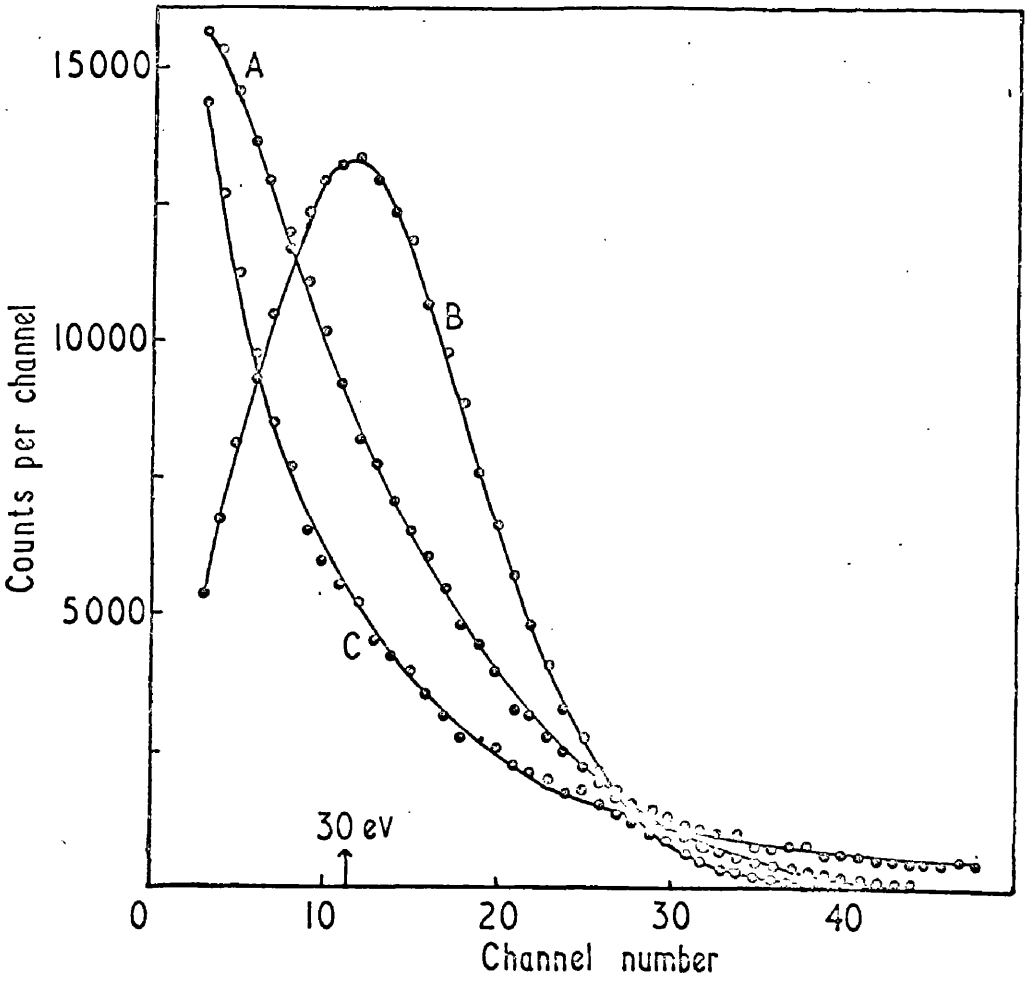


Fig. 11. Single electron spectra: A, argon-propane filling, $\bar{A} = 10^5$; B, argon-propane, $\bar{A} = 10^6$; C, argon-methane, $\bar{A} = 10^5$.

the same amplifier gain as for the single electron measurements, the position of the 5.3 MeV peak in the pulse height analyser spectrum was noted; the ratio of 5.3 MeV to 30 eV then gave the approximate number of ion pairs corresponding to this pulse height channel. From this figure the gas gain of the proportional counter could be deduced for any given output pulse height. The measurement was checked by feeding pulses of height v from a generator onto the counter wire through a capacity C (1.4 pf) small compared with the total counter and amplifier input capacity of about 30 pf. The charge (Cv) collected then gave the number of avalanche electrons corresponding to the output pulse from the system. The results obtained by the two methods were in good agreement.

3. RESULTS AND DISCUSSION

No significant variation in the shape of the single electron spectrum was observed as the gas gain was increased from about 10^4 to 10^5, but at higher gains a marked change in shape occurred as shown in fig.11, curves A and B. This change was accompanied by a progressive reduction in the observed ratio of the mean pulse height in the ^{37}A K peak to that in the L peak. The response of the counter was therefore no longer linear for these energies at gas gains greater than 10^5. These observations are in agreement with the "good behaviour criterion" reported by Hanna, Kirkwood and Pontecorvo (1949), which requires the product $\bar{A}E$ (where E is

the incident energy in eV) to be less than about 10^8; this figure is obtained by requiring the electron charge collected by the wire to be less than a few percent of the charge stored on the counter capacity. When this condition is not fulfilled, the counter field is distorted and effectively reduced by the space charge sheath of positive ions surrounding the wire. Obviously the single electron spectrum only violates this criterion at very high gas gains.

Since the gas gain used in the work on low energy line shapes was less than 10^5, (the expected ratio of the argon peaks being maintained) the avalanche spectrum to be associated with these distribution should be very similar to the form shown in fig. 11, curve A. The relative variance of this spectrum was found to be 0.651 from the average of several results. The mean energy of the ^{37}A L peak was estimated as 250 $\pm$ 15 eV by taking weighted contributions from L_I capture and from $L_{II,III}$ events resulting from the escape of K x rays following K capture (the revised value of 269 eV proposed by Fahlman et al, 1965, was used for the L_I energy). From this, the mean pulse height of the spectrum was found to correspond to an energy of 27.5 $\pm$ 1.7 eV. This may be taken as a measure of the energy required to form an ion pair, and, as such, is in general agreement with the results of other workers. The measured relative variance, although considerably smaller than that predicted by Snyder and Frisch, agrees well with that observed by Curran et al; however, the fall in the spectrum at low energies

reported by these workers was, as stated above, only observed at high gas gain where it would appear that space-charge effects were becoming significant. It was not found possible to fit the spectrum A with a simple function.

Taking the measured values for the energy per ion pair $E/\overline{N}$ and for $(\sigma_A/\overline{A})^2$ and taking Fano's constant k as 2.5, it can be shown from equation (27) that c should have the value 0.170 ± 0.005. This is in excellent agreement with the value of 0.169 ± 0.004 observed for the x-ray pulse height distributions and is a further indication that the spectrum A is the avalanche distribution appropriate to the latter.

Similar behaviour was observed with the argon-methane mixture, although the spectrum shapes were slightly different and the variance at a gas gain of 1.5×10^5 was slightly greater than in the case of argon-propane. It would appear that the spectrum is dependent on the gas mixture used, although it must be pointed out that the purities of the mixtures employed here may have been different.

The author concludes that the observed variation in spectrum shape with $\overline{A}$ demonstrates the dependence of ionisation probability on avalanche size, especially when space-charge is no longer negligible. The model proposed by Byrne and by Lansiart and Morucci represents a considerable advance on the earlier treatment of Snyder and Frisch, but would appear to require extension to explain the variation of shape, probably via a more complicated dependence

of the ionisation probability on the avalanche size than that given in equation (35).

Shortly after the publication of the above results, Gold and Bennett (1966) published an account of similar investigation for various argon-methane mixtures. Their results and conclusions are broadly similar to those of the author, although the measured values of relative variance at corresponding gas gains are rather larger than those obtained here, possibly because of the presence in their counter of nitrogen, introduced to afford an energy calibration using the 615 keV protons from the ^{14}N (n,p) ^{14}C reaction.

CHAPTER FIVE

THE K/β^+ RATIO IN THE DECAY OF CARBON 11

1. INTRODUCTION

Carbon 11 is an eminently suitable isotope for a measurement of K/β^+ at low Z, since it can be produced in the required quantities by the irradiation of an ordinary counter quenching gas such as methane or propane by bremsstrahlung x rays from an electron accelerator.

The cross-section for the reaction $^{12}C\ (\gamma,n)^{11}C$ exhibits a pronounced maximum, the well-known giant resonance, at a γ-ray energy of about 20 MeV. The integrated cross-section has been measured by Barber et al (1955) and by Cook et al (1966), the respective values being 80 MeV-mb (up to 250 MeV) and 77 MeV-mb (up to 65 MeV). The target nucleus ^{12}C forms 99% of natural carbon.

The half-life of ^{11}C is 20.70 $\pm$ 0.10 minutes (Kundu et al, 1953) and the decay

$$^{11}_{6}C \rightarrow {}^{11}_{5}B + e^+ + \nu$$

occurs between mirror nuclei, with zero spin change. From Table 3, the transition is a mixture of Fermi (63%) and Gamow-Teller components, being a simple allowed decay with a linear Fermi-Kurie plot.

The K/β^+ ratio of ^{11}C has been measured before, by Scobie and Lewis (1957), who determined a value of $(1.9 \pm 0.3) \times 10^{-3}$, in

agreement with a theoretical ratio of 2×10^{-3}. These workers used a multiwire counter, filling it with argon together with irradiated propane, counting for about two half-lives and then refilling. (In a separate experiment with a plastic counter similar to that described in Chapter Two, the K peak was not sufficiently well-defined to give a measurement of the K/β^+ ratio.) The statistics obtained by this method were poor, and the present author feels from his own experience that the uniform mixing of the counter gases necessary in such a measurement would not be attained under these conditions. Further, the theoretical ratio used by Scobie and Lewis was based on the early K-electron wave function given by Marshak (1942) and on a value of 970 keV for the maximum positron kinetic energy. This ratio is altered considerably when the more recent wave function given by Depommier et al (1960) is used, together with a positron end point derived from the precision experiments performed to determine the Q value of the reaction $^{11}B(p,n)^{11}C$. By a least squares adjustment of a large set of experimental data, Mattauch et al (1965) have derived a Q value which gives a value of 958.2 $\pm$ 1.4 keV for the positron end point.

It was felt therefore that the experiment could be repeated profitably, using some of the improved counter techniques discussed in Chapter Two, together with the detailed knowledge of counter response obtained by the work described in Chapters Three and Four.

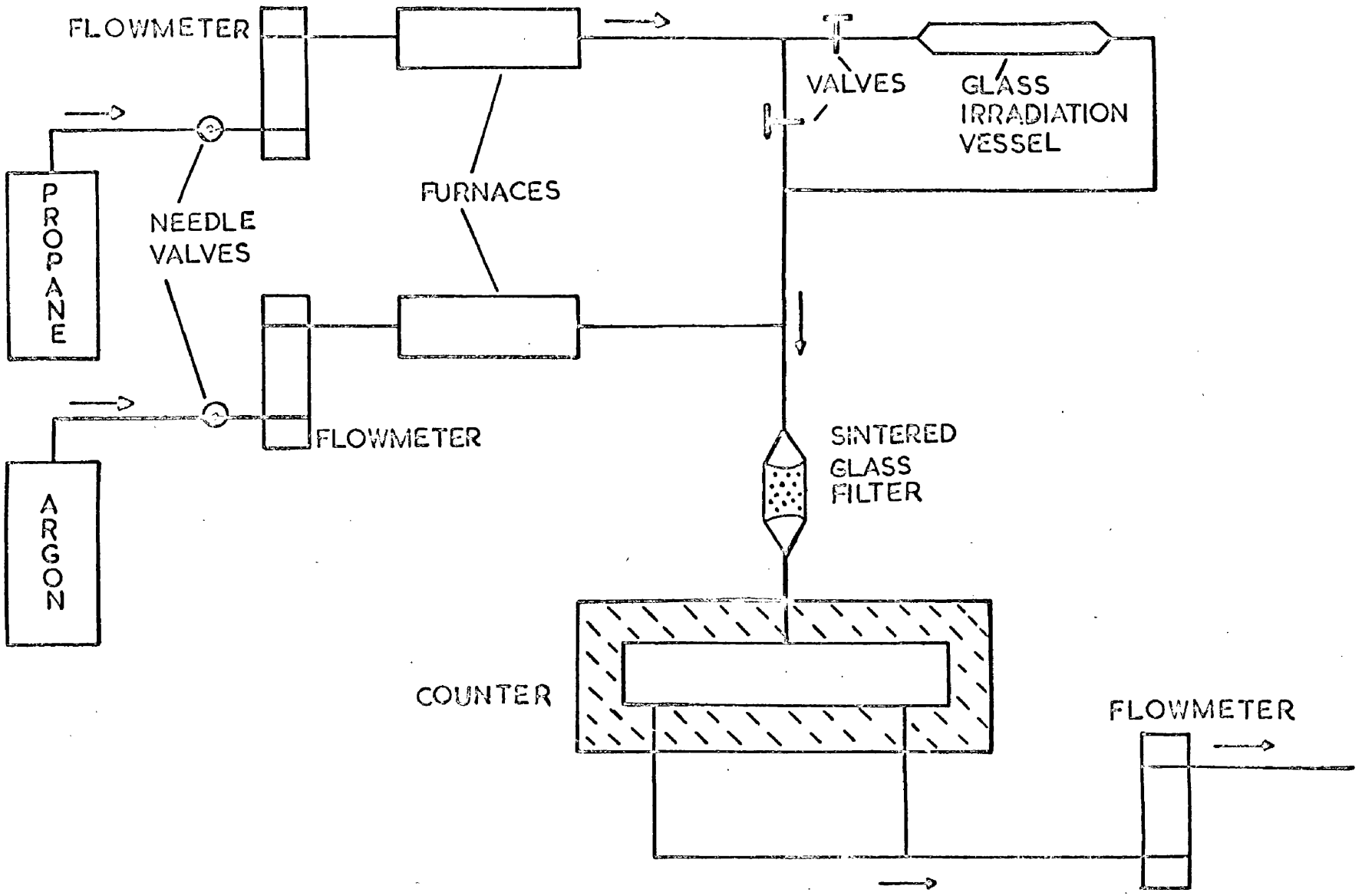


Fig. 12. Cas Flow System.

2. EXPERIMENTAL TECHNIQUE

The Gas Flow System

On account of its higher carbon content, propane was chosen in preference to the more usual counter gas methane, the main filling gas being argon. The flow system is displayed in fig. 12, the pressures required to drive the gases through the system being obtained by the use of commercial high-pressure cylinders. Constant pressure regulators on the cylinders supplied a rough control of the flow rates, while sensitive needle valves provided the fine control necessary to prevent long-term drifts. The flow rates from the cylinders were measured on calibrated Rotameter flowmeters whose readings could be monitored using microscopes.

To reduce the number of spurious pulses caused by the presence of impurities in the counter gases, these were passed through stainless steel furnaces containing copper turnings at temperatures of 550 and 450°C for the argon and propane respectively. This process was intended to remove oxygen, the principal contributor to negative ion formation.

A second needle valve in the propane line controlled the relative amounts of propane flowing to the irradiation vessel and to the bypass, the former flow rate being measured on a third Rotameter. The argon and propane lines then met at the input to the counter, where a sintered glass filter removed any dust particles which might enter the counter and cause sparking. The total flow rate was measured on a fourth Rotameter at the counter output.

The tubing used in the flow lines was copper of 1 mm bore,

the narrow bore ensuring that decay of ^{11}C in the 16 metre distance between the counter and the irradiation vessel was kept to a minimum.

Source Production

The irradiation vessel was a glass tube placed behind the tungsten target of the 300 MeV Glasgow electron synchrotron. To ensure maximum efficiency of irradiation, the vessel was aligned accurately along the x-ray beam, its diameter (2.5 cm) being equal to the diameter of the beam, and its length the maximum permitted by the geometry of the target region, namely 55 cm. The resultant volume was about 300 ccs and therefore at flow rates of a few ccs per minute the gas was irradiated for long enough (relative to the 21 minute ^{11}C half-life) for the necessary activity to be produced.

Calculations of activities produced by other photo-nuclear reactions on carbon, taking into account the flow rates and the relative volumes of the vessel and the tubing, indicated that only ^{11}C would reach the counter in any significant quantity; all other possible products were either stable or present to only a negligible degree due to half-life and cross-section considerations.

The gas mixture entered the counter by a small inlet halfway along its length, and left by outlets at each end. The flow rates were chosen so that the ^{11}C decayed considerably before leaving, this giving a high concentration of activity near the centre of

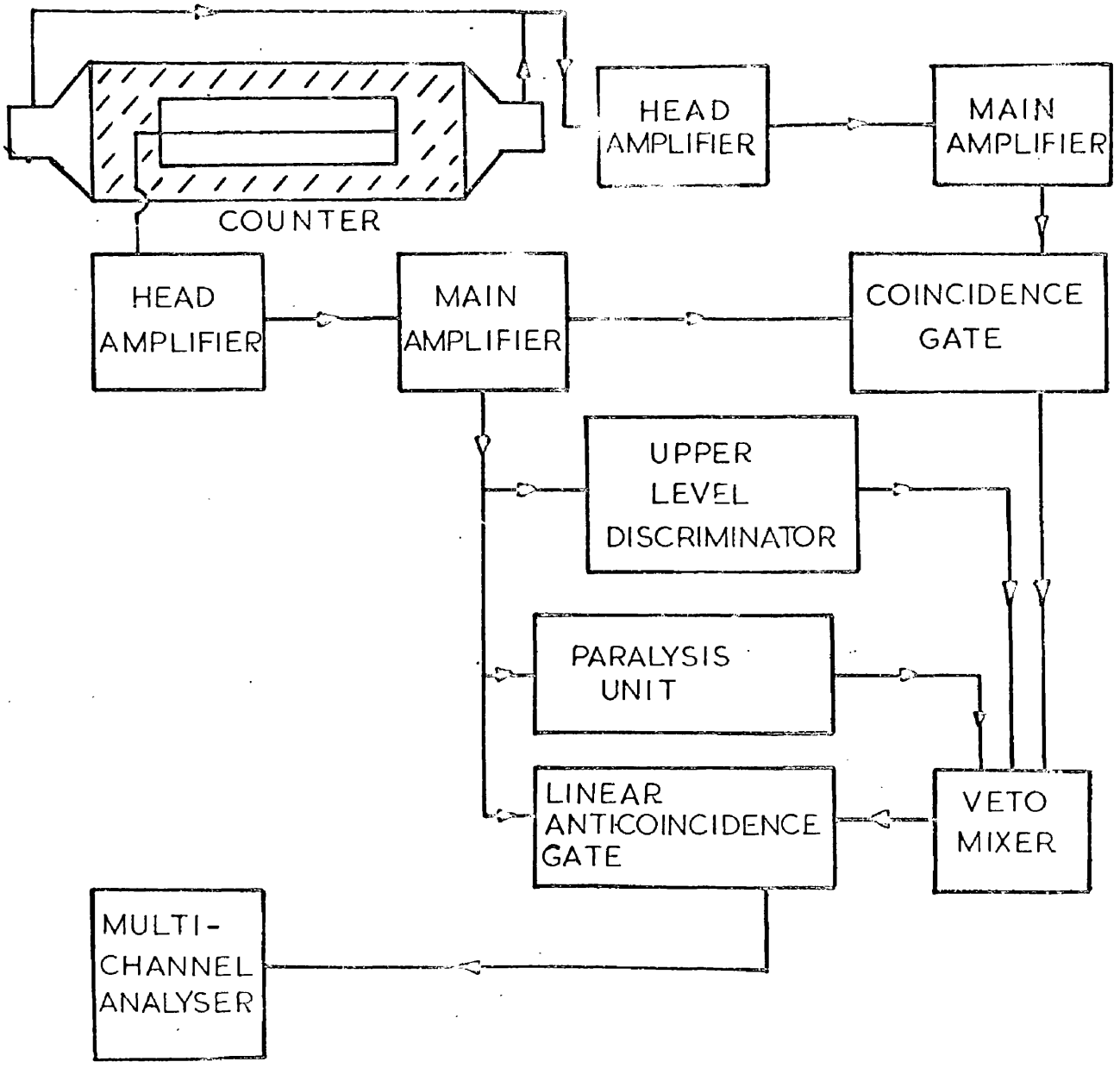


Fig. 13. Electronic arrangement used in ^{11}C experiment.

the counter and a low concentration at the ends. By this procedure end effects are considerably reduced, and a counter with no field correcting tubes may be used without introducing appreciable error.

With the synchrotron beam at its maximum intensity, activities of 30,000 counts per minute could be attained in the central counter.

The Counter

The plastic anticoincidence counter has been described in Chapter Two. For this experiment it was shielded by four inches of lead, which was covered by a layer of cadmium sheet and surrounded by three inches of concrete; these precautions ensured that neutrons generated by a Cockroft-Walton H.T. set operating in the vicinity did not interfere with the measurement.

Electronic Circuitry

The electronic arrangement employed in the final measurement is displayed diagramatically in fig. 13. The high voltage for the proportional counter was derived from an extremely stable Fluke 6 kV power supply, and the pulses from the counter were amplified by a Dynatron 1008 amplifier; a scaler at the amplifier output measured the total counting rate, mainly due to positrons from carbon 11.

Light pulses in the plastic scintillator were detected by

two five-inch condenser-coupled E.M.I. 6099F photomultipliers whose voltages were supplied by two potentiometers connected to the same 2 kV power pack. The relationship between gain and voltage differed slightly for the two tubes, and the gains were therefore matched by calibrating with ^{65}Zn 1.114 MeV gamma rays. The multiplier pulses were amplified by a NE5202 non-overloading amplifier, whose gain was chosen such that positrons of energies above about 40 keV would be detected. The resulting background counting rate was large (about 250,000 per minute), due to photomultiplier noise.

The two sets of pulses were then fed to the anticoincidence gate described on page 35 and the output pulses from the gate were displayed on a C.D.C.100 channel pulse height analyser, the resultant spectrum including the K-capture peak. This arrangement had been used in several previous experiments on electron capture performed by Drever et al, but in the present experiment the high counting rate in the veto circuit necessitated certain modifications. Tests of the system with a linear pulse generator revealed that when the photomultipliers were operating, about 5% of the signal pulses were degraded in size, giving rise to a flat spectrum of spurious pulses. This effect was due to random pulses from the multipliers closing the gate during the 7 μsecond rise of a signal pulse, as illustrated in fig. 5. Similar degradation during the 20 μsec decay of a pulse is dealt with by a section of the circuitry which detects coincidences between veto pulses and the back edges

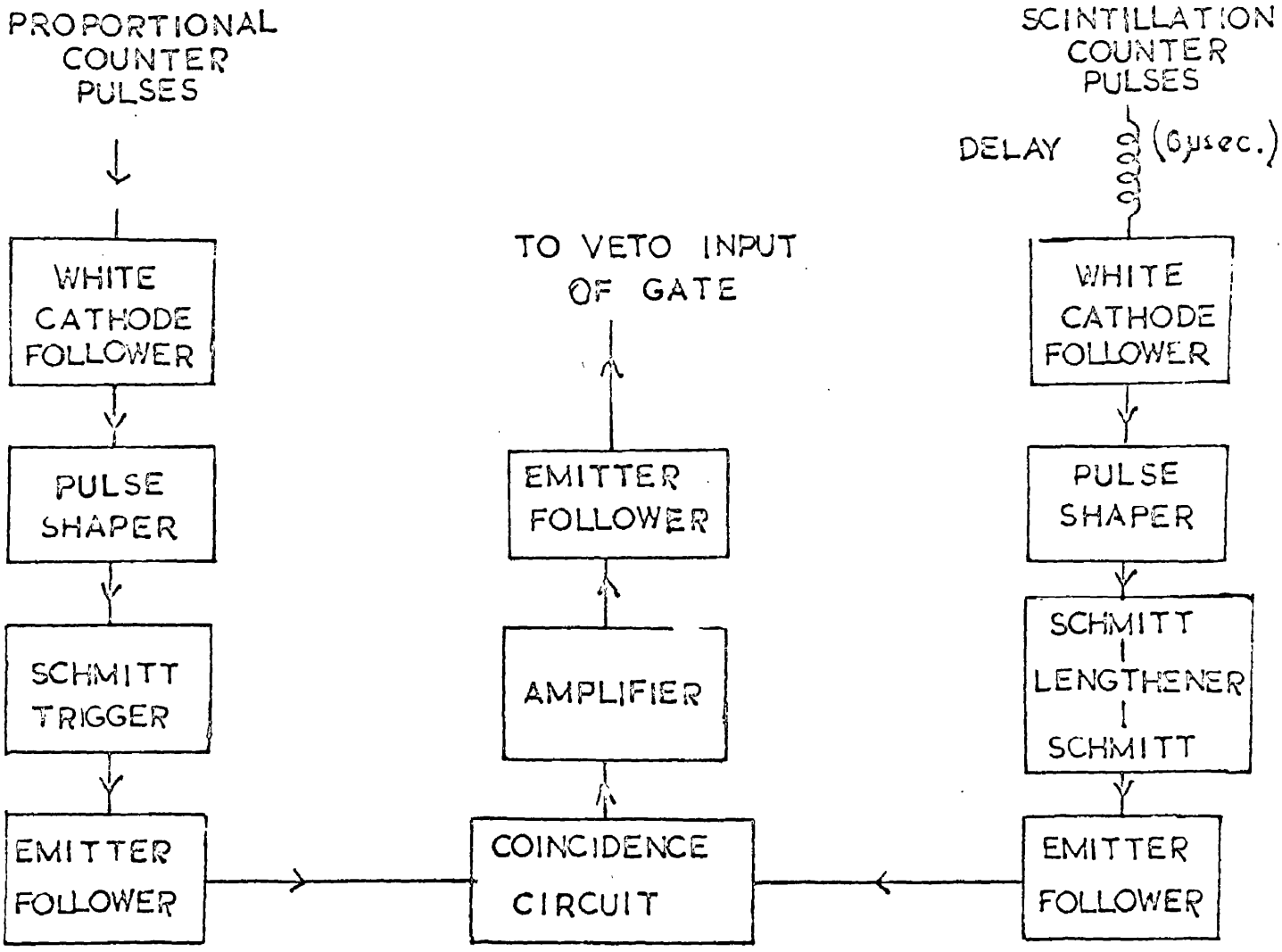


Fig. 14. Coincidence Unit.

of signal pulses; a further coincidence unit was therefore built to remove front edge chopping; this unit was placed between the NE5202 amplifier and the veto input of the gate, and only those veto pulses in coincidence with signal pulses were allowed to enter and close the gate.

The unit is shown in fig. 14. The two sets of pulses are limited, shaped by Schmitt trigger circuits and fed to a double diode gate. To ensure maximum efficiency, the fast scintillator pulses were delayed while the slow-rising proportional counter pulse reached the triggering level of the Schmitt; a delay of 6 μsec was found to give the maximum coincidence counting rate, the degradation being removed entirely. The resolving time of this unit made necessary a small correction to the registered open time of the gate; this is discussed later.

The linearity of the electronic system was tested with a flat spectrum generator. With the counter still connected, pulses were fed by capacitive coupling into the pre-amplifier and the resulting spectrum recorded on the C.D.C. analyser. By comparing this spectrum with the generator spectrum undistorted by the electronics, the relative widths of the analyser energy channels were evaluated.

3. EXPERIMENTAL PROCEDURE

With flow rates of 35 ccs per minute of argon and 12 ccs per minute of propane, and an x-ray beam of about 5×10^9 equivalent

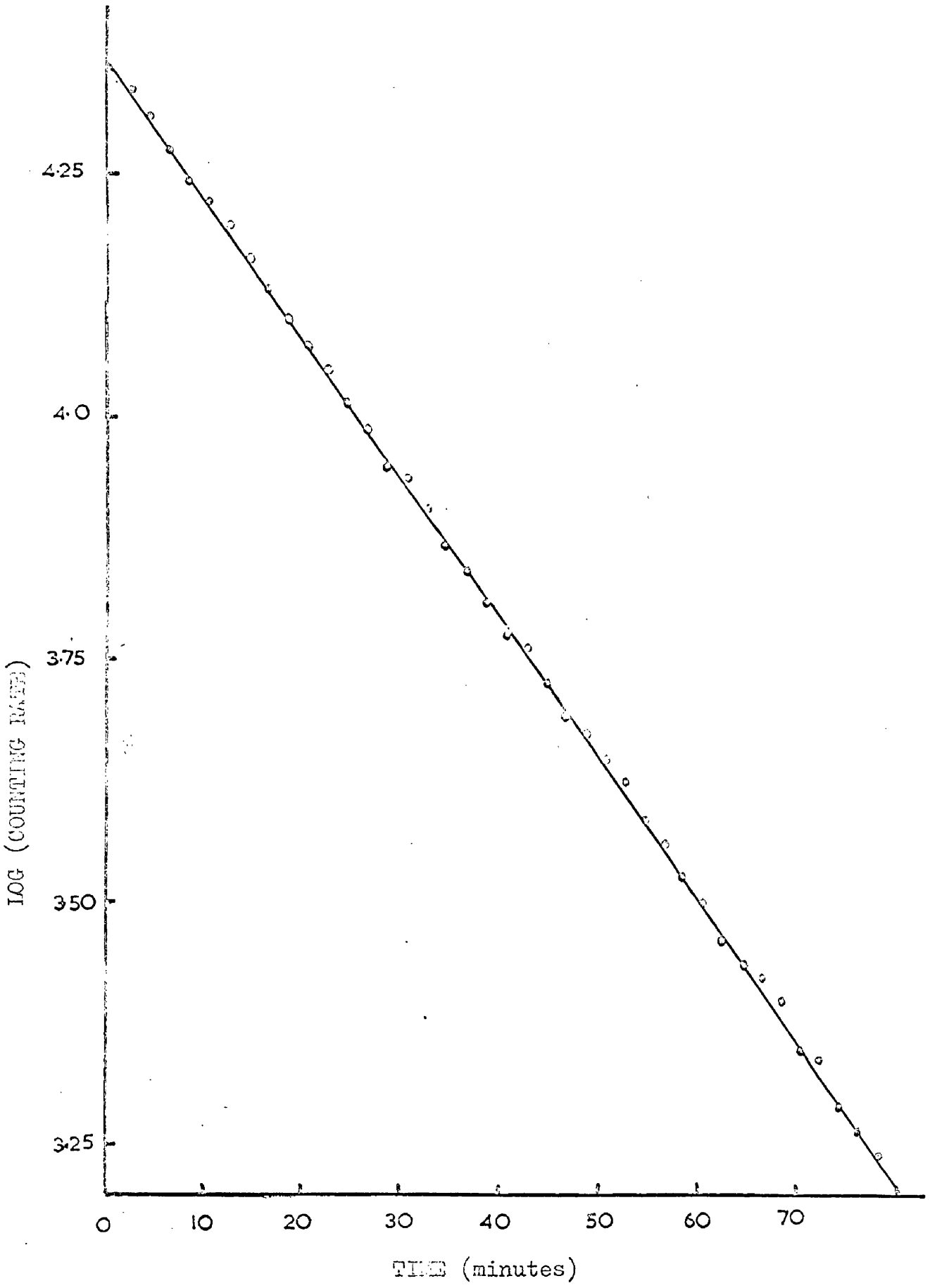


Fig. 15. Decay curve for ^{11}C.

quanta per minute, activity of the order of 15,000 counts per minute was obtained in the proportional counter. The counter was operated at about 3.2 kV. The gain was adjusted by using the 9.2 keV electron capture x rays from an external ^{71}Ge source; the ^{71}Ge peak was placed at channel 60 and the amplifier gain then increased by a factor of 25 to analyse the energy region below 1 keV.

Source Purity Check and Halflife measurement

The synchrotron beam was increased to its maximum, the counting rate increasing to 30,000 counts per minute. The counter was then sealed and the decay of the enclosed source was monitored on a scaler (before the gate) for one hour. The resulting decay curve, shown in fig. 15, was a straight line, there being no evidence for the presence of undesired activities. From a least squares fit analysis, the ^{11}C half-life was found to be 20.80 $\pm$ .06 minutes, in agreement with Kundu's measurement of 20.70 $\pm$ 0.10 minutes.

Measurement of the K/β^+ ratio

The K peak was recorded with the paralysis time set at various values from 300 μsec to 9 msec in an attempt to remove some of the pulses below the peak. A 2 msec paralysis was finally adopted; with lower paralysis times the number of these pulses increased, but the use of values greater than 2 msec led to no appreciable improvement in the definition of the low energy side of the peak.

With a positron counting rate of 15,000 per minute and a

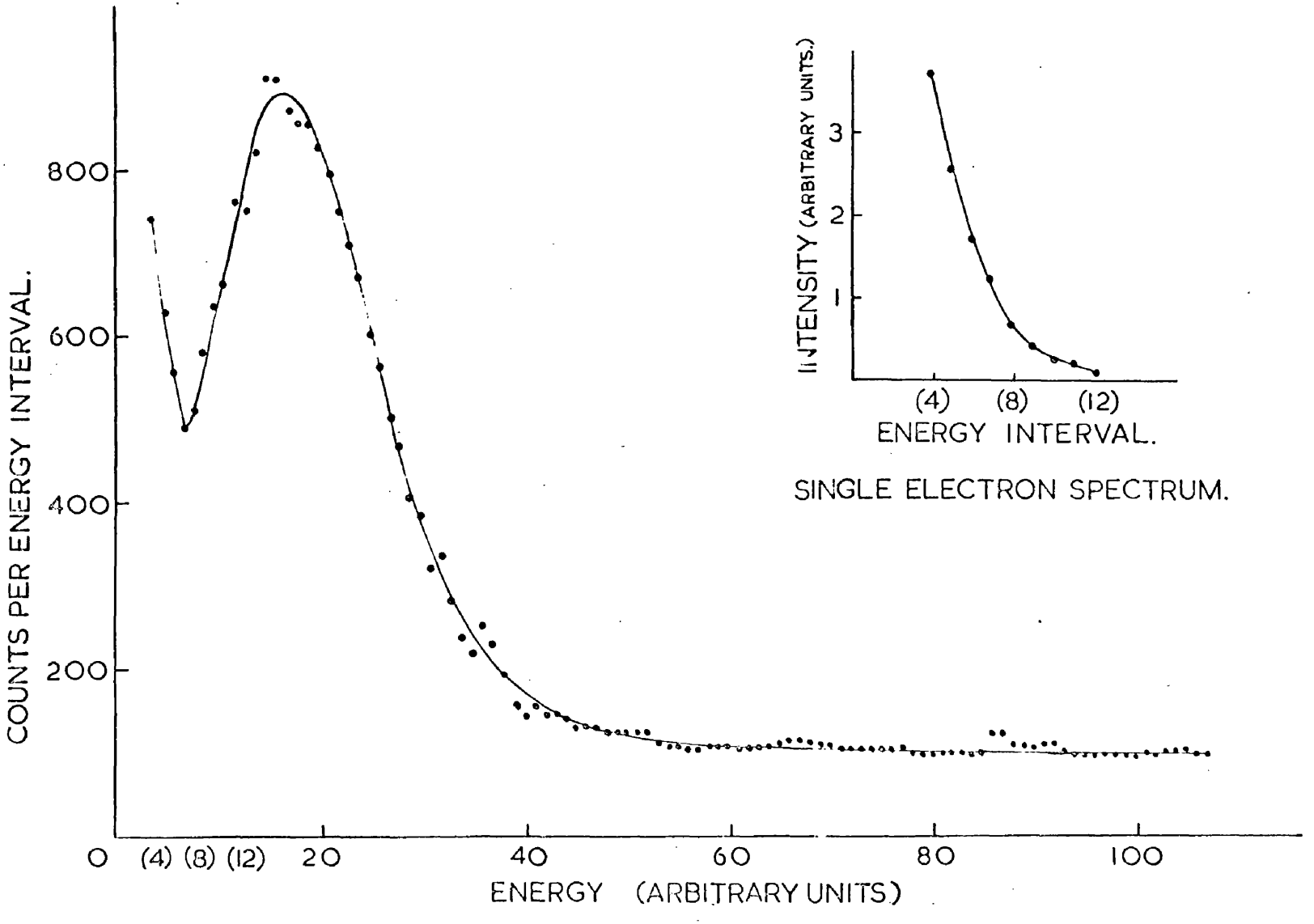


Fig. 16. Pulse-height spectrum from central counter in anticoincidence with ring counter; insert shows single electron spectrum (from Chapter Four) normalised to same energy scale.

paralysis time of 2 msec, the gate was closed for about 60% of the time. The counting rate in the ^{11}C K region was about 30 counts per minute, the contribution from natural background being about 1 count every three minutes.

Counting was performed in short runs of one to two hours duration. Between these runs the counter voltage could be adjusted to compensate for any small drifts in gain, using the ^{71}Ge calibration source. Since the ^{11}C peak in a proportional counter is intrinsically very wide (about 90% resolution), the peak was recorded in two series of runs at different amplifier gains. The spectra at these gains were taken over about twelve hours each, and corresponding background runs were carried out with the propane flow diverted out of the x-ray beam. The resulting K spectrum, corresponding to a total of the order of 10^7 pulses registered on the scaler before the gate is shown in fig. 16. (A very small correction was applied to this peak to correct for the slight non-linearity of the electronic system.)

4. ANALYSIS OF RESULTS

From the number of pulses recorded at the input to the gating circuit, along with the live time fraction (0.59) of the anticoincidence gate, the total number of pulses detected in the central counter during this live time was found to be 6,527,000.

The K capture intensity is considerably more difficult to assess; the K peak is superimposed on a spectrum of pulses which is linear above channel 60 but which rises steeply at low energy. The processes thought to contribute to this background spectrum have been outlined in Chapter Two. For the purpose of analysis, it was assumed that the low energy rise, interpreted as being due to single electron pulses arising from L capture in ^{11}C, heavy ions and very low energy positrons, could be represented by the single electron spectrum investigated in Chapter Four. The experimental spectrum was thus taken to contain three contributions of unknown intensity, namely the ^{11}C K peak, a single electron spectrum and a linear spectrum.

The single electron spectrum, normalised to the gain employed in the present experiment, is shown in the inset in fig. 16. The ^{11}C K peak was taken to be a Poisson distribution,

$$P(x) = m^x e^{-x} / \Gamma(1 + x) \qquad (37)$$

where the symbols have the same meanings as before. A straight line was fitted to the points above channel 65 by the method of least squares. The total background was then considered to consist to a first approximation of this linear spectrum, extrapolated to zero, with a single electron spectrum of unknown intensity superimposed upon it.

A computer programme was written in K.D.F.9 Algol to compare the K peak remaining after subtraction of this background with

a suitably normalised Poisson distribution, the goodness-of-fit being determined by the chi-squared criterion. This criterion requires that for the closest fit between theory and experiment, the sum of the squares of the differences between theory and experiment for each channel, weighted according to the standard deviation of that experimental point should be a minimum. The programme evaluated chi-squared for various combinations of the following parameters:-

1) the scale factor of the pulse height variable x;

2) the height of the Poisson peak;

3) the parameter m which determines both the centroid of the peak and its standard deviation ($\sqrt{m}$):

4) the intensity of the single electron spectrum.

Each parameter was varied over a range of values, the limits of which were defined as the points where the lowest value of chi-squared given by all possible combinations of the remaining parameters corresponded to a significance level of 5% (Fisher and Yates, 1957). The number of pulses in the K capture peak was evaluated in each case. To make some allowance for error in the assumed linear background under the peak, the whole procedure was carried out three times, first with the linear background given by the least squares fit to the high energy points, and then with backgrounds higher and lower than this by an amount equal to the standard error in the least squares fit.

It is found that, when one considers all combinations of the

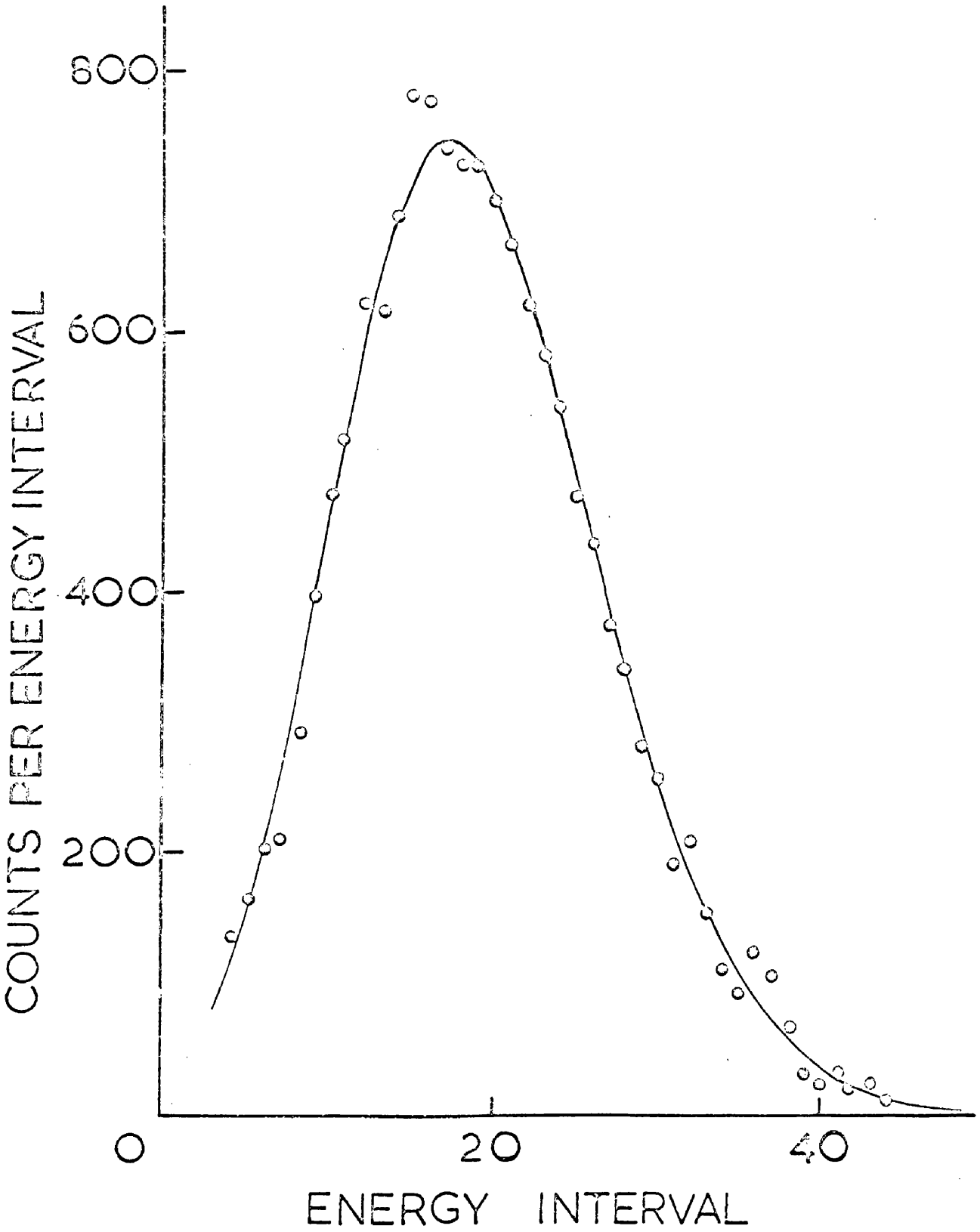


Fig. 17. Comparison between reduced experimental data (full circles) and the Poisson distribution which gives the minimum value of χ^2.

parameters which give a fit to the Poisson distribution with a significance level greater than 5%, then the corresponding value for the number of pulses in the K peak lies in the range between 12,850 and 14,210. This range is considered to give an indication of the probable error involved in the process of extracting the number of K capture pulses from the observed pulse height distribution.

The combination of parameters which gives the best fit to the Poisson distribution results in a value of 13,400 for the number of counts in the K peak, and this is regarded as the most probable value. The corresponding spectrum is shown in fig. 17.

The energy corresponding to the centroid of this peak was measured as 207 $\pm$ 15 eV, using the ^{71}Ge calibration and the values of m and scale factor given by the computer programme as being allowed. This is in agreement with the expected value of about 200 eV; the kinetic energy of the Auger electrons, calculated from the relevant binding energy data (Handbook of Chemistry and Physics, 1963) is about 170 eV, and to this energy should be added another 30 eV contributed by the Auger electron itself on reaching the avalanche region.

The number of positron decays was found by subtracting the number of counts in the K peak from the total number of pulses detected in the central counter during the same live time.

Two corrections were applied to the K/β^+ ratio calculated from these figures in order to compensate for counting losses:-

1) End correction:-

Since the counter is not fitted with field tubes, the insensitive end region is not well defined and the requisite correction has to be determined experimentally.

A K Auger electron born in the end region of the counter will not be detected, while less than half the positrons born here are registered. The ratio of the dead space to the total volume was measured previously by Ledingham et al as $(10 \pm .5)\%$, by investigating the loss of intensity in the ^{79}Kr L peak (1.79 keV) when the counter gas was shared with another counter of accurately known sensitive volume. In the case of a flow experiment on a short-lived source, however, the correction is reduced considerably owing to the decay of the activity before reaching the ends of the counter. In this case, the fall-off in activity along the counter was measured as 3.5 by placing a small proportional counter in series with the main counter at both inlet and outlet. The resulting correction to the K/β^+ ratio is $(2.4 \pm 2)\%$, the stated error being due mainly to uncertainty as to the amount of diffusion which takes place into the end regions.

2) Open time correction:-

The true dead time of the anticoincidence gate is in fact slightly larger than the value measured by the parallel gate timing unit; this is due to the coincidence unit. A noise pulse from the photomultipliers would be removed by the coincidence unit, and

although the presence of this pulse would prevent any signal pulse reaching the gate, the latter being effectively dead, the noise pulse would not pass to the timing unit to register a dead time. Counting rates and pulse lengths indicate that this further dead time fraction should be about 10%, being slightly dependent on pulse height. The correction was measured as a function of channel number by passing artificial pulses through the system while the ^{11}C decay was being recorded, and noting the fractional loss of pulses. The resulting correction to the K/β^+ ratio was $(9.3 \pm 0.6)\%$.

With the inclusion of these corrections. the errors being added orthogonally, a figure of $(2.30 \pm {}^{.14}_{.11}) \times 10^{-3}$ is obtained for the K/β^+ ratio.

The theoretical ratio was computed, there being no accurate tables of K/β^+ ratios available for very light isotopes. At the author's suggestion, a programme was first written by Mr. W. Leiper to compute the Fermi function F(Z,W) for 256 values of positron momentum p. This was based on the formulae given by Bhalla and Rose (1960) who published tables of F(Z,W) at limited values of W.

A second programme, written by the author, then employed an iterative Simpson's rule process to evaluate the total probability of positron emission. Previous workers in this field have

simply drawn the spectrum as a function of W on squared paper at about 30 values of W, and then used graphical integration to find the total intensity. The method devised here is more accurate and less tedious, the integration being performed several times, with the number of momentum steps dp being increased successively by a factor of two, until the integral takes on a constant value. It has the added advantage of being immediately applicable to other isotopes of interest with the requirement of no further effort. The end point kinetic energy was taken as 958.2 $\pm$ 1.4 keV.

The resulting value for the theoretical ratio is (2.18 $\pm$ 0.01) x 10^{-3}, where the error quoted is due only to the uncertainty in the end point.

The experimental and theoretical ratios for ^{11}C are therefore in reasonably good agreement. A value for the Fierz term could be derived from these results, but the author feels that this would be of doubtful validity at the present time due to the lack of information on overlap and exchange effects, and to the uncertainties in available calculations of electron wave functions.

To obtain a b-value of 0.2 as given by Langer, the theoretical ratio would have to decrease to 1.7 x 10^{-3}. There is therefore no evidence at present to corroborate Langer's observations, but the need for adequate theoretical information is again underlined.

Ledingham et al found agreement between theory and experiment in the case of ^{13}N, by flowing nitrogen, irradiated by 14 MeV neutrons

through the plastic counter. This experiment had various disadvantages, including degradation of pulses by front-edge chopping, and worsening of resolution due to the presence of nitrogen in the counter. The background spectrum was removed by recording the low energy portion of the spectrum from a high energy gamma source and subtracting this, suitably normalised, from the experimental spectrum; although the spectra produced in this way appeared acceptable, the author feels that the procedure cannot be entirely justified and that more reliance may be placed on the ^{11}C result obtained by the analysis described here.

CHAPTER SIX

A NEW TECHNIQUE FOR THE MEASUREMENT OF K/β^+ RATIOS IN VERY SHORT-LIVED SOURCES:

DETECTION OF K CAPTURE IN NEON 19

1. PRELIMINARY

Attention was drawn in Chapter One to the dearth of measurements of K/β^+ ratios in the atomic number range ($14<Z<20$) where the effects discussed by Bahcall should be most pronounced. Two of the isotopes in this range, ^{34}Cl and ^{35}A, (of further interest on account of their large Fermi fractions) might be studied with proportional counters. The main drawback in such an investigation is the short half-lives of these isotopes, and this led Dr. R. W. P. Drever to suggest the "direct irradiation" technique which was outlined in Chapter Two. If successful, this technique would allow one to dispense with the complicated anticoincidence systems described earlier, and to use a conventional, static counter.

The work described in this chapter was undertaken in order to investigate the possibilities afforded by this new method, with a view to applying it at a later date when the Glasgow linear accelerator should become operational.

Various data relevant to the production of ^{34}Cl and ^{35}A (and also ^{11}C and ^{19}Ne) by placing a proportional counter containing a

TABLE 5

DATA RELEVANT TO YIELDS OF ^{34}Cl, ^{35}A, ^{19}Ne and ^{11}C

PRODUCED BY (γ,n) REACTIONS

Isotope	$T_{\frac{1}{2}}$	Gas to be irradiated	A (%)	B (%)	Integrated cross-section (MeV.mb)
^{35}A	1.8 sec	Argon	0.34	90	--
^{34}Cl	1.6 sec	CH_3Cl	75.5	1	140 †
^{19}Ne	17.5 sec	Ne	91	90	115 *
^{11}C	21 min	C_3H_8	99	30	80

A = %age abundance of target isotope in natural element;

B = %age of gas which can be tolerated in counter filling.

*Gorbunov et al (1962); †Ferrero et al (1959).

suitable target gas in a bremsstrahlung x-ray beam are given in Table Five. It will be seen from the table that each of ^{34}Cl and ^{35}A has a particular disadvantage; the percentage abundance of the target isotope ^{36}A in natural argon is only 0.34%, while in the case of ^{34}Cl an admixture of more than one or two percent of a suitable gas, e.g. methyl chloride, in a counter ruins its operation. Finally, the beam-handling facilities of the synchrotron dictate that a counter placed along the beam cannot be closer than twelve feet to the x-ray target. The collimation required to produce a narrow enough beam at this distance then reduces the intensity by a large factor relative to that available in the ^{11}C experiment, where the irradiation vessel was placed beside the target and exposed to the full, uncollimated beam.

Rough calculations, based on the above data and on the ^{11}C activities obtained previously, indicate that the maximum activities of ^{34}Cl and ^{35}A which could be produced with the synchrotron beam, would be of the order of a few counts per minute. The projected linear accelerator intensity of 6×10^{14} equivalent quanta per second, would, however, be adequate for the purpose.

It was decided therefore to carry out a preliminary experiment with the synchrotron on the isotope ^{19}Ne in order to gain some experience in the new technique before designing experiments on ^{35}A and ^{34}Cl for the linear accelerator. Calculations indicated that useful quantities of ^{19}Ne could be produced in the synchrotron beam, and a preliminary experiment verified this, although the counting

rates were, as expected, considerably smaller than those obtained in the ^{11}C experiment. One drawback which had to be accepted was the 17.5 sec halflife, an order of magnitude greater than those of ^{35}A and ^{34}Cl, which might allow some diffusion of the activated gas to the counter walls during the decay, with a consequent increase in wall effect.

TABLE 6

INTEGRATED CROSS-SECTIONS FOR PHOTO-REACTIONS IN NEON 20

Reaction	$\int_0^{170\,MeV} \sigma\,dE$ (MeV.mb)	Product	Decay of product	$T_{\frac{1}{2}}$
(γ,p)	165	^{19}F	stable	--
(γ,n)	115	^{19}Ne	β^+,ec	17.5 sec
(γ,pn)	66	^{18}F	β^+,ec	110 min
(γ,α)	6	^{16}O	stable	--
$(\gamma,\alpha n)$	38	^{15}O	β^+,ec	2 min

The cross-sections for various photo-reactions in ^{20}Ne (integrated from zero to 170 MeV) have been measured in a cloud chamber experiment by Gorbunov et al (1962); the results, together with the product isotopes are summarised in Table Six. ^{11}C will be produced, of course, in the counter quenching gas. The relative

quantities of impurity isotopes produced are discussed later.

The decay

$$^{19}_{10}Ne \rightarrow ^{19}_{9}F + e^{+} + \nu$$

is a mirror transition with zero spin change. Interest in the ft values of such "super-allowed" transitions has led to several measurements of the half-life, with results ranging from 16.7 to 20.3 seconds. Precise determinations, by Earwaker et al (1962) Hermannsfeld et al (1959) and Penning and Schmidt (1957), suggest a value of 17.5 ± .1 sec. The transition is a mixture of Gamow-Teller (69%) and Fermi (31%) components. Mattauch et al give a value of 2.217 ± 0.005 MeV for the maximum positron kinetic energy, based on various (p,n) reaction measurements.

Electron capture has not previously been observed in ^{19}Ne and no attempt has been made to measure the K/β^{+} ratio. The energy of the Auger electrons emitted following K capture is about 657 eV, calculated from the fluorine K_{α} x-ray energy of 677 eV given by Bearden and the L_{II} binding energy of about 20 eV (H.C.P. 1963). The K peak should therefore correspond to an energy of around 680 eV in a proportional counter.

If the value of 1.47×10^{-3} is taken for g_k^2 (calculated by Depommier, 1966, using his 1960 work), the K/β^{+} ratio obtained from the programmes described in Chapter Five is $(9.5 \pm .1) \times 10^{-4}$. The error quoted is due solely to the spread in the positron end-point; as mentioned earlier, the wave functions in this region of

atomic number may not be accurate.

2. PRELIMINARY EXPERIMENTS ON GAS PURITY AND COUNTER RESOLUTION

In view of the difficulties encountered previously with rising spectra at low energies, an attempt was made to find an efficient method of purification of the gas contained in a static counter. For this work a brass counter, of length 45 cm and diameter 15 cm was employed; this was fitted with both earth tubes and field-correcting tubes. Teflon was chosen as the insulator for the end assemblies since it does not outgas appreciably, and stainless steel wire, diameter 0.07 mm, chosen for its uniform cross-section (variation in diameter less than 0.5%) was used for the anode.

Since it would be necessary in the final experiment to keep production of ^{11}C to a minimum, methane was chosen, rather than propane, as quenching gas. It was then necessary to determine the minimum permissible methane concentration. A 9.2 keV ^{71}Ge x-ray source was placed outside the central window and the energy resolution, together with the relative intensity of pulses in the energy region below 1 keV, was noted for various Ne/CH_4 ratios, the total pressure being 1 atmosphere. Before each filling the methane was purified by exposure to sodium-lead alloy in a glass vessel at 100°C for 6 hours. The counter response, in terms of resolution and intensity of small pulses, was observed to worsen at methane concentrations below 10%, and a 12% concentration was finally adopted.

As expected, increase of the paralysis time after each pulse (using the circuitry described earlier) resulted in decreased intensity of small pulses. If these pulses are due to negative ions, then the drift velocities indicate that a 15 msec paralysis would be required to eliminate all of them; since the maximum feasible paralysis is only a few msec, it was clear that any further improvement must lie in more efficient removal of impurities such as oxygen and water vapour from the counter gas.

Two circulatory purifiers were therefore built to provide alternative methods of continuous purification of the gas contained in the counter. Both relied on the heat generated by the purifying agent in a vertical pyrex tube attached to the counter to drive the gas round the system by convection. The first device contained a thick copper spiral heated to 600°C by a high electric current; after some hours of operation, the black copper oxide could be removed by reduction in a hydrogen atmosphere. No improvement in counter response was observed.

The second system contained four gauze trays of sodium-lead alloy, one above the other, at 100°C. This was exceedingly effective; over 100 hours of operation, the ^{71}Ge resolution improved steadily from over 20% to just under 17%, while the intensity of small pulses (below 1.4 keV) produced by an external ^{65}Zn γ-ray source attained a constant minimum value after 24 hours. This system was adopted for use in the present (and in future) experiments.

3. THE COUNTING SYSTEM FOR NEON 19

The counter employed in the measurement of K/β^+ in ^{19}Ne consisted of a 4.7 metre brass tube, of internal diameter 12.5 cm, flanged to fit the end assemblies used on the test counter and wired with the same .07 mm stainless steel. In order that the x-ray beam might pass as close to the counter axis as possible, the earth tube assemblies were made as narrow as possible, and the field tube voltage supplied through a second teflon insulator half-way along a radius.

The length of the counter was the maximum compatible with the size of the experimental area, the counter being in fact the longest reported. The purpose of this length was simply to expose the maximum amount of neon to the x-ray beam, in order to maximise the ratio "$(\text{source})^2$/background" (Moljk et al, 1957); this could not be done by resorting to high pressure, since thin windows were necessary to admit the beam without any attenuation and without activation of the window material.

A survey of available window materials suggested that nickel foil, thickness 50 μm, would be strong enough to permit safe evacuation of the counter, and would not give rise to any short-lived activity. Mylar was rejected on account of its porosity, and copper, aluminium and stainless steel due to the production of short-lived positron emitters. The windows were placed as close to the centres of the end plates as possible, being attached to the latter by Araldite.

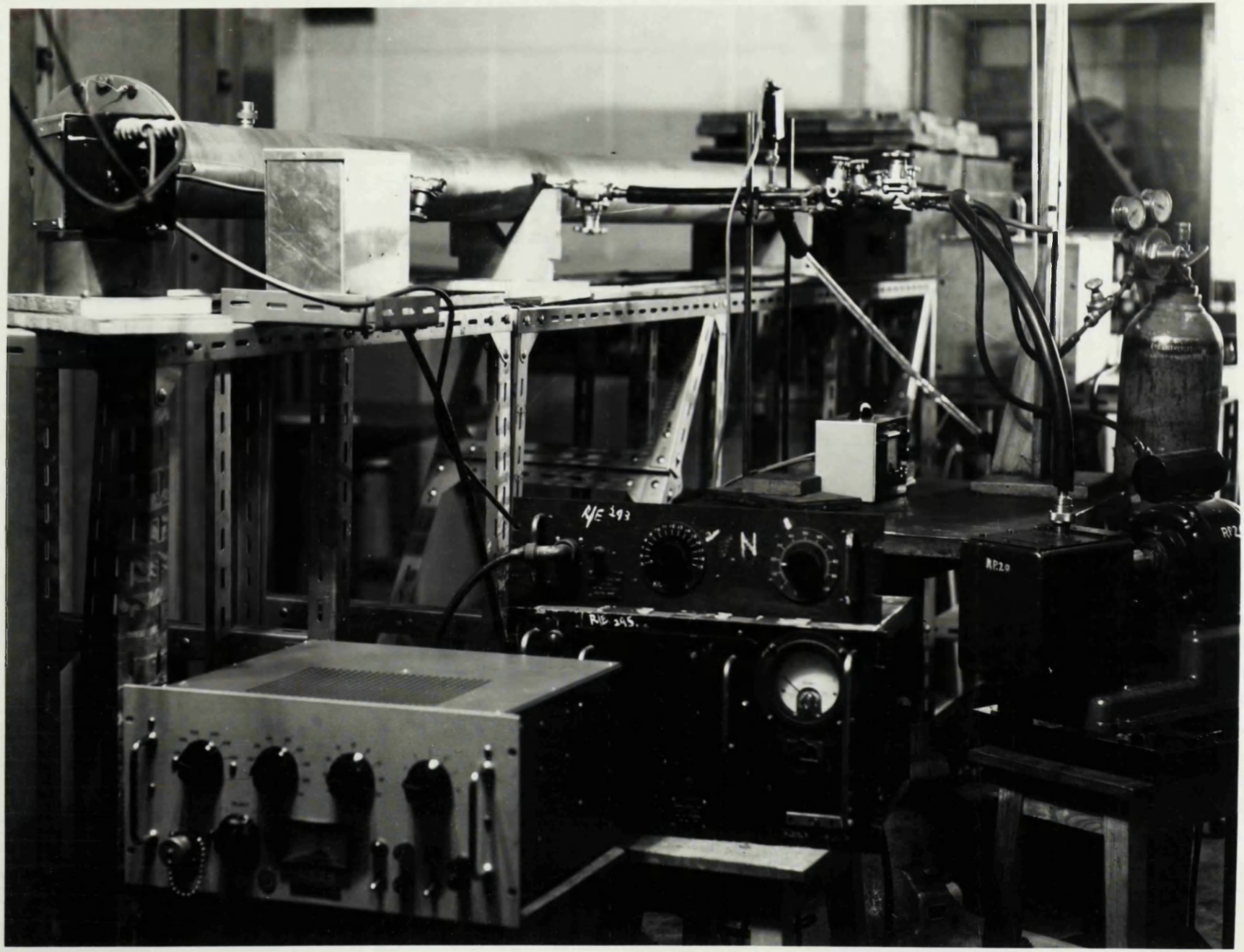


Fig. 18. Experimental arrangement employed for investigation of K capture in ^{19}Ne.

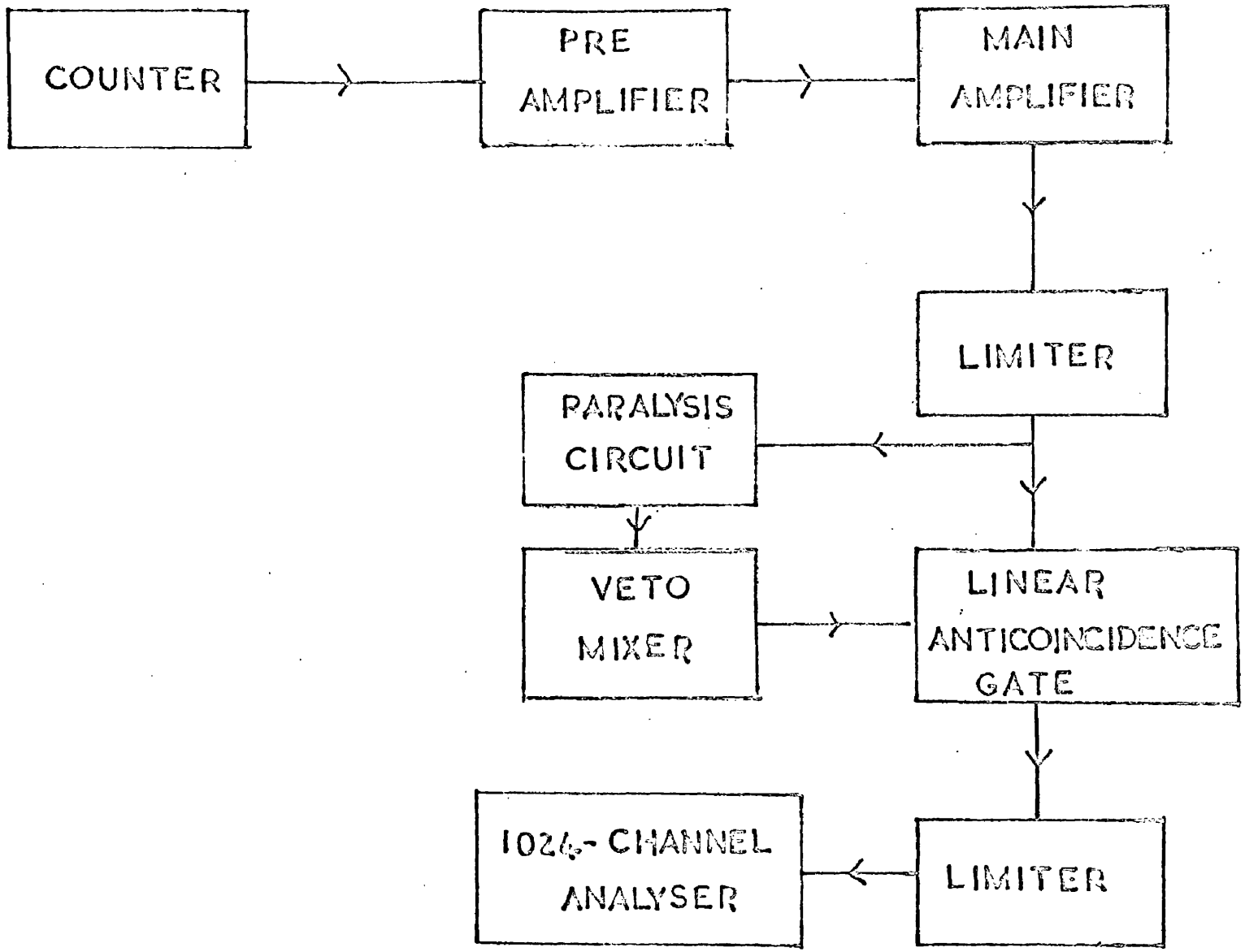


Fig. 19. Electronic circuitry employed in conjunction with directly irradiated counter.

At the entrance to the counter the x-ray beam diameter was 1.6 cm and at the exit, 2.5 cm. The counter and window dimensions were such that some of the activated cone of gas might diffuse to the nearest part of the wall during the measurement of the ^{19}Ne decay. However, the increase in counter diameter required to counteract this effect would have increased the cosmic ray background enough to cancel any improvement in terms of accuracy of separation of the ^{19}Ne K peak from the backgrounds of cosmic ray and wall effect pulses. The advantages to be gained from the high intensity linac beam in terms of a much shorter and broader counter are obvious.

The counter was aligned in the synchrotron beam with the aid of x-ray photographs placed over the windows, the alignment being adjusted to place the beam as close to the axis as possible. Four inches of lead shielding was used to reduce cosmic ray background. Fig. 18 is a photograph of the experimental arrangement in the beam room, with part of the shielding removed.

A counter of this length is extremely prone to electromagnetic interference and considerable care was taken to ensure that spurious pulses were not induced by the pulsing magnetic field of the synchrotron.

The high voltage (approximately 2100V) was supplied by a Fluke 6 kV power pack, and the pulses were amplified by a 1008 amplifier. The electronic system is depicted in fig.19. The paralysis facility of the anticoincidence gate was employed to inject a 2.2 msec dead

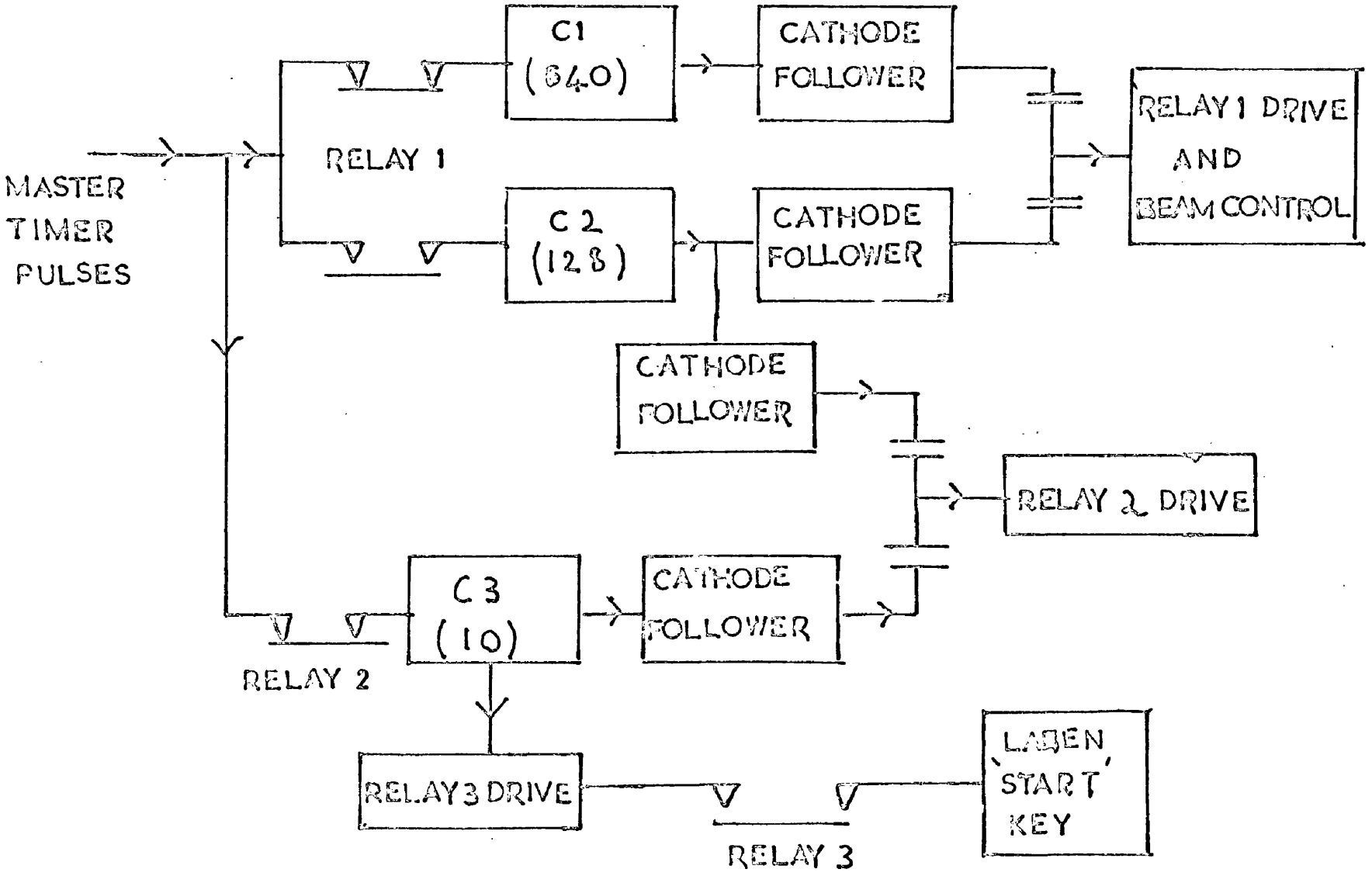


Fig. 20. Automatic control system for ^{19}Ne experiment.

time after each signal pulse. The pulses from the gate were fed to a Laben 1024-channel pulse height analyser, whose memory was split up to provide 16 independent subgroups of 64 channels each. Diode limiters were placed before and after the gate to limit the large pulses produced by positrons to a height of 38V, corresponding to channel 60 of an analyser sub-group; the lower channels were available for recording the K spectrum of ^{19}Ne in the counter.

Automatic Control System for Experiment

The timing system designed by the author to control irradiation of the counter and analysis of the resultant activity as a function of both time and pulse height is displayed in fig. 20. The system used as a "clock" the synchrotron master pulse generator which maintains the machine repetition frequency at 250 beam pulses per minute.

Pulses from this timer were fed, via relays 1 and 2 to pulse counters C1, C2 and C3, set to give output pulses for 640, 128 and 10 input pulses respectively. The cycle of events is as follows:-

(a) With the x-ray beam on, pulses passed to C2, until after about 30 sec, an output pulse was produced;

(b) This pulse operated relay 1, which switched off the beam and directed the master pulses to C1. It also operated relay 2 starting the counter C3;

(c) The output pulse from C3 operated relay 2 again, thus stopping C3 and also (via relay 3) actuating the "START" key of the

Laben analyser. The 2.5 sec delay between the beam going off and the start of the analysis gave ample time for the counter to recover from the intense ionisation produced by the beam.

(d) The analyser then recorded the pulse height spectrum from the counter in 16 consecutive subgroups, spending 9 sec in each subgroup, and thus following the decay of the ^{19}Ne produced.

(e) After 144 sec, the analyser passed into the "WAIT" condition, and a few seconds later, the output pulse from C1 operated relay 1 to restart the cycle.

The irradiation time was chosen on the basis of calculations of the relative quantities of ^{19}Ne, ^{11}C and ^{15}O as functions of irradiation time; these indicated an optimum irradiation time of just over 30 sec. Any ^{18}F activity produced can be regarded as constant in value over the 144 sec analysis period. The 2.5 sec value for the delay was larger than required (see later) but was immediately available from a simple dekatron tube counter.

4. EXPERIMENTAL PROCEDURE

As the activity produced by any 30 sec period of irradiation decays, the dead time fraction of the anti-coincidence gate changes, the dead time being different for each analyser subgroup. To determine the relationship between dead time and counting rate, scalers were connected to the clock and live-time outputs of the parallel gate described on page 36, and the open time fraction measured for various

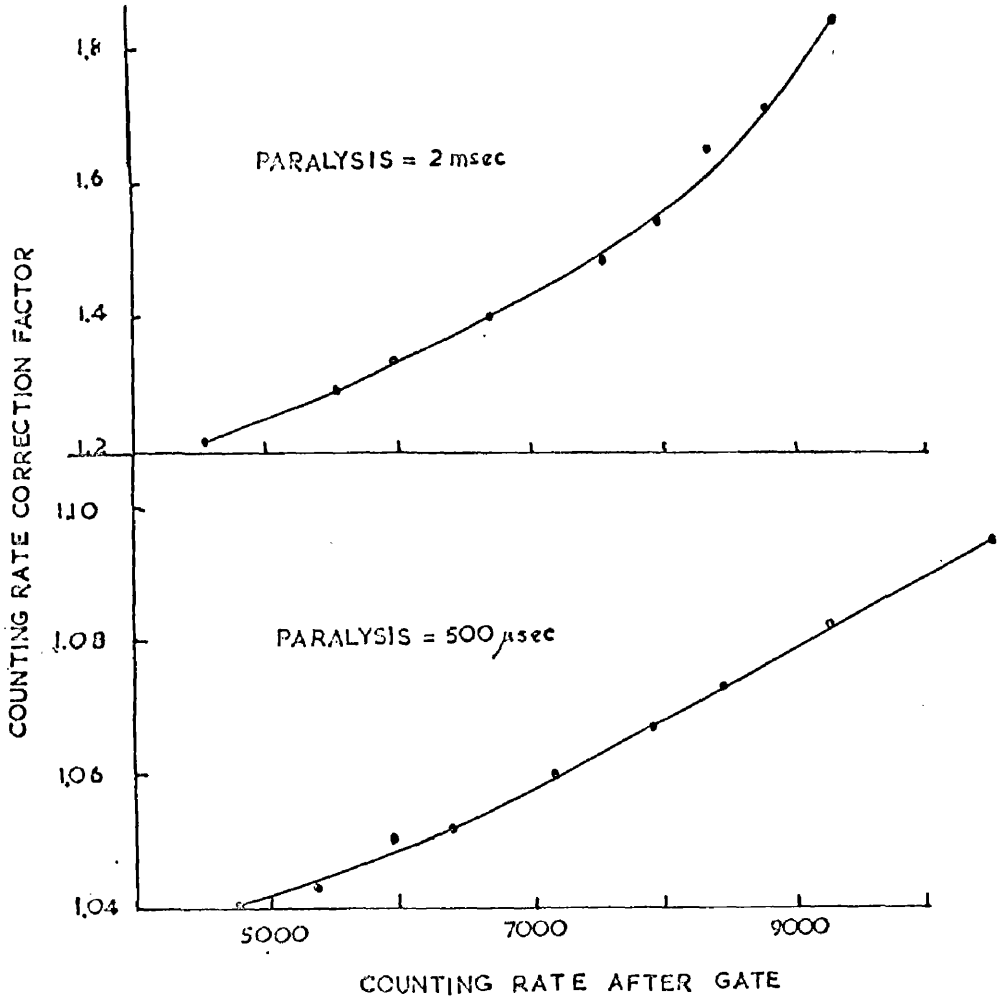


Fig. 21. Correction factors applied to counting rates to compensate for dead time of gate.

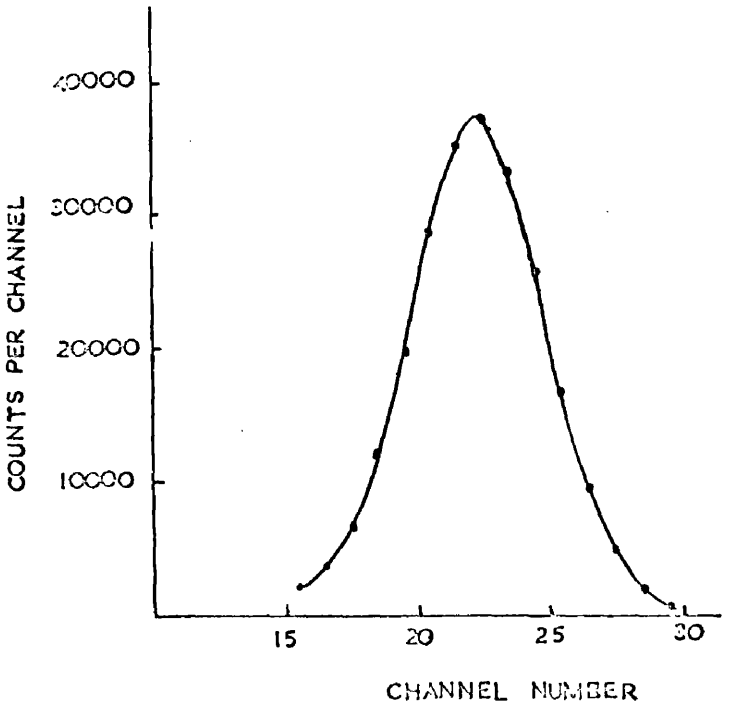


Fig. 22. Calibration peak from electron capture in ^{37}A (2.82 keV).

counting rates from an external gamma ray source. The correction factors for paralysis intervals of 2.2 msec and 500 μsec are plotted in fig. 21.

The counter was evacuated and allowed to outgas for several days before the final gas filling was introduced. The sodium-lead purifier was attached.

In a preliminary experiment to detect the time required for the counter to recover from the ionisation produced by passage of the x-ray beam, the 2.5 sec delay between the irradiation and analysis periods was removed and the time spent in each analyser sub-group reduced to 100 msec. A 9.2 keV ^{71}Ge x-ray peak was then recorded under actual running conditions, i.e. with the equipment under the control of the system shown in fig. 21. It was observed that the resolution of the peak became normal within 100 msec after passage of the beam, but that during the first 200 msec, there were spurious pulses in the region below 1 keV. The recovery time of the counter was therefore about 0.2 sec, considerably less than the 2.5 sec used during the runs on ^{19}Ne.

The overall gain of the system was set by placing the ^{71}Ge 9.2 keV peak in analyser channel 25, and increasing the amplifier gain by a factor of approximately 13.7 (measured by feeding pulses from a mercury pulse generator into the pre-amplifier) to analyse the energy region below 1.4 keV. Pulses corresponding to larger energies than this value were limited by the diode circuits and counted in analyser channel 60.

The main experiment was conducted over a running time of three weeks and runs were carried out at three different values of paralysis time as sown in Table 7. The counter was calibrated with the ^{71}Ge source at the start of each day's run, which lasted typically for about 9 hours, the apparatus performing about 170 cycles of irradiation and analysis. The average intensity of the x-ray beam, as recorded on a quantameter behind the counter was 1.5×10^{8} equivalent quanta per minute.

TABLE 7

DETAILS OF RUNNING TIME ON NEON 19 EXPERIMENT

Paralysis time	Number of Cycles	(Pulses in K region)/ (total pulses)
2.2 msec	1076	(7.1 ± .15) %
500 μsec	768	(7.4 ± .15) %
150 μsec	894	(7.6 ± .15) %

At the conclusion of the experiment, a small amount of argon 37 was injected into the counter and the electron capture peak at 2.82 keV recorded to obtain an accurate measurement of the counter resolution. The peak is shown in fig. 22.

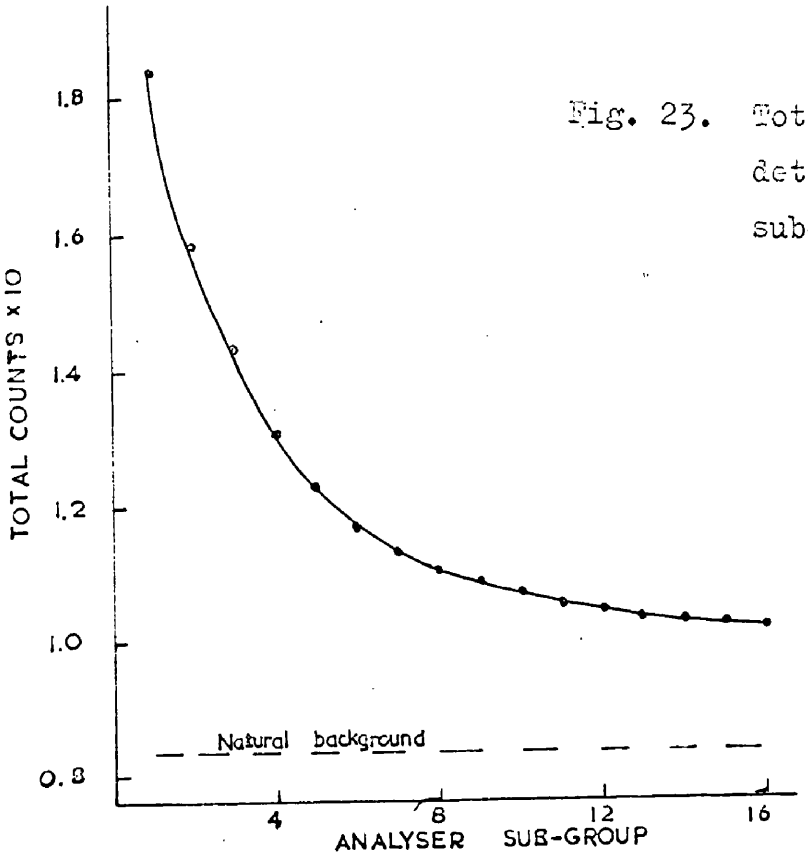


Fig. 23. Total number of pulses detected in each 9-sec sub-group.

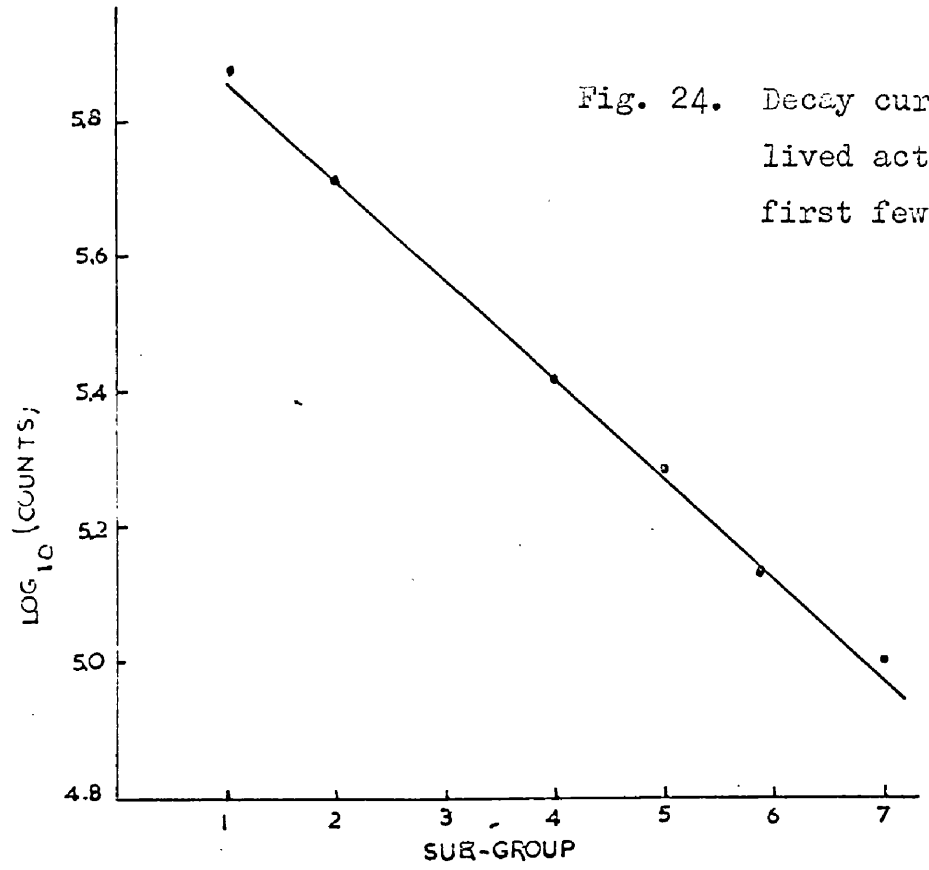


Fig. 24. Decay curve for short-lived activity in first few sub-groups.

5. ANALYSIS OF RESULTS

Three sets of experimental results were obtained, one for each value of the paralysis time. Each set of results consisted of 16 pulse height spectra (each of 64 channels) recorded in consecutive 9-second intervals during the decay of the activity produced. The results for the 2.2 msec paralysis time are discussed first.

Each subgroup was summed over all the channels to find the total number of pulses detected and therefore the average counting rate in each 9-sec. interval. The loss of pulses due to the action of the paralysis circuit was corrected for by multiplying each of the 16 spectra by the relevant time-normalisation factor obtained from the graph of fig. 21.

The corrected total number of pulses in each subgroup is plotted against sub-group number (i.e. time), in fig. 23; it is evident that the contribution from long-lived activities is not large, the totals in the last few subgroups exceeding natural background by about 20%. The decay of the long-lived contribution during the first three half-lives of ^{19}Ne being negligible, the total background intensity was taken as the average intensity in the final four sub-groups. When this background is subtracted from each of the earlier sub-group totals, the decay curve of the short-lived activity, shown in fig. 24, is obtained. The half-life measured from this curve is 18.5 sec, corresponding closely to the

half-life of ^{19}Ne, the small increase over the expected value probably being due to the presence of a small amount of ^{15}O.

Since this procedure appeared to separate the ^{19}Ne activity accurately, it was then applied to individual channels of the pulse height spectra to isolate the ^{19}Ne spectra.

It can be shown that in a case such as this, maximum accuracy is achieved by considering the activity detected in, roughly, the first two half-lives of the decay. Accordingly, the first four sub-groups were added together at each channel to give the pulse height spectrum due to ^{19}Ne and background, and the last four sub-groups to give that due to background only. The difference of these two spectra then gave the pulse height spectrum from ^{19}Ne in the first 36 seconds of analysis time, the K peak appearing in the centre channels and limited pulses in channel 60. It was checked that the half-life of the activity in the K region (below 1.5 keV) was the same as that of the activity giving limited pulses.

This procedure was repeated for the results obtained at lower paralysis, all the analysis being carried out on the KDF9 computer by the author.

The only difference observed in the final three spectra lay in the ratio of the number of pulses in the K region (up to channel 60) to the total number of pulses. As shown in Table 7, this ratio increased slightly with decreasing paralysis, perhaps indicating a contribution from negative ions. The three ^{19}Ne spectra were finally added to give the spectrum shown in fig. 25, the channel

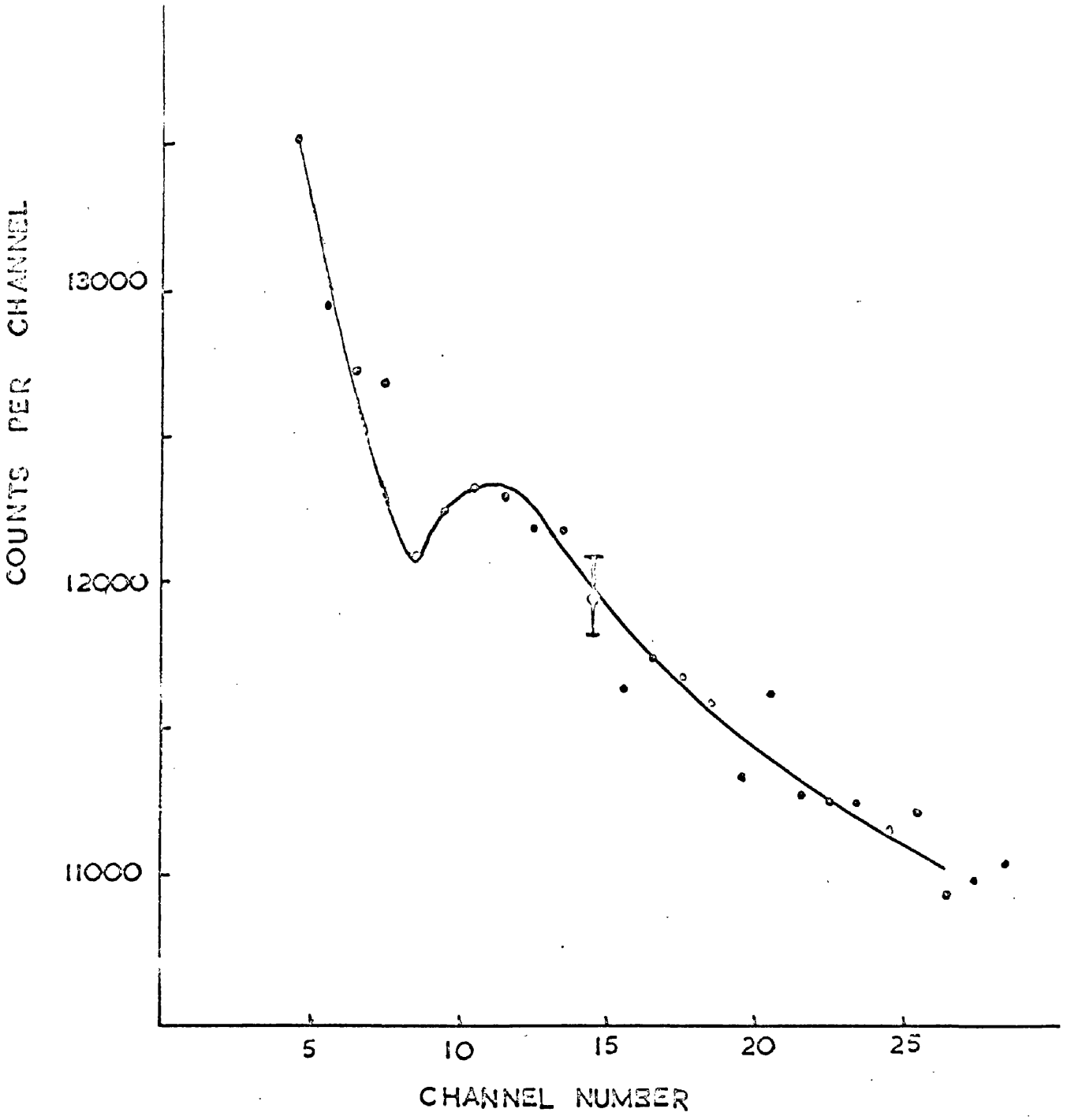


Fig. 25. Pulse height spectrum from ^{19}Ne in energy region below 1.5 keV.

width being doubled to improve the definition of the K peak; the number of limited pulses corresponding to this spectrum is 4.2×10^6.

One of the main points of interest concerning this final spectrum is the origin of the low energy background, constituting about 7% of the total intensity upon which the K peak is superimposed. To check that this was not due to a short-lived activity produced in the entrance and exit windows, the window thickness was doubled during an 8-hour run; no increase in the low energy background was observed. It would appear therefore that the background is due to ^{19}Ne positrons which have spent less energy than expected in the counter gas.

An approximate calculation indicated that the mean energy loss of a 1 MeV positron travelling from the centre of the activated cone directly towards the nearest wall is about 2 keV. Further calculations, taking into account the distribution of positron energies and the variation in path length from the activated cone to the walls, indicate that the intensity of small pulses due to Landau fluctuations in the energy loss (Ritson, 1961) should be less than one percent of the total counting rate.

It would appear therefore that the low energy background is, as anticipated, caused by diffusion of radioactive atoms towards the walls. This conclusion is not discouraging, however, since for isotopes of half-lives an order of magnitude smaller than

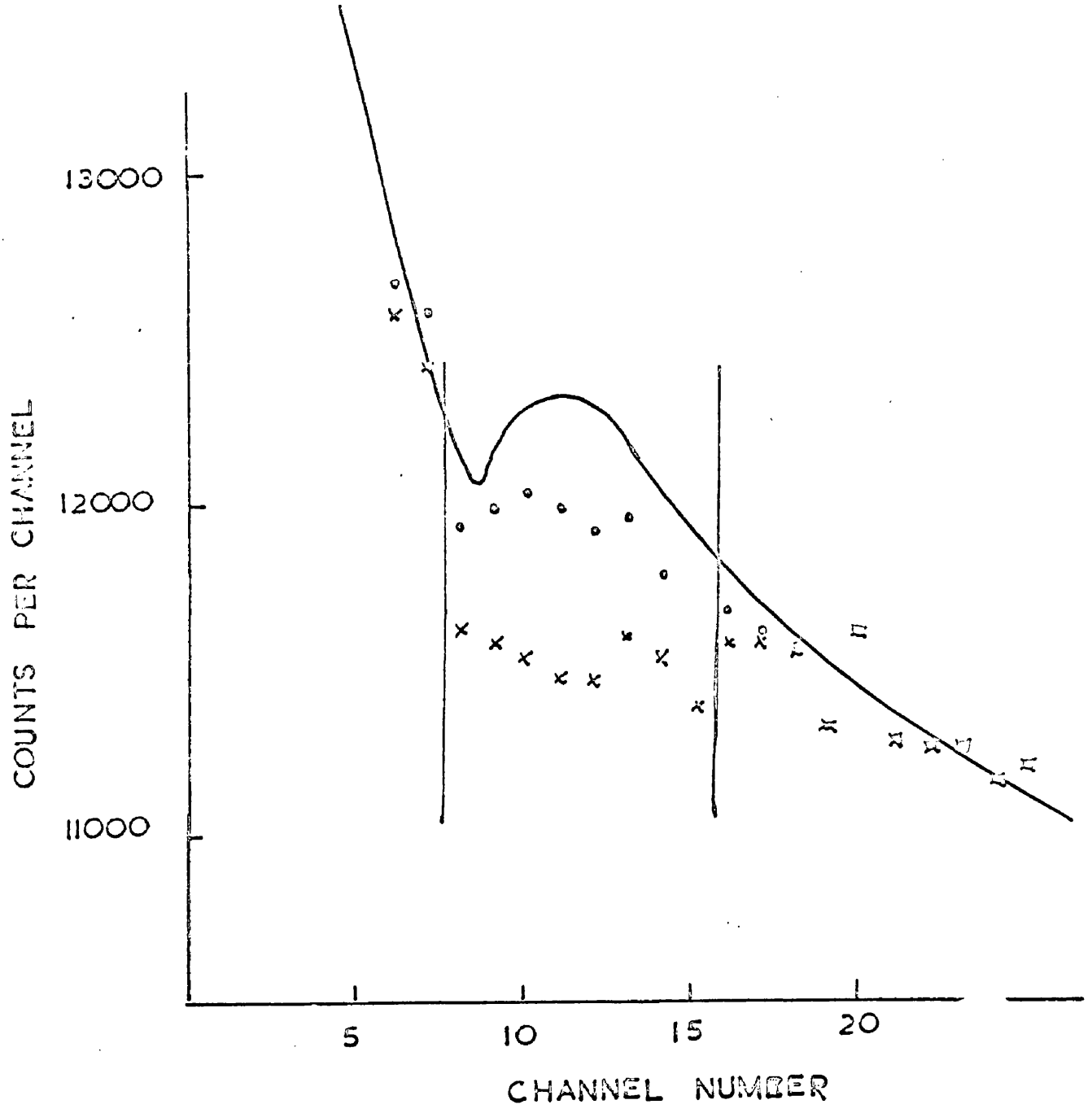


Fig. 26. Examples of "remainder" spectra (crosses and circles) obtained by subtraction of different Poisson distributions from the experimental ^{19}Ne spectrum. Both examples were rejected as corresponding to distributions which were respectively too high and too low.

that of ^{19}Ne, the effects of diffusion will be considerably less.

The Poisson parameter m for the expected ^{19}Ne electron capture pulse height distribution was obtained from the ^{37}A calibration peak shown in fig. 22. For the latter, the value of m (equal to $1/(\sigma_p/\bar{p})^2$) was calculated as 80 ± 6, and therefore, taking m proportional to energy, the value for the ^{19}Ne K peak should be 19.5 ± 1.5. The value of 26.4% for the ^{37}A resolution is a few percent greater than the values obtained in the work on line shapes and resolution, (perhaps due to the length of the counter) and accordingly the m value suggested for ^{19}Ne is lower than the optimum value; this is reflected by the fact that the mean pulse height of the ^{19}Ne peak is slightly less than would be expected from the ^{71}Ge calibration and the measured value (13.7) of amplifier gain.

In view of the large background below the K peak, it was not thought worthwhile to carry out any rigorous analysis of the results. A programme was written to evaluate Poisson distributions of various heights and with the above values of m, and to subtract these from the experimental spectrum. The resulting pulse height distributions were then examined for any departure from smoothness, and those Poisson distributions which resulted in a background spectrum which departed significantly from a monotonic decreasing function were rejected as being too high or too low. Examples of such backgrounds are shown in fig. 26. The areas under the acceptable Poisson distributions were calculated, the resulting

K/β^+ ratio being $(0.8 \pm 0.17) \times 10^{-3}$ (this value includes a 2% end-correction) in agreement with the theoretical ratio of 0.95×10^{-3} given earlier.

The accuracy of this measurement of the K/β^+ ratio of ^{19}Ne does not appear to afford a particularly useful comparison with theory; nevertheless, this is the only measurement on ^{19}Ne, and the value of the result depends to a considerable extent on the, as yet uncalculated, overlap and exchange corrections.

The result is, however, encouraging when viewed as a test of the new technique. One might reasonably expect a considerable increase in accuracy, other factors being equal, on applying the direct irradiation technique to isotopes of half-lives an order of magnitude less than that of ^{19}Ne.

CHAPTER SEVEN

CONCLUSIONS

The proportional counter is at present finding an increasing number of applications in the 10-100 Å wavelength range. It would appear that the work described here on low energy pulse height distributions helps to consolidate the role of the instrument as a reliable, quantitative detector of soft x rays and electrons. The author hopes to extend this work shortly into the ultra-violet region.

As regards K/β^+ ratios, the work described here, together with that of Ledingham (1963), augments the experimental data at very low atomic number and does not indicate any serious failure or omission in the available theory, although the necessity for further theoretical work has been emphasized. The flow technique, with the improvements made during the ^{11}C experiment, and the direct irradiation technique as developed in the ^{19}Ne experiment, are both now available for future application and the choice between these techniques, for the investigation of the isotopes ^{34}Cl and ^{35}A, is at present under active consideration.

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SUMMARY OF THESIS

PROPORTIONAL COUNTER STUDIES OF ELECTRON CAPTURE TO POSITRON EMISSION RATIOS IN LIGHT ISOTOPES

By John L. Campbell B.Sc.

The introductory chapter of this thesis comprises a brief review of the theory of nuclear beta decay, with emphasis on electron capture to positron emission ratios in allowed transitions. Reasons for interest in the ratio of K capture to positron emission are discussed, and the techniques which have been employed to measure this quantity are outlined. A summary is given of the available experimental data for isotopes of low atomic number.

There have been few measurements of K/β^+ ratios in Fermi decays, and few in decays where recent calculations have suggested that inclusion of atomic matrix elements in the conventional theory might alter the K/β^+ ratio by an observable amount. The need for such measurements, and also for measurements at very low atomic number where, as yet, there is little accurate theoretical information, is emphasised.

The proportional counter techniques used by the author to investigate gaseous electron capture sources of low atomic number are described in the second chapter. Different types of continuous flow counters designed to minimise the "wall effect" are discussed, and methods of overcoming the difficulties arising from the short half-lives and weak electron capture intensities of the isotopes of interest are outlined.

Two chapters are devoted to a study of the response of proportional counters at very low energy, undertaken to provide the information necessary for the analysis of pulse height distributions produced by electron capture sources. The distributio produced by monochromatic x-rays of energies below 3 kev were investigated and found to approximate closely to the Poisson functi a further experiment determined the contribution to the width of such distributions from statistical variations in the gas multiplication process.

A measurement of the K/β^+ ratio in the 21 minute decay of carbon 11 is described[2]; the special plastic anticoincidence counter used was designed earlier by Drever for carbon dating work, but was well suited to the present application, where propane, irradiated by the x-ray beam of the Glasgow synchrotron was flowed through it. The result was consistent with the available theory.

Finally, the development of a new technique for application to very short-lived sources is described; this method, in which a proportional counter is exposed directly to the activating x-ray beam to produce the isotope of interest, was suggested for use in conjunction with the high intensity beam of the new Glasgow linear accelerator. A preliminary experiment, using the synchrotron to irradiate a neon-filled counter, gave a result for the K/β^+ ratio of neon 19 in agreement with theory, and indicated that the technique

would be well suited to half-lives of the order of a second.

The results on K/β^{+} ratios obtained here do not indicate any serious failure or omission in the available theory, while the study of proportional counter response helps to consolidate the role of the instrument as a reliable detector of soft x-rays and ultra-violet radiation.

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2. Nuclear Physics, (1967), to be published.