STUDIES OF ORBITAL ELECTRON CAPTURE USING PROPORTIONAL COUNTERS

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PREFACE.

This thesis contains an account of experimental work performed by the writer at Glasgow University during the period between October, 1953 and October, 1956.

In the first chapter the theory of orbital electron capture is briefly summarised, and experimental data available on K/β^+ ratios and L/K capture ratios are surveyed. The accuracy of many of the experimental results is poor, but it is pointed out that the development of proportional counter techniques for studying low energy radiations has made better measurements possible. These techniques are discussed in the following chapter, with particular emphasis on the use of gaseous sources. The material of this introductory section of the thesis is drawn mainly from the literature, though an appreciable part of the discussion on gas source counters is based on the writer's own experience.

The next two chapters relate to the development of new proportional counter techniques, made with the aim of overcoming some of the limitations to the use of gas sources. An investigation into the operation of proportional counters at high temperatures, initiated by Dr. A. Moljk and carried out jointly by him and the writer,

is described; and the development of a multiwire proportional counter system with small wall effects is discussed, particularly in relation to the measurement of weak sources. The principle of the latter counting system was due to Dr. Moljk and the writer, following an initial suggestion by Dr. S.C. Curran, and the experimental work and its interpretation was shared equally by us. The detailed design of the counter and of the associated electronic equipment (described in an appendix) was due largely to the writer.

The final three chapters, forming the main part of the thesis, deal with studies of electron capture carried out with the aid of gas source techniques. An investigation of the decay of Cl^{36}, leading to the discovery of a previously undetected K-capture branch, is described in Chapter 5. This work was done jointly with Dr. Moljk.

Chapter 6 concerns a measurement of the K/β^+ ratio in the decay of F^{18}, which enables limits to be set on Fierz interference effects in Gamow-Teller interactions. This experiment was performed in collaboration with Dr. Moljk, assisted by Mr. J. Scobie; though the writer takes the main responsibility for the results and their interpretation.

Chapter 7 deals with a study of L-capture in Ge^{71}. The L/K capture ratio was measured by a new direct method using a multiwire counting system; and the results obtained show that previous work suggesting serious disagreement with theory is not valid. The writer was entirely responsible for the original conception of this experiment, its performance, and the interpretation of the results.

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PUBLICATIONS.

The work described in this thesis has been published in the following papers:-

Operation of Proportional Counters at High Temperatures;

A. Moljk, R.W.P. Drever and S.C. Curran,
Rev. Sci. Inst., 26, 1034, (1955).

A Proportional Counter System with Small Wall Effect;

R.W.P. Drever, A. Moljk and S.C. Curran,
Nucl. Inst., 1, 41, (1957).

The Background of Counters and Radiocarbon Dating;

A. Moljk, R.W.P. Drever and S.C. Curran,
Proc. Roy. Soc., A 239, 433, (1957).

K-Capture in the Decay of Chlorine 36;

R.W.P. Drever and A. Moljk,
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The Ratio of K-Capture to Positron Emission in Fluorine 18;

R.W.P. Drever, A. Moljk and J. Scobie,
Phil. Mag., 1 (8th S.), 942, (1956).

The L/K Capture Ratio of Germanium 71;

R.W.P. Drever and A. Moljk,
Phil. Mag., 2 (8th S.), 427, (1957).

Other work carried out during the same period but not discussed in this thesis is described in the following publications:-

Measurement of Tritium as Water Vapour;

R.W.P. Drever and A. Moljk,
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Neutron Activation Applied to Potassium-Mineral Dating;

A. Moljk, R.W.P. Drever and S.C. Curran,
Nucleonics 13/2, 44, (1955).

Proportional Counters for Demonstration Experiments;

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CONTENTS

INTRODUCTION

Chapter 1.

RADIOACTIVE DECAY BY ORBITAL ELECTRON CAPTURE

General Description.

The apparent violation of the principles of conservation of energy and momentum in the beta decay of radioactive nuclei has long been a stimulus to the study of this type of process. The early suggestion by Pauli that an unobserved particle might be involved indicated a way of avoiding some of the difficulties; and the subsequent development of the neutrino theory by Fermi and others, with the explanations of spectrum shapes and half-lives which have followed, have well justified the original hypothesis.

The process of beta decay may be taken to involve a transformation of a neutron into a proton with a simultaneous production of the two light particles, the electron e^- and the neutrino ν :-

$$N \rightarrow P + e^- + \nu \ .$$

Decay by positron emission might be considered as an analogous event:-

$$P \rightarrow N + e^+ + \nu' ,$$

where ν' represents an antineutrino, not necessarily identical

with a neutrino. However, positron decay may also be regarded from another point of view as the direct inverse of negative beta emission, an electron and neutrino being drawn from negative energy states and being absorbed in the nucleus:-

$$P + e^- + \nu \longrightarrow N,$$

the absorption of the electron and neutrino corresponding here to the emission of a positron and an antineutrino.

This way of looking at positron decay suggests that electrons in positive energy states, such as those in the atomic shells, might also be absorbed in a radioactive decay; and this possibility was pointed out in 1935 by Yukawa and Sakata, and by Bethe and Backer. Such a process of decay by electron capture was difficult to detect experimentally, but two years later Alvarez demonstrated its existence in the decay of vanadium 48.

The initial energy of the electron absorbed by a nucleus in a K-capture event is $mc^2 - w_K$ where mc^2 is the rest energy of an electron and w_K is the K-shell binding energy; while in the case of positron decay the corresponding initial electron energy is $- mc^2$. Thus K-capture is energetically possible whenever positron emission takes place. Taking the atom as a whole, K-capture can in fact

occur when the masses M_1 and M_2 of the parent and daughter atoms satisfy the inequality

$$M_1 - M_2 > w_K$$

while positron decay is only possible when

$$M_1 - M_2 > 2mc^2 + w_v \ .$$

Here w_v is the binding energy of the valency electron in the parent atom, which is almost always negligible.

Capture of electrons from the L and M atomic shells can also take place, and is energetically slightly favoured over K-capture. The probability of occurrence of these outer electrons at the nucleus is relatively small, though, and in practice decay constants for L-capture are usually only about one tenth of the corresponding K-capture decay constants. There is, however, considerable interest in accurate measurement of the relative probabilities of L-capture and K-capture in the decay of a given isotope, since this provides a method of determining the energy difference between the initial and final nuclei.

The detailed theory of beta decay and electron capture has been well described by many authors (for example Blatt and Weisskopf, 1952; Konopinski and Langer, 1953; Rose, 1955) and it is not proposed to recount it here. It may be useful, though, to summarise the main results on

decay constants and branching ratios in allowed transitions which are of interest in connection with the present experimental work.

Decay Constants for Electron Capture and Positron Emission.

Both electron capture and beta decay are taken to depend on the same interactions between the neutron, proton, electron and neutrino so the theoretical treatment of the two processes is very similar. The Fermi theory gives in either case a decay constant which involves the wave functions of the electron and the neutrino at the nucleus and a matrix element describing the nuclear transition, though for beta decay a factor arising from the statistical distribution of energy and momentum between the electron and neutrino has to be included. In allowed decays, which are the only ones considered here, the calculations can be greatly simplified by neglecting all but the first terms in the expansion of the matrix elements.

In the case of K-capture, the decay constant λ_K obtained may be written in the form

$$\lambda_K = \frac{1}{4\pi^2} (W_o + W_K)^2 \cdot g_K^2 \cdot |M|^2 \qquad \text{— (1)}$$

where Wo is the total decay energy;

Wk is the energy of the K atomic electron $|(W_K = 1 - w_K)$;

g_K is the large component of the Dirac wave function of the electron, taken at the nucleus;

and M is the matrix element for the transition.

To evaluate g_K^2 hydrogen-like wave functions may be used, as was done by Marshak (1942) in the original calculations on electron capture. The value for g_K^2 is then given by the expression

$$g_K^2 = \frac{1 + W_K}{2\Gamma(2\gamma+1)} \cdot (2\alpha Z_K)^3 e^{-2\alpha Z_K R} \cdot (2\alpha Z_K R)^{2\gamma-2} \qquad -(2)$$

where $\gamma = (1-\alpha^2 Z_K^2)^{1/2}$;

$Z_K = Z - 0.3$ is the effective nuclear charge corrected for screening;

$R = 3.9 \times 10^{-3} \cdot A^{1/3}$ is the nuclear radius (in units of $\hbar/mc$);

and $\alpha = 1/137$ is the fine-structure constant.

Since the work of Marshak, more accurate results for electron wave functions have become available, and for all but low values of atomic number it is more appropriate to use the values of g_K^2 computed by Rose and Jackson (1949) and Brysk and Rose (1955).

In the case of positron decay it is necessary to integrate over electron energy W in finding the total decay constant. The expression then obtained for the decay constant λ_+ for positron decay is

$$\lambda_+ = \frac{1}{2\pi^3} \int_1^{W_0} p\, W\, (W_0 - W)^2 \cdot F(Z,W) \cdot |M'|^2 \, dW \qquad -(3)$$

where p is the momentum of the positron, corresponding to energy W, and F(ZW) is the Fermi function describing the

effect of the Coulomb field on the electrons. Values of the Fermi function have been computed by Rose (1955) and are available in tabular form.

In Fermi's original work the matrix element was taken to describe an interaction involving only a scalar coupling between the particles. This gave selection rules for allowed transitions

$$\Delta J = 0, \text{ no change of parity.}$$

Gamow and Teller showed that an interaction involving the spins of the particles is also possible, giving selection rules

$$\Delta J = 0 \text{ or } \pm 1, \text{ (no } 0 \rightarrow 0\text{) no change of parity.}$$

A complete relativistic treatment of beta decay indicates that altogether five types of interaction might occur: scalar, vector, tensor, axial vector, and pseudoscalar. The scalar and vector interactions both give Fermi selection rules for allowed decay, and the tensor and axial vector interactions give Gamow-Teller selection rules. The pseudo-scalar interaction corresponds to different selection rules, which seem to be contradicted by experimental data. De Groot and Tolhoek (1946) have developed a general theory involving a linear combination of all five interactions, with relative strengths C_S, C_V, C_T, C_A, and C_P respectively.

If the pseudo-scalar interaction is omitted, then the expression found for the matrix element $|M|^2$ in equation (1) is

$$|M|^2 = G^2\left[(C_S^2 + C_V^2)|M_F|^2 + (C_T^2 + C_A^2)|M_{GT}|^2 \cdot (1 + b)\right] \qquad -(4)$$

and the expression for $|M'|^2$ in equation (3) is

$$|M'|^2 = G^2\left[(C_S^2 + C_V^2)|M_F|^2 + (C_T^2 + C_A^2)|M_{GT}|^2 \cdot \left(1 - \frac{\gamma b}{W}\right)\right] \qquad -(5)$$

In these two expressions G is a universal beta decay constant, and M_F and M_{GT} are matrix elements corresponding to the interactions obeying Fermi and Gamow-Teller selection rules, respectively. The term b arises from an interference effect when both scalar and vector, or tensor and axial vector, interactions are present. This Fierz interference term is given as

$$b = \frac{2\left[(C_S \cdot C_V)|M_F|^2 + (C_T \cdot C_A)|M_{GT}|^2\right]}{(C_S^2 + C_V^2)|M_F|^2 + (C_T^2 + C_A^2)|M_{GT}|^2} \qquad -(6)$$

The same interference constant b enters also into expressions for spectrum shapes in allowed beta decay, where it modifies the normal statistical spectrum by a factor proportional to $1/W$.

The determination of the values of the coupling constants has been for a long time one of the main problems in the study of beta decay. The position has been reviewed by Wu (1955) and Konopinski (1955). The experimental data

require that both Fermi and Gamow-Teller interactions occur, with approximately equal strengths. Measurements of the shapes of allowed beta spectra give no sign of a term in 1/W, indicating that the Fierz constant b is certainly not large. There have been several estimated upper limits to the magnitude of this constant. In the case of Gamow-Teller interactions Konopinski and Langer (1953) put a limit $|b| < 0.04$; though Davidson and Peaslee (1953) set the limit as $|b| < 0.08$. Winther and Kofoed-Hansen (1953) pointed out the difficulties in detecting a small deviation from an allowed spectrum shape and concluded that a value of $|b|$ as high as 0.40 could not be ruled out. There is less experimental data on the Fermi interactions, and here Davidson and Peaslee have put an upper limit to $|b|$ of 0.20.

Further information on the value of the Fierz term b can be obtained from study of the relative probabilities of K-capture and positron emission in an allowed decay, as will be discussed later.

From the expression (6) for the Fierz interference term and the experimental results it is possible to make some conclusions about ratios of coupling constants. In the case of the Gamow-Teller interactions, for example, a

limit for $|b|$ of 0.08 implies that either $|C_A/C_T|$ or $|C_T/C_A|$ is less than 0.04. Experiments on the angular correlation between the beta particle and the neutrino emitted in the decay of helium 6 (Rustad and Ruby, 1955) indicate that $|C_A| < |C_T|$, so it may be concluded that $|C_A/C_T|$ is less than 0.04.

It may be remarked that the recent discovery that parity is not conserved in beta decay renders the interpretation given here rather less definite, as will be indicated later.

Relative Probabilities of K-Capture and Positron Emission.

From equations (1), (3), (4) and (5) the ratio, R, of the decay constants for K-capture and positron emission in transitions between the same initial and final nuclei may be shown to be

$$R = \frac{\lambda_K}{\lambda_+} = \frac{\pi g_K^2 (W_0 + W_K)^2 (1+b)}{2\int_1^{W_0} p W (W_0 - W)^2 . F(Z,W) . (1 - \frac{\gamma b}{W}) dW} \qquad -(7)$$

If b is taken to be zero, this ratio reduces to

$$R_0 = \left(\frac{\lambda_K}{\lambda_+}\right)_{b=0} = \frac{\pi g_K^2 (W_0 + W_K)^2}{2\int_1^{W_0} p W (W_0 - W)^2 . F(Z,W)\, dW} \qquad -(8)$$

The latter equation does not involve matrix elements, so the value of R_0 for any particular decay can readily be calculated accurately. Thus if b is assumed zero from

the results on spectrum shapes, then a close agreement between R_0 and a measured K/β^+ ratio would provide a check of the theory. Alternatively, assuming the relevant theory to be correct, comparison of R_0 with a measured K/β^+ ratio would enable limits to be set on b independently of the data on spectrum shapes. Sherr and Miller (1953) have in fact made a measurement of the K/β^+ ratio in sodium 22 for this purpose, and conclude that $|b|$ is less than 0.04. There may, however, be some doubt whether this particular decay is allowed.

Relative Probabilities of L-Capture and K-Capture.

Decay constants for L-capture were calculated first by Marshak (1942). In the case of allowed decays capture takes place mainly from the L_1 subshell, with a small contribution from the L_2 subshell. The decay constant λ_L for L-capture is given as

$$\lambda_L = \frac{1}{4\pi^2}(W_0 + W_L)^2 (g_{L_1}^2 + f_{L_2}^2) . |M|^2 \qquad -(9)$$

where g_{L_1} and f_{L_2} are the large and small components of the Dirac wave functions corresponding to the L_1 and L_2 electrons. In computing these L-shell parameters hydrogen-like wave functions do not give a good approximation, and the results calculated by Brysk and Rose are preferable.

The ratio of the probabilities of L-capture and K-capture

in a given decay, λ_L/λ_K, is found from equations (1) and (9) as

$$\frac{\lambda_L}{\lambda_K} = \frac{(W_0 + W_L)^2}{(W_0 + W_K)^2} \cdot \left(\frac{g_{L_1}^2 + g_{L_2}^2}{g_K^2} \right) \qquad -(10)$$

This L/K capture ratio does not involve nuclear matrix elements, so, provided the transition energy and the atomic wave functions are known, it is a suitable quantity for experimental checking. However, transition energies for nuclei which decay only by electron capture are difficult to measure, and in fact the majority of measurements of L/K capture ratios have been made with the aim of determining transition energies. In the few cases when L/K capture ratios and transition energies have been measured separately, large discrepancies between theory and experiment have appeared, and these are stimulating more detailed study.

Note on Recent Developments in Beta Decay Theory.

Difficulties in interpretation of the two separate modes of decay of the apparently identical τ and θ mesons recently led Lee and Yang (1956) to suggest that parity may not always be conserved in weak interactions. It was subsequently demonstrated experimentally by Wu et al (1957), and later by Postma et al (1957), Schopper (1957), Grace et al (1957) and others, that parity is in fact not conserved in the beta decay of several nuclei. The angular distribution of beta particles emitted from nuclei with their spins aligned in a known direction was found to be asymmetrical, emission being more likely in a direction antiparallel to the nuclear spin direction than parallel to it in the case of negative electrons, and in the opposite direction in the case of positrons. It has also been found, in violation to conservation of parity, that the beta particles from certain nuclei are longitudinally polarised (Fraunfelder et al, 1957; Cavanagh et al, 1957; and others), the spins being predominantly parallel to the direction of flight for positrons, and antiparallel to it for negative electrons.

The experimental work of the author, described in this thesis, was all carried out prior to this important development, and the relevant theoretical results have

naturally been described in terms of the interpretation then current. It is interesting to inquire, however, how the results concerning K/β^+ ratios and L/K capture ratios are affected by the recent extension of beta decay theory.

In developing a theory of beta decay without assuming conservation of parity, Lee and Yang (1956, 1957) introduced parity non-conserving interaction constants C_S', C_V', C_T', C_A' and C_P' in addition to the five previous parity-conserving constants C_S, C_V, C_T, C_A, and C_P. The breakdown of conservation of parity has caused other assumptions made in the earlier theory to be questioned also, but if it is assumed that the interactions are still invariant to time reversal, which is not in contradiction with experiment at present, then all the interaction constants are real. The expressions then obtained for decay constants and spectrum shapes are rather like the previous ones, but C_S^2 in the earlier expressions is replaced by $\frac{1}{2}(C_S^2 + C_S'^2)$, $C_S.C_V$ is replaced by $\frac{1}{2}(C_S.C_V + C_S'.C_V')$, and there are similar changes in the other constants. It is evident that such alterations have no effect on the equations for K/β^+ ratios and L/K capture ratios, (7), (8) and (10) given already.

The only material change that results is in the interpretation of the Fierz term b. A small value for b in a Gamow-Teller decay does not now necessarily imply that either C_A or C_T is small, and in fact it indicates instead that

$$C_A/C_A' \doteq -C_T'/C_T \ .$$

However, the two-component neutrino theory developed by Lee and Yang, and also by Salem (1957) and Landau (1957), requires that either $C_S' = -C_S$, $C_V' = -C_V$, etc., or that $C_S' = C_S$, $C_V' = C_V$, etc. Such relationships are not in disagreement with the experimental data, and if they are taken to be correct then the new expression for b reduces exactly to the old one, (6), and the previous results remain unchanged.

It may be concluded, then, that the introductio n of parity-nonconserving theories of beta decay has little effect on the work on K/β^+ and L/K ratios described in this thesis, though it may render conclusions drawn about ratios of coupling constants rather less definite. It may be remarked though, that in the present unsettled state of beta decay theory reliable results on combinations of the coupling constants, such as given by K/β^+ measurements, may have even greater value than before.

EXPERIMENTAL METHODS OF MEASURING ELECTRON CAPTURE.

The information which can be obtained from ratios of decay constants for K-capture, L-capture, and positron emission has now been indicated, and it may be useful to review the experimental techniques which are employed for measuring these ratios. The only observable results of an electron capture event are the disappearance of an electron from an inner atomic shell, leading to emission of X-rays or Auger electrons as the vacancy is refilled; and the appearance of a daughter nucleus, which may subsequently emit gamma radiation. Both these effects of electron capture have been used in experimental studies of the process.

Measurement of K/β^+ Ratios.

The most obvious way of determining decay constants for K-capture and positron emission is by direct measurement of the X-rays and positrons themselves, and this has in fact been done for a number of isotopes. The K/β^+ ratios in Cu^{61} and Cu^{64}, for example, were measured in this way by Bouchez and Kayas (1949) and Huber et al (1949), the X-rays being detected by Geiger counters; while similar work on Fe^{52} has been carried out by Friedlander and Miller (1951) with a thin-window proportional counter. All these measurements suffer, though, from the difficulties of

determining absolute counter efficiency and the corrections required for absorption of X-rays in the source and the counter window.

To avoid absolute counting some workers have measured the Auger electrons emitted after K-capture rather than the X-rays themselves, so that the same instrument can be used to determine both K-capture and positron intensities. Cook and Langer (1948) used this method for Cu^{61} and other nuclei, with a magnetic spectrometer; but the small penetrating power of the K Auger electrons (of energy 8 keV) made scattering and absorption serious. Avignon (1953, 1955) measured Zn^{65} in a similar way, though with a proportional counter; and even Na^{22} has been investigated as a solid source in a 4π Geiger counter by Charpak (1955). In the latter case the maximum Auger electron energy is 0.8 keV so scattering and self-absorption is severe, and this probably accounts for the fact that the K/B^+ ratio obtained is much lower than that given by other measurements.

Chemical or mass-spectrometer analysis of decay products provides a completely different method of determining branching ratios and this has been applied in measurements of K/β^+ ratios in Cu^{64} and Na^{22}. In the former case the source decays by negative beta emission as well as

positron emission and K-capture, and the K/β^+ ratio was found (Reynolds, 1950) by comparing the rates of production of the two daughter elements with the known β^+/β^- ratio; while in the work on Na^{22} (Allen et al, 1955) the rate of production of neon was compared with the rate of decay by positron emission, determined by absolute counting of the positrons. The technique seems rather specialised, however, and hardly suitable for general application.

The difficulties of direct measurement of the low energy atomic radiations accompanying K-capture have led to the development of a number of indirect methods of determining K/β^+ ratios, based on gamma emission from excited states of the daughter nuclei. In such measurements the total rate of decay, by positron emission and electron capture, is compared with the rate of decay by positron emission alone. Sodium 22 was measured in this way by Sherr and Miller (1953, 1954), who used a scintillation counter to determine the gamma ray intensity, and a 4π Geiger counter for the positron intensity. Here the absolute efficiencies of the two counters had to be found separately, which made the experiment complex and introduced a large number of corrections. Other workers have arranged to use the same detectors for both positron decay and gamma

emission so that fewer corrections are required. Thus Good et al (1946) measured the positron annihilation radiation instead of the positrons themselves, employing a coincidence technique with two gamma counters to distinguish between the annihilation radiation and the nuclear gamma rays. On the other hand, Bradt et al (1945) measured positrons, and internal conversion electrons associated with the gamma transition, using a magnetic spectrometer. In both these experiments, corrections still had to be made for variations of detection efficiency with energy.

The values for K/β^+ ratios obtained by the various indirect methods seem generally to be more reliable than those which have been found by direct measurements of X-rays or Auger electrons. Nevertheless, the indirect measurements themselves are not entirely satisfactory. They usually involve a considerable number of instrumental corrections; and moreover they determine the ratio of the total decay constant to the decay constant for positron emission alone, so that values for L/K capture ratios have to be assumed. Further, the indirect techniques are clearly inapplicable to simple ground-state to ground-state decays which are the most suitable for comparison with theory. Direct methods of measuring K/β^+ ratios, if of

sufficient accuracy, would therefore be preferable.

The development of proportional counter techniques for measuring radioactive sources in gas form has made possible more accurate experiments on low energy electrons and X-rays, and thus, in principle, on electron capture intensities also. These low energy proportional counter techniques will be discussed in detail later in this thesis, but it may be remarked here that their application seems generally to have been rather neglected. The only published measurement of a K/β^+ ratio made with a gas source in a proportional counter is that of Townsend (1951), in which Zn^{65} was studied in the form of the gas $Zn(C_2H_5)_2$. The value found for the K/β^+ ratio in this work was twice that obtained in other experiments, and it appears that the counter was not arranged to detect the positrons efficiently. The final result was obtained by extrapolation of observed ratios measured at different pressures, but it seems likely that this procedure was invalidated by the effects of escape of X-rays from the counter, which do not vary linearly with pressure. In principle, though, this type of measurement can be made accurate and reliable, and similar techniques have been used in much of the work of the present author.

It should be mentioned for completeness that a gas source technique of a different kind has been used by Radvanyi (1952, 1955) in a measurement of the K/β^+ ratio of Kr^{79}. Here the gas was put into a cloud chamber, and the number of short tracks due to X-rays and Auger electrons was compared with the number of positron tracks photographed. Individual inspection of each track was necessary, so it was difficult to analyse enough events to give good statistical accuracy.

Measurement of L/K Capture Ratios.

The energies of the L X-rays from any particular atom are of the order of one tenth of those of the corresponding K radiations, so measurement of L-capture intensity is considerably more difficult than measurement of K-capture. In certain cases, however, L/K capture ratios can be found by indirect methods analogous to those used for K/β^+ ratios.

Iodine 125, for example, decays to an excited state of Te^{125}, and the L/K capture ratio can be determined by measuring the K X-ray intensity and comparing it with the total rate of decay indicated by the intensity of gamma emission. This particular experiment was done by Friedlander and Orr (1951), and the same method has been

applied to Pd^{103} by Avignon (1953). A somewhat similar approach has been used by Der Mateosian (1953) in work on I^{125} and Cd^{109}, though here an ingenious scintillation counter technique was employed in the measurement of the K and gamma radiations. The NaI(Tl) crystal used was grown from a solution of the radioactive material being studied, so that the source was uniformly distributed throughout the detecting medium, and errors from scattering and self-absorption were eliminated.

It might be mentioned also that Heintze (1954) has reported detection of the 3 keV Auger electrons from K^{40}, as a solid source in a proportional counter; and from their apparent low intensity has deduced an unusually high L/K capture ratio. It seems to the writer that the low K-capture intensity observed was more probably due to scattering difficulties in the measurement than to a high L-capture probability.

A general difficulty with indirect methods of finding L/K capture ratios is the relatively low intensity of L-capture, and, together with the uncertainties in determinations of the K-capture intensities themselves, this tends to make the accuracy of the results low. Direct measurements of L/K capture ratios have their own difficulties, however.

The most straightforward direct method consists of a measurement of intensities of L and K X-rays from the electron capture source, as has been done, for example, by Fink and Robinson (1955) and by Bisi, Terrani and Zappa (1955) with thin-window proportional counters. Here, though, it is necessary to take into account not only the variation of detection efficiency with energy, but also the possibility of simultaneous emission of L and K α X-rays following K-capture events. In practice it is often not possible to distinguish between the L X-rays due to L-capture itself and those due to K-capture, and the latter are usually by far the more numerous. The correction for this "background" of L-radiation requires knowledge of the K fluorescence yield of the daughter atom, and the value of the L/K capture ratio then obtained depends strongly on this factor. Fluorescence yields are not yet known sufficiently accurately, so there is considerable uncertainty in the results. Similar troubles arise if the L and K Auger electrons are measured instead of the X-rays, as has been done by Radvanyi (1952), Langevin and Radvanyi (1954) and Langevin (1954b) for Kr^{79} and Ge^{71}. These latter experiments were made with proportional counters containing gas sources, and will be discussed in detail later.

It is possible to avoid spurious recording of L-capture events following K-capture with K_α emission if both X-rays and Auger electrons are measured at the same time. This can be done, for example, with a gas source in a proportional counter, provided that the counter is large enough to detect essentially all the X-rays emitted by the source. X-rays and Auger electrons emitted simultaneously after a K-capture process are then recorded as a single pulse, which corresponds to the total K-absorption energy of the daughter atom. Thus only true L-capture events are registered in the L-peak, and the L/K capture ratio is given directly by the ratio of the counting rates in the two peaks. This technique was employed by Pontecorvo, Kirkwood and Hanna (1949) in the original discovery of L-capture in A^{37}, and has also been used in a recent remeasurement of the L/K capture ratio in this isotope by Langevin and Radvanyi (1955). In a suitable case, such as that of A^{37}, this total-absorption technique can probably give a more accurate value for an L/K capture ratio than can any other method.

Summary of Experimental K/β^+ and L/K Capture Ratios for Allowed Decays.

To give an idea of the kind of results which have been obtained by measurements such as those discussed above

it may be useful to summarise some of the experimental data available. Fairly detailed reviews of the work on K/β^+ ratios have been given by Cook (1951a), Bouchez (1952), and Radvanyi (1955); while the results available on L/K capture ratios have been reviewed by Robinson and Fink (1955) and by Radvanyi (1955).

Experimental results from some of the main measurements of K/β^+ ratios are given in Table 1, together with the values expected theoretically. Many of the measurements have been made by indirect methods which give total capture probabilities, and in these cases the K/β^+ ratios given here have been deduced using the theoretical graphs of L/K capture ratios given by Brysk and Rose (1955).

It will be seen from the table that there is a considerable amount of scatter in the experimental results, and the accuracy of most of the measurements is low. The experiments with the smallest stated errors are those of Sherr and Miller on Na^{22}, and of Good, Peaslee and Deutsch on Co^{56}; but in both these cases there is some uncertainty about the type of beta transition involved. The majority of the measured K/β^+ ratios lie in the neighbourhood of the expected values, but it is evident that more work on simple, well-understood decays is

TABLE 1.

EXPERIMENTAL AND THEORETICAL VALUES OF K/β^+ RATIOS.

Nucleus	Measured K/β^+ ratio	Theoretical K/β^+ ratio	Author	Remarks
Na^{22}	0.0 ± 0.05 0.09 ± 0.05 0.04 ± 0.03 0.060 ± 0.009 0.104 ± 0.006 0.116 ± 0.010	0.107 (allowed) or 0.08 (1st forbidden) or 0.05 (L-forbidden)	Good et al (1946) Bouchez (1952) Major (1952) Charpak (1955) Sherr and Miller (1954) Allen et al (1955)	Type of decay uncertain; allowed, 1st. forbidden, or L-forbidden.
Mn^{52}	1.7 ± 0.2	1.6	Good et al (1946)	Allowed
Fe^{52}	1.5 ± 0.4	3.2	Friedlander and Miller (1951)	Allowed
Co^{58}	5.4 ± 0.2	5.0 (allowed)	Good et al (1946)	Allowed or 1st. forbidden
Cu^{61}	0.55 ± 0.06 0.44 ± 0.07) 0.39 ± 0.05) 0.32 ± 0.03 0.47	0.52	Bouchez and Kayas (1949) Bouchez (1952) Huber et al (1949) Cook (1951b)	Complex decay scheme

TABLE 1 (Continued).

Nucleus	Measured K/β^+ ratio	Theoretical K/β^+ ratio	Author	Remarks
Cu^{64}	2.6 ± 0.3	2.1	Bouchez and Kayas (1949)	Allowed
	3.5 ± 1.0		Cook and Langer (1948)	Complex decay scheme
	2.32 ± 0.28		Reynolds (1950)	
	1.75 ± 0.2		Huber et al (1949)	
	1.81 ± 0.2 or 2.07		Plasman and Scott (1951)	This result depends on fluorescence yield
Zn^{65}	32.5 ± 6	29 (allowed) or 18 (1st forbidden)	Sakai and Hubert (1953)	Many other scattered results
	17.5 ± 2		Avignon (1953)	
	37 ± 6		Perrin (1953)	
	28.0 ± 3.2		Perkins and Haynes (1953)	Allowed or 1st. forbidden
	130 ± 25		Townsend (1951)	
Ga^{68}	1.9	2.2	Mukurji and Preiswerk (1952)	Allowed
Br^{80}	2.0 ± 0.2	1.9	Laberrique (1951)	Allowed
	1.3 ± 0.2		Mims and Halban (1951)	
Cd^{107}	280 ± 30	320	Bradt et al (1945)	Allowed

TABLE 2.

EXPERIMENTAL AND THEORETICAL VALUES OF L/K CAPTURE RATIOS.

Nucleus	Measured L/K ratio	Theoretical L/K ratio	Author	Remarks
A^{37}	0.085 ± 0.02 $0.092 \,^{+\,0.010}_{-\,0.005}$	0.082	Pontecorvo et al (1949) Langevin and Radvanyi (1955)	Allowed decay. Total-absorption proportional counter measurements.
K^{40}	1.35 ± 0.23	$0.21 \,^{+\,0.33}_{-\,0.06}$	Heintze (1954)	Experimental result doubtful. 2nd. forbidden decay.
Kr^{79}	0.26 ± 0.03 ($F=\bar{0}.57$) or 0.22 ± 0.03 ($F=\bar{0}.586$)	0.101	Langevin and Radvanyi (1954) Radvanyi (1955)	Allowed decays. Experimental results depend on fluorescence yield F. The second value in each case is that suggested by Labberique-Frolow, Radvanyi and Langevin (1956)
Ge^{71}	0.30 ± 0.02 ($F=\bar{0}.45$) or 0.25 ± 0.02 ($F=\bar{0}.477$)	0.106	Langevin (1954b, 1956)	
Pd^{103}	0.79 ± 0.22	0.120	Avignon (1953, 1956)	Experimental result may be affected by internal conversion X-rays. Allowed decay. Complex decay scheme.

desirable for a reliable check of the theory.

Experimental results on L/K capture ratios are less numerous than those on K/β^+ ratios, and most are unsuitable for comparison with theory since the relevant decay energies are not known. However, there are a few nuclei for which both L/K capture ratios and transition energies have been measured, and the observed and calculated ratios for these nuclei are summarised in Table 2.

It is evident from the data that there is considerable disagreement between theory and experiment, but, as already noted, most of the measurements are not free from uncertainty. The only completely reliable L/K capture ratio seems to be that for A^{37}, and this is in satisfactory agreement with the theoretical result. It is possible that this one well-established agreement is more significant than the apparent discrepancies in the four other cases, but more experimental work is necessary before definite conclusions can be drawn.

(It should be mentioned that some results for heavy elements are not included in Table 2. The decay schemes involved are complex and uncertain, while the measurements may be affected by X-rays due to internal conversion).

To conclude this chapter it may be remarked that the scarcity of accurate electron capture data which has been emphasised is due largely to the difficulty of measurement of low energy electrons and X-rays. It has been pointed out that improved proportional counter techniques can, in principle at least, make better measurements possible. Much of the work of the author has been concerned with the development of proportional counter techniques for these, and other, purposes; and it may be useful to review now some general features of proportional counter measurement of low energy phenomena.

Chapter 2.

THE MEASUREMENT OF LOW ENERGY ELECTRONS AND X-RAYS WITH PROPORTIONAL COUNTERS

The brief review of experimental data on electron capture given in the previous chapter is sufficient to indicate how the experimental study of this type of decay has been hampered by the difficulties of measuring the low energy X-rays and Auger electrons emitted. The introduction of proportional counter techniques has now enabled these difficulties to be largely overcome, particularly when gas sources are used. The present chapter will therefore be devoted to some aspects of the use of proportional counters in low energy measurements. As the techniques are valuable in several other fields besides that of electron capture, the discussion will not be restricted entirely to this particular application.

Good general reviews of proportional counter techniques have been given by Curran and Craggs (1949), Rossi and Staub (1949), Wilkinson (1950), Pontecorvo (1950), West (1953) and Curran (1955).

Operating Mechanism of the Proportional Counter.

A normal proportional counter consists essentially of a thin anode wire mounted along the axis of a relatively

large cylindrical cathode, with the intervening space filled by a suitable mixture of gases. Any electrons produced in the gas move, under the influence of the applied electric field, towards the anode wire, where the field strength increases rapidly. With a suitable voltage applied to the counter, the field becomes large enough at a few tenths of a millimetre from the wire to cause the electrons to ionise. The number of electrons present then doubles at the end of each subsequent free path, and the number finally collected at the wire may be of the order of 10^4 times the number initially produced by the primary particle.

Photons are emitted in this multiplication process, and to prevent these producing photoelectrons at the cathode it is usual to include a quenching gas, such as methane or carbon dioxide, in the filling. These molecular gases strongly absorb ultraviolet photons, their molecules breaking up in the process. The quenching gas also has the important function of preventing positive ions created in the avalanche from releasing secondary electrons at the cathode. A process of charge exchange takes place at collisions between positive ions and molecules of the quenching gas, and the resultant molecular ions dissociate at the cathode instead of extracting electrons.

The collection at the wire of the electrons produced in the avalanche leads to a negative voltage pulse, with an initial rise time of the order of a tenth of a microsecond. This part of the output pulse is not large, though, since most of the electrons move through only a relatively small difference of potential; and the main component of the pulse comes from the movement of the positive ions away from the wire. When short resolving times are not required it is convenient to differentiate the pulse by a circuit with a time constant of a few microseconds, so that almost all of the positive ion component is measured and the maximum pulse height is obtained. When beta particles having long tracks in the counter are being studied it is sometimes necessary to increase the differentiating time constant further so that the orientation of the tracks does not affect the pulse height, but it is seldom that a time constant of more than 15 microseconds is required.

The multiplication factor in a proportional counter can be made large if necessary; and it is possible, for example, to count single thermal electrons produced at the cathode. The effects of space charge in the avalanche set an upper limit, however, to the gain which

can usefully be employed with primary particles of a given energy. These effects depend on the total ionisation density in the avalanche, and not on the gas multiplication itself; and it can always be arranged that they are unimportant if a low-noise amplifier is used.

The fundamental restriction in low energy measurements is the deterioration of energy resolution which occurs when the number of primary ion pairs becomes small. This is due principally to statistical fluctuations in the production of ions by the primary particle, and to a lesser extent to statistical fluctuations in the multiplication process itself. These effects set a lower limit of about 200 eV to the energies which can be satisfactorily measured. At high energies the energy resolution of a proportional counter is generally inferior to that of a magnetic spectrometer, but in low energy work this limitation is outweighed by other advantages, such as the source arrangements possible and the good counting geometry. The energy resolution is, however, always better than that of a scintillation spectrometer.

The gas multiplication in a proportional counter is only uniform throughout the sensitive volume if the field strength at the wire does not vary along its length. In

a simple counter this condition is not fulfilled, since end effects tend to make the field fall off towards both ends of the wire. Several devices have been introduced to overcome this difficulty, but the most satisfactory is the use of field tubes (Cockroft and Curran, 1951). Metal tubes are fitted at both ends of the counter, coaxial with the wire, and are maintained at a potential equal to the undisturbed potential at a distance from the wire equal to their radii. With this arrangement the gas multiplication is modified by end effects only in the region inside the field tubes, the volume of which can easily be made negligible.

Source Arrangements.

Proportional counters fitted with thin windows are sometimes employed for measurements of beta particles or X-rays from sources mounted externally; but this technique is not suitable for radiations of weak penetrating power, and even with high energy beta particles corrections for absorption in the window have to be made. Since it is mainly in the low energy region that the proportional counter has advantages over other types of electron or X-ray spectrometer, it is usually desirable to avoid the deterioration in performance resulting from the use of a window, and the radioactive sources are normally put inside the counter itself.

A common method of arranging the source is to coat it over the inside of the cylindrical cathode. This gives almost 2π counting geometry, and the large surface area available permits the use of thin sources even when the radioactive material is of low specific activity. Both these features of this technique make it particularly suitable for studies of radioactive isotopes of long half-life, and it has been extensively used for this purpose by Curran, Wilson and others at Glasgow (for references see Curran, 1955). With a source on the wall of a cylindrical proportional counter there is, though, a relatively high probability that beta particles of long range may pass out of the counter before spending all their energy. When the spectrum shape has to be measured accurately the source is therefore often mounted on a thin plastic film supported on a hollow probe near the centre of the counter. By using a high gas pressure, and applying a magnetic field parallel to the wire if necessary, electrons with energies up to about 500 keV can then easily be prevented from passing out of the gas.

In low energy work with proportional counters the use of solid sources tends to impose some limitations that prevent the good low-energy performance of the counter

itself from being fully exploited. The most obvious trouble is self-absorption in the source material. The range of a 2 keV electron, for example, is about 24 $\mu g/cm^2$, while that of a 500 eV electron is only 4.5 $\mu g/cm^2$. For measurements extending down to energies of this order, sources with a thickness of not more than a few micrograms per square centimetre would therefore be required. The preparation and checking for uniformity of such thin sources is extremely difficult, if not impossible, and material of very high specific activity is essential.

An even more serious difficulty is the large amount of scattering suffered by low energy electrons. Measurements of backscattering from material supporting radioactive sources indicate that at energies of a few hundred keV about 40% of the electrons may be reflected, and at very low energies the fraction backscattered may become even larger. Another related effect is the scattering of electrons from the filling gas back into the source. There does not seem to have been any experimental study of this phenomenon, but from the tortuous nature of the tracks of low energy electrons observed in cloud chambers it would appear to be of considerable importance.

These various scattering and absorption effects tend to make measurement of electrons of energies less than

about 5 keV, coming from solid sources, difficult and inaccurate. This may be illustrated by some work of the writer, carried out during a study of Cl^{36} which will be described later. Chlorine 36 decays partly by K-capture, and with a source in the form of methyl chloride gas mixed with the filling of a proportional counter, the peak in the pulse spectrum due to the 2.5 keV Auger electrons was plainly apparent. With a thin solid source on the wall of the counter, however, no definite peak could be detected; and only a broad bump in the spectrum, representing a counting rate less than 10% of that in the true peak, was observed. In this experiment the solid source was made by reaction of chlorine gas with the cathode of the counter, and if a uniform distribution of the radioactive material is assumed, the thickness was less than 0.03 $\mu g/cm^2$.

Gas Sources.

The scattering and absorption phenomena that limit low energy measurements with solid sources become unimportant if the radioactive material is converted into suitable gaseous form and is put into the counter with the filling gas. The source is then part of the detecting medium itself, and all the energy spent in ionising the source contributes to the observed pulse. Thus scattering

of electrons is of no consequence, and in a suitable case the low energy performance of the system is determined only by the fundamental limitations of statistical fluctuations in the ionisation and multiplication processes.

If the counter is large enough, and the filling gas dense enough, to make effects of electrons passing out of the gas negligible, then the beta spectrum of a gas source may be obtained directly from the pulse height distribution. With an electron capture source a similarly direct result can be obtained provided that escape of X-rays from the gas is also negligible. The counting geometry is effectively 4π and the energies of all the atomic radiations associated with any single electron-capture event are integrated by the counter; so a K-capture process gives a pulse corresponding to the K absorption energy of the daughter atom, and an L-capture event gives a readily-distinguishable pulse corresponding to the L absorption energy. Thus in suitable cases a proportional counter with a gaseous source can give direct information on beta spectra and electron capture which is difficult to obtain by other methods. This technique has therefore been used in several beta decay studies (such as, for example, that on C^{14} and S^{35} by Moljk and Curran, 1954), as well as

in the work on electron capture already referred to.

With a gaseous beta source it can usually be arranged that almost every disintegration which occurs in the working volume of the counter is recorded, and this high counting efficiency is valuable in measurements of radioactive materials of low specific activity. Such measurements are necessary in studies of long-lived isotopes, but a more extensive application of low level counting techniques has grown up recently in connection with the dating of archaeological specimens by the abundance of C^{14} in their carbon content (see Libby, 1952). The endpoint of the beta spectrum of C^{14} is 150 keV, and the specific activity of the carbon measured in this work is always less than 15 disintegrations per minute per gm, so equipment sensitive for beta particles of both low energy and low intensity is required. Proportional counters are now commonly employed in this work, with the carbon sample in the form of carbon dioxide or some other suitable gas used as the filling itself.

Having indicated now some of the main features of proportional counter measurement of radioactive sources in gaseous form, it may be useful to discuss some of the difficulties which tend to restrict the application of this technique.

Wall Effects.

Many of the limitations of proportional counters arise from the low stopping power of a gas and the practical limits to the size and working pressure of the apparatus. The pulses from a proportional counter represent the energy of the radiations emitted by the source only if the radiations are completely absorbed by the gas in the sensitive volume, and this condition is seldom satisfied perfectly. With a gas source there is a relatively high probability for escape of radiations emitted in the region adjacent to the wall of the counter, so wall effects have always to be considered.

In general the escape of any radiation results in a decrease in the number of large pulses in the measured spectrum and an increase in the number of small ones, but the detailed effects of this depend on the kind of source being studied. In the case of an alpha or beta source, more than one quarter of the particles of a given range r originating within a distance r from the wall, pass out of the gas before spending their whole energy. Thus, in a counter of diameter D large compared with r, more than $(1 - r/D).r/D$ of the particles of range r emitted by the source give pulses which correspond to energies lower

than their true energy. In a measurement of a beta source, this results in a decrease in counting rate in the high energy part of the observed spectrum, and a rapid rise in the observed spectrum at the low energy end. This type of wall effect, besides disturbing spectrum studies, also hinders investigation of other phenomena, such as electron capture, which may occur in the presence of beta emission.

In measurements of electron capture, escape of X-rays from the counter leads to rather similar effects. The intensity of the K-capture peak in the pulse spectrum is reduced, and that of the lower-energy L-peak is increased. The relative effect on the L-peak is usually the greater, and for accurate measurements of L/K capture ratios it is desirable to make X-ray escape effects considerably smaller than those which could be tolerated in studies of K-capture intensities alone.

Escape of beta particles or X-rays from the sensitive volume of a proportional counter can take place through the ends as well as through the sides. In the case of the end effect, though, radiations may also enter the sensitive volume from the part of the source outside it. In counters of normal proportions these end effects

are usually considerably smaller than the wall effects, so they will not be considered further here.

All the wall effects mentioned above can be reduced by increasing the diameter of the counter or the pressure of the filling gas, though there are practical limits to the improvement which can be obtained in this way. In measurements of beta sources the wall effects can be further reduced by the application of a magnetic field to curl up the tracks of the particles. The use of filling gases of high atomic number, such as krypton or xenon; may also give some improvement; but their high fluorescence yield is a disadvantage in X-ray measurements.

Chemical Restrictions to the Use of Gas Sources.

Although wall effects lead to some of the major restrictions in the use of gas sources with proportional counters, another limitation, more chemical in nature, arises from the difficulty of obtaining many of the elements in a suitable gaseous form. Some elements, such as the alkali metals, have no simple stable compounds which are gases at room temperature, and so are outside the scope of normal gas source technique.

Further, when a gas compound is available, it must not have properties which would disturb the operation

of the counter. Thus a gas with a high electron attachment coefficient is undesirable, since electrons moving towards the wire may be captured to form negative ions, leading to a reduction in pulse height. In this case there is also a deterioration in energy resolution due to statistical fluctuations in the loss of electrons, and to variation of the probability of attachment for electrons originating at different distances from the wire. In certain applications of proportional counters it may be difficult to avoid using a gas which has an appreciable attachment coefficient, and in this connection some work of the author (Drever and Moljk, 1956) on the measurement of tritium as water vapour may be of interest. The attachment coefficient of water vapour depends strongly on the agitation velocity of the electrons, and it was found that by suitable choice of the proportions of argon and methane in the filling it was possible to bring the agitation velocity throughout the main part of the counter into a region of minimum attachment coefficient, thus obtaining satisfactory energy resolution.

It is evident that a gaseous source, in addition to having a low attachment coefficient, should also be

chemically inert with respect to other materials present in the counter. This condition is often more difficult to satisfy than might be expected, since physical adsorption, or reaction with substances adsorbed on the wall of the counter, is possible even when there should be no reaction between the gas and the wall material itself. With a radioactive gas of high specific activity, even a monomolecular layer deposited on the wall may correspond to an appreciable fraction of the total quantity of source in the counter.

The limitations to gas source measurements which have just been mentioned tend to restrict the energy region and the range of isotopes which can be investigated. However, within these limits the use of gas sources with proportional counters enables measurements of electrons and X-rays to be made at energies lower than can be reached by other methods, and has some special advantages in studies of decay by electron capture.

In view of this, the work of the author to be described in this thesis has had two principal objectives. One has been the further development of proportional counter gas source techniques in general; while the other has been

the application of these techniques to measurements of orbital electron capture.

THE DEVELOPMENT OF SOME NEW GAS SOURCE PROPORTIONAL COUNTER TECHNIQUES.

Chapter 3.

THE OPERATION OF PROPORTIONAL COUNTERS AT HIGH TEMPERATURES.

It has been pointed out in the Introduction that accurate measurement of electrons with energies of less than a few keV is difficult when solid sources are used, and that for many investigations on electron capture and beta decay it is more satisfactory to convert the radioactive material into gaseous form and include it in the filling of a proportional counter. The applicability of this technique is, however, severely restricted by the difficulty of obtaining suitable gas compounds of many of the elements, and this prevents low energy measurements on a number of interesting isotopes. Most of the elements, though, do possess compounds which have an appreciable vapour pressure at temperatures of a few hundreds of degrees Centigrade, and it might therefore be possible to extend the scope of gas source techniques if the experiments could be performed at such temperatures. With this in view, a study has been made of the possibility

of operating proportional counters at elevated temperatures.

There has been no previous published work on heated proportional counters, though some investigations on the properties of Geiger tubes at temperatures up to 200°C have been reported. Meunier, Bonpas and Legrand (1955) found that the background of their Geiger counters increased considerably above a certain critical temperature of the order of 50°C, and suggested that this was due to the emission of sodium ions from the cathode. Similar effects have been observed by Om Parkash (1950) who ascribed them to secondary emission by residual mercury ions, and by Joshi (1953).

Preliminary Considerations.

The mechanism of a proportional counter is distinct from that of a Geiger counter in that it depends only on ionisation processes in the gas itself. These mainly involve energy transfers of the order of 15 eV, which is large compared with the mean thermal energy of a gas molecule, even at a temperature of 1000°C. Change of temperature might thus be expected to have little direct effect on the operation of a proportional counter, provided that the counter is sealed off so that the mean free path for electrons in the gas does not alter.

However, any thermal process by which electrons or negative ions are produced at the cathode would clearly prevent normal counting. One possible process of this type is thermionic emission of electrons, and this phenomenon, at least, might be expected to set one limit to the working temperature. Prediction of the magnitude of thermionic emission is difficult since the presence of the filling gas and of surface impurities may alter the effective work function of the cathode material appreciably. If, however, the work function of the cathode were 4.3 eV, which is the published value for copper, then it would emit one electron per cm^2 per second at $540^{\circ}C$, and about 10^6 electrons per cm^2 per second at $770^{\circ}C$. The latter electron flux would certainly disturb the working of a normal proportional counter.

It was decided in the present work to make a general study of the factors limiting the operating temperature, and to determine whether counting is in fact possible at temperatures for which thermionic emission from normal materials becomes appreciable. To this end a number of different types of proportional counter were developed, and their operation at high temperatures was investigated.

Experiments on Glass Counters with Internal Cathodes.

In the first experiments on heated proportional counters, glass counters of fairly conventional design were used. A diagram of such a counter is given in Fig. 1. The cathode is made of nickel, since this material has a higher work function than most other common metals. Electrical leakage from the cathode to the anode wire is prevented by painting bands of Aquadag on the inner and outer surfaces of the glass and keeping them at the same potential as the wire. Such guard rings are often omitted in proportional counters for use at normal temperatures, but the rapid decrease in the electrical resistivity of glass with rise in temperature made them essential in the present work. The glass used here was a hard borosilicate glass (P-9, by B.T.H. Ltd.) with a resistivity of 10^{14} ohm. cm at 20°C, which falls to 10^{7} ohm. cm at 400°C.

Before filling and sealing off, each counter was thoroughly degassed at a temperature of about 540°C. The main fillings used were normal gas mixtures, usually argon and carbon dioxide at partial pressures of 30 cm Hg and 7 cm Hg, respectively. Methane, the more common quenching gas, was not employed since it decomposes at

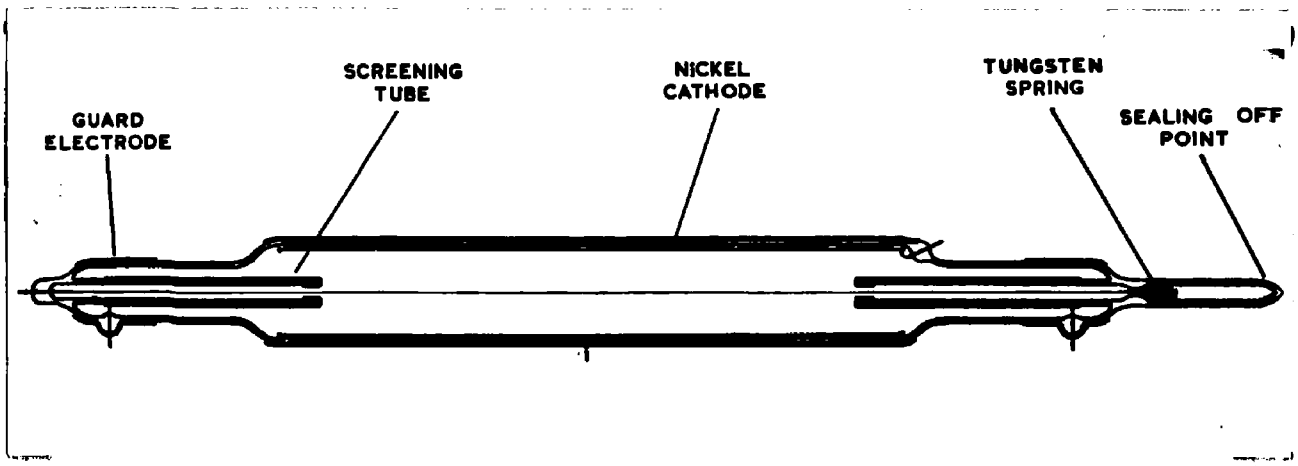


Fig. 1.

Glass proportional counter for use at high temperatures.

Thickened lines indicate conducting coatings.

about 500°C.

It was desired to check energy resolution, gas multiplication, and the absence of spurious pulses at various temperatures, so a small quantity of A^{37} was included in the filling of each counter. This electron-capture source emits the 2.8 keV K radiation and the 250 eV L radiation of chlorine, and analysis of the pulse spectra corresponding to these low energy X-rays and Auger electrons provides a sensitive test of the operation of a proportional counter. The pulses were analysed by a multichannel kicksorter, and were also examined visually on an oscilloscope; while total counting rates were monitored by a scaler.

A thermostatically controlled furnace was used to heat the counters, and it was lined internally with stainless steel to reduce electrical interference from the heater windings. Connections to the counter inside the furnace were enclosed in thick-walled silica tubes, and the signal from the anode wire was brought out through a screened lead with ceramic insulation.

The first experiments showed that energy resolution and counting rate do not change appreciably with temperature in the range from 20°C to 170°C. However,

at a temperature slightly above 170°C some spurious pulses were noticed, and these rapidly became more numerous with further increase of temperature. The amplitude of the spurious pulses was found to be almost independent of the gas multiplication in the counter, which suggested that they were not due to electrons or ions coming from the cathode. Further investigation showed that the effect was due to sparking between the nickel cathode and the partially-conducting glass envelope; and it could readily be prevented by coating the outside of the glass with Aquadag in the neighbourhood of the cathode, and keeping the coating at the same potential as the cathode. This spurious effect seems in some ways rather similar to the effects which have been observed with Geiger counters, and as the conductivity of heated glass seems to have been overlooked in the previous work, it is not impossible that this may have been partly responsible for the great increase of counting rate with temperature which has been reported.

When practical difficulties, such as the one just mentioned, had been overcome it was found that a glass counter like that in Fig. 1 could be operated satisfactorily at temperatures up to 450°C. This is demonstrated by the pulse spectrum in the region of the A^{37} K-peak shown in

Fig. 2, and of the L-peak in Fig. 3, both of which were measured at a temperature of 450°C. No changes in counting rate were detected throughout.

At temperatures above 450°C the energy resolution of counters like that in Fig. 1 usually deteriorated, due to evolution of gases from the internal Aquadag coatings; but a more serious difficulty arose from the reduction of the resistivity of the glass. At the working voltage of 2.2 kV, the leakage current flowing in the glass from the cathode to the guard rings was about 1 ma at 450°C, and it rose to 4 ma at 540°C. This leakage current did not have any immediate effect on the operation of a counter, but after some time electrolysis effects in the glass became apparent. The graphite guard rings became less efficient, and when about 1 coulomb of electricity had passed through the glass, direct leakage to the wire occurred. This may be explained by the removal of metal ions from the glass adjacent to the guard rings, with a resultant local increase in resistivity. Thus the guard rings became effectively insulated from the glass by a layer of silica.

Other effects of the conductivity of hot glass were also observed, though these were of a less serious

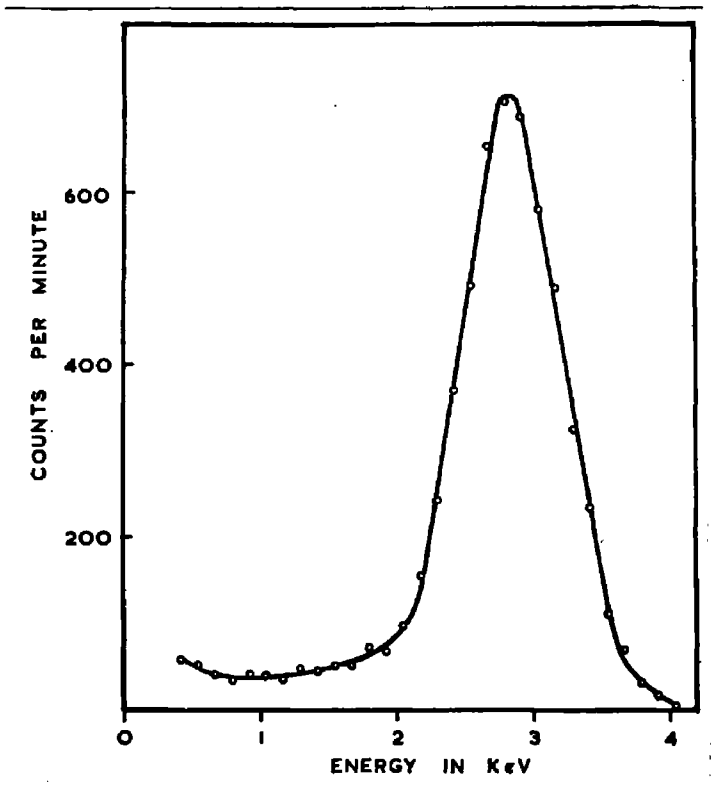


Fig. 2. K-peak of A^{37} taken at 450° C.

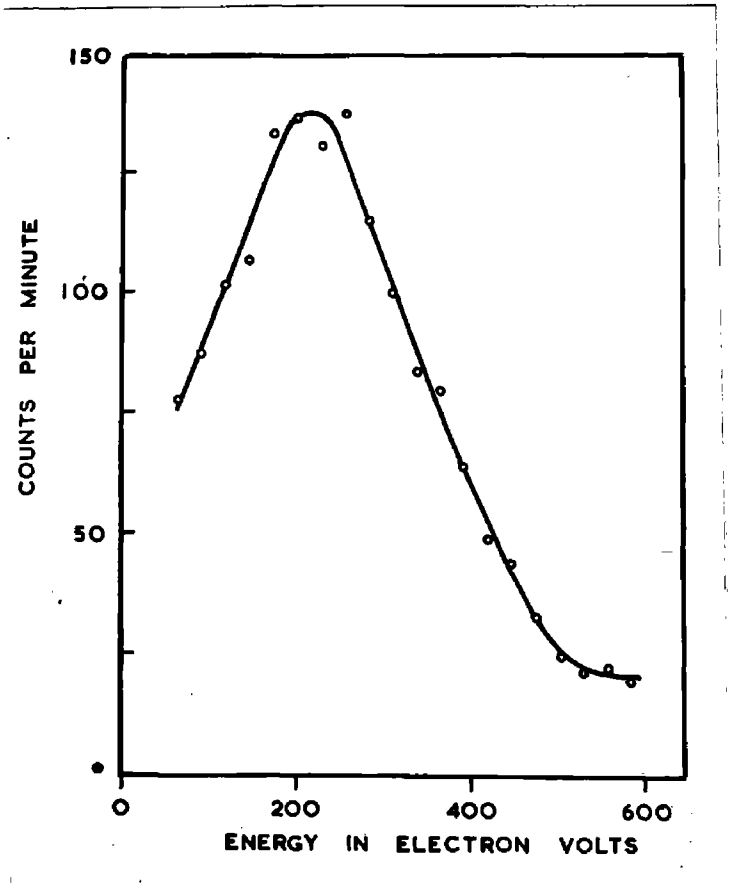


Fig. 3. L-peak of A^{37} taken at 450° C.

nature. At about 450°C, for example, the fall in resistance between the anode wire and the guard rings reduced the time constant of the input circuit to a value comparable with the time constant of the differentiating circuit used to shape the pulses in the amplifier, so the waveform of the output pulses altered and overshoots of the baseline became apparent after each pulse. It was noticed also that the anode current of the input valve in the head amplifier decreased at about the same temperature. This was found to be due to the appearance of an E.M.F. of 0.5 V between the anode wire and the guard rings, generated by a cell formed by the graphite and the tungsten wire, with the glass as electrolyte. In later experiments, condenser coupling was used at the input of the amplifier to prevent the grid bias of the input valve from changing.

The experience gained with these glass counters showed that there was no fundamental difficulty in operating proportional counters at temperatures up to 450°C, and that the main limitations were purely technical ones arising from the conductivity of the glass envelope. To extend the work to higher temperatures it was necessary to use another insulating material, and fused quartz was chosen. At room temperature fused quartz is one of the best insulators,

and its resistivity only falls to 10^7 ohm. cm at 700°C.

Experiments with Quartz Counters.

Although the high melting point of silica makes it a very suitable material for high-temperature apparatus, this property also makes its manipulation rather difficult, and it would be impracticable to construct a counter like that shown in Fig. 1 from fused quartz. It was therefore necessary to simplify the design considerably; and in the first quartz counter built the anode wire was made thick (0.02 cm) and was supported at one end only. The connections to the anode, the nickel cathode, and a metal guard tube, were brought out through a long quartz extension with Araldite seals kept cool by an air blast. It was found that this counter could be operated at temperatures higher than those reached with the glass counters, but there were a number of practical difficulties associated with the extended air-cooled seals, and with the non-uniform distribution of temperature in the counter. Further, a counter of this type is clearly unsuitable for measurements on vapourised sources, which would condense on the colder parts. A more satisfactory solution to the problem of designing a proportional counter simple enough for construction in quartz was found in the use of external cathodes.

Maze (1946) has shown that it is possible to build Geiger counters with cathodes which consist only of graphite coatings on the outside surface of the glass. The resistivity of soda glass is about 5×10^{11} ohm.cm at room temperature, which is low enough to permit the discharge current of a counter to pass through the envelope with negligible voltage drop. A similar type of construction is practicable for proportional counters also, as has been demonstrated by Cockroft and Valentine (1950).

The resistivity of fused quartz comes down to 5×10^{11} ohm.cm at a temperature of 200°C, so an external cathode could be used in a counter of quartz for operation at this temperature and above. The construction of such a counter might be further simplified by using conducting coatings on the outside of the quartz as guard electrodes also, so that the only electrical connection required to pass through the envelope would be that to the anode wire. To investigate these possibilities, several quartz counters were constructed, and the type of counter finally developed is shown in Fig. 4. Clear fused quartz, 1 mm thick, was used for the envelope, and the anode connection was made through

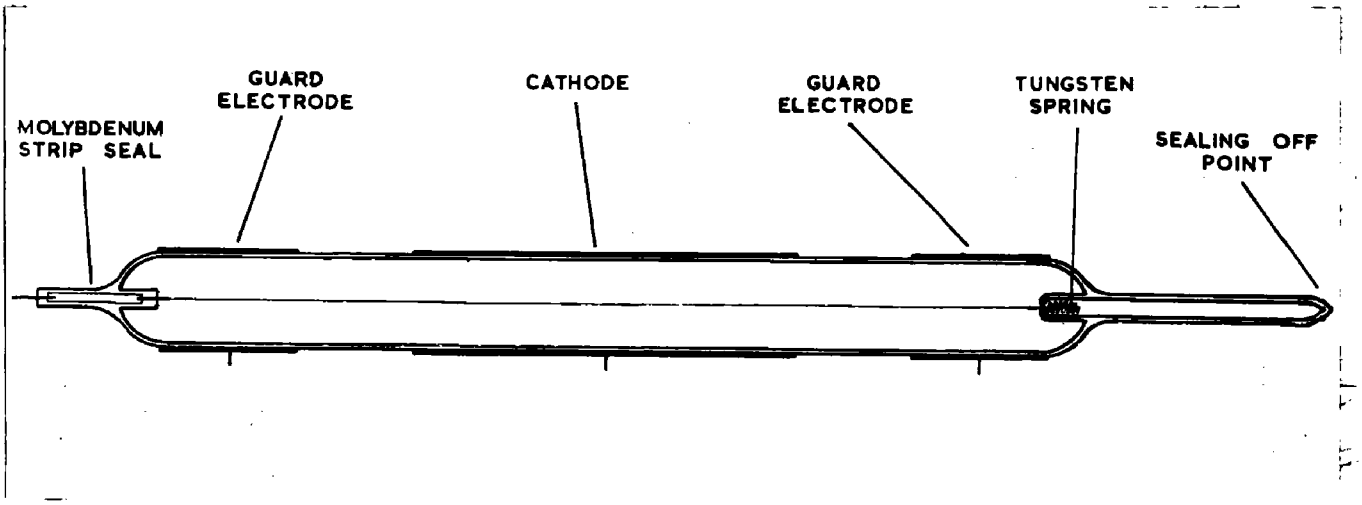


Fig. 4. Quartz proportional counter with external cathode.

The glass external-cathode counters are similar, apart from the type of anode seal used.

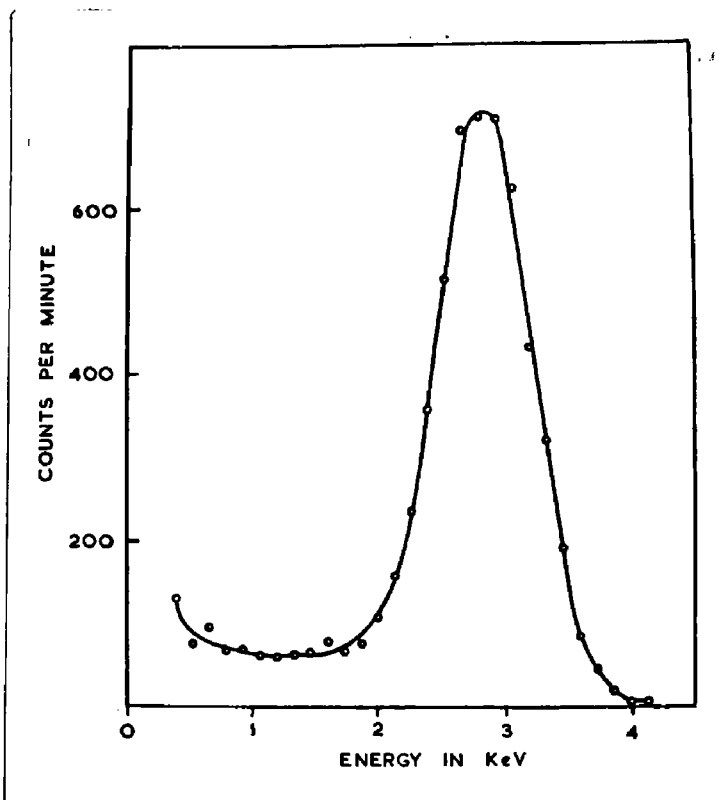


Fig. 5. K-peak of A^{37} taken at 810° C with the above counter.

a quartz-molybdenum strip seal. The anode and guard rings were formed by coatings of graphite on the outside of the quartz, covered by a layer of heat-resisting cement to prevent oxidation.

The experimental arrangement used to test the quartz counters was similar to that used with the glass counters, and the same filling gases were employed. It was found that quartz counters of the type illustrated could be operated at temperatures up to approximately 800°C, and a pulse spectrum of the A^{37} K-peak taken at 810°C is shown in Fig. 5. The leakage current through the quartz from the anode to the guard rings was more than 4 milliamps at this temperature, but this did not disturb the working of the counter and no electrolysis effects were noticed.

When this type of proportional counter is operated at room temperature, the resistance of the quartz envelope under the cathode coating is no longer negligible. The counter then behaves as if a condenser were connected in series with the cathode. During counting, charge collects on this condenser, which is formed in fact by the inner and outer surfaces of the quartz, and the effective voltage across the counter gradually falls.

The sensitive volume changes also. In spite of these effects, counting was found to be still possible at room temperature provided the quartz was discharged periodically by irradiating with X-rays, with the high voltage supply turned off. At temperatures above 180°C, however, the conductivity of the quartz is sufficient to prevent charges collecting, and the counter behaves as if it has a normal internal cathode.

No changes in energy resolution or counting rate were detected as the temperature was varied between 180°C and 770°C, but above the latter temperature small pulses corresponding to energies below about 80 eV were observed. These rapidly increased with further rise in temperature, and at 870°C the part of the A^{37} pulse spectrum below 1 keV was completely obscured by piling up of the small pulses. On altering the voltage applied to the counter it was found that the amplitude of the additional pulses remained proportional to the gas multiplication, as indicated by the A^{37} K-peak. This showed that the pulses were caused by electrons or ions appearing in the gas, and not by any spurious effect such as breakdown of insulation, or sparking. It appears, in fact, that the phenomenon is due to thermal emission

of electrons from the walls of the counter; and the thermionic work function estimated from these observations is close to the published value for silica of 4.8 eV. It may be remarked also that similar effects were noticed in the single-ended quartz counter mentioned before, when a nickel cathode coated with carbon was used. In this case the pulses appeared at a temperature between 500°C and 680°C, which is in agreement with the published work function for carbon of 4.4 eV.

Glass External-Cathode Counters.

Following the satisfactory performance given by the quartz external-cathode counters, some proportional counters were made of borosilicate glass to the same design. The experiments previously performed with counters like that in Fig. 1 were repeated with these external-cathode counters, and in this case there was no trouble with evolution of gases inside the counters at high temperatures. These counters could be operated at temperatures up to the softening point of the glass, 540°C, without much difficulty; though at the maximum temperature electrolysis of the glass made the working life rather short. This type of proportional counter seems well suited to measurements of vapourised sources

at temperatures in the range from 100°C to 300°C, and the very simple construction is an important practical advantage.

It might just be mentioned here that counters of similar design made from soda glass have been extensively employed by the writer in later work with radioactive gases at room temperature, when there has been some possibility of contaminating the apparatus. These counters are built so rapidly and cheaply that they can be regarded as expendable, and they were always used to check the properties of new gas sources before more accurate measurements were made in larger, and more expensive, metal counters.

Summary and Discussion.

The results obtained with the external-cathode quartz counters confirm the previous expectations, and demonstrate that it is quite practicable to operate proportional counters at temperatures as high as 800°C. The principal limit to the maximum working temperature is the fundamental one of thermionic emission of electrons, and the operating mechanism of the counter is not itself appreciably affected by increase in temperature.

The hard-glass proportional counters developed have proved satisfactory at temperatures up to 300°C, and from a practical point of view are very convenient and simple to use. Their working life is, however, limited when external guard rings are employed.

It may be worth mentioning that when the investigation on heated counters was begun, one of the isotopes whose study was in mind was iron 55. This is a pure electron-capture source of known decay energy, and a measurement of its L/K capture ratio would therefore be interesting. The K and L energies are 6.5 keV and 760 eV respectively, so a gas source technique is essential.

An attempt was made to measure Fe^{55} in the form of ferric chloride in a heated external-cathode glass counter. It was found, though, that the source became deposited on the walls of the counter by chemical reaction with water adsorbed on the glass, or perhaps diffusing through it. Another practical difficulty was the small size of the furnace available, which very much restricted the dimensions of the counter. With the low gas pressure which can be employed in a glass counter, this would have led to appreciable escape of X-rays, making the result dependent on fluorescence yield.

For these reasons, work on Fe^{55} with high temperature counters was discontinued, and attention was directed to the general problem of avoiding escape effects in gas source counters. However, in spite of this lack of immediate success in the particular case of Fe^{55}, it seems likely that the use of high temperature counters will permit considerable extension of the range of elements which can be measured by gas source proportional counter techniques.

Chapter 4.

A PROPORTIONAL COUNTER SYSTEM WITH REDUCED WALL EFFECTS AND ITS APPLICATION TO LOW LEVEL MEASUREMENTS.

The work on high temperature counters just described was one of several general investigations of proportional counter methods for measuring gas sources. Another such investigation was concerned with wall effects in gas source counters. This work originated in a study of problems arising from background counting rate in measurements of radioactive gases of very low specific activity, such as are required in radiocarbon dating. A method for reducing wall effects was developed, and the same technique proved valuable in later measurements of electron capture. In view of these subsequent applications, as well as the results obtained on low level counting itself, it may be worth while to give an account of this work here.

It may be useful to begin by reviewing briefly some general aspects of the measurement of materials of low specific activity.

General Considerations in Low Level Counting.

In the measurement of weak radioactive sources statistical fluctuations in the recorded counting rates due to the source and to the natural background of the counter are of considerable importance, and the accuracy and range of the measurements are, in fact, usually limited by statistical errors.

When finding the activity of a source two measurements have to be made: one without the source to determine the background counting rate, B, and one with the source present to find the total counting rate, S+B, due to the source plus the background. The statistical errors in the measurements depend on the duration of the counting. In practice the total time available for the complete measurement is usually limited, and it may be shown (for example, Browning, 1951) that when this is the case the minimum error in the result is obtained if the total time, t, is shared between the source and background measurements in the ratio $(S/B)^{\frac{1}{2}}$. If the source is very weak, so that $S \ll B$, then the standard error in each measurement is $(Bt/2)^{\frac{1}{2}}$, and the relative standard error in the final result obtained for S is $2(B/t)^{\frac{1}{2}}/S$. In many experiments the specific activity, A, of the material has

to be found rather than the total activity of a given specimen. In this case the relative error in the result obtained for A is

$$\frac{\sigma_A}{A} = \frac{2}{M.E.A} \cdot (B/t)^{\frac{1}{2}} \qquad \text{.....(11)}$$

where M is the mass of source material in the counter, and E is the overall detection efficiency for disintegrations occurring in this source material.

The minimum detectable specific activity may be taken as that for which the difference between the counting rates obtained with and without the source is just significant. A safe condition for this is $A = 3\sigma_A$, and with this definition the minimum detectable specific activity is

$$A_{min} = \frac{6}{M.E} \cdot (B/t)^{\frac{1}{2}} \qquad \text{.....(12)}$$

It may be seen from equations (11) and (12) that the accuracy of measurement of a low specific activity, and the lowest specific activity which can be detected, both depend upon the factor $M.E/B^{\frac{1}{2}}$. This factor should be large for a counter intended for measuring materials of low specific activity, so it is necessary to have a high counting efficiency over a large mass of source, along with a low background counting rate.

The same considerations hold in the application of low level counting for radiocarbon dating, an application which was particularly in mind during the present work. The maximum age which can be measured is set principally by the sensitivity of the counting arrangement. The archaeological importance of accurate dating of ancient specimens has therefore led to a considerable development of low level counting techniques. Much early work was done using Geiger counters with the source material spread over the inside of the cathode, but a higher detection efficiency can be obtained in a proportional counter with a gas compound of the sample as the filling, and such counters are now more widely used. (De Vries and Barendsen, 1953; Crathorn, 1953; and others). To enable a large mass of source material to be accommodated the pressure of the filling may be several atmospheres, or a gas with more than one atom of carbon per molecule, such as acetylene, may be used (see e.g. Barker, 1953).

The background counting rate of a normal proportional counter with a volume of 6 litres is of the order of 1000 counts/min., so a considerable reduction of background is necessary for measurements of natural carbon 14 (maximum specific activity 15 | disintegrations/min.gm.)

and other low level work. Standard methods of reducing the background counting rate include the use of lead or iron shielding to absorb gamma radiation from outside, and a ring of Geiger counters surrounding the main counter and connected in anticoincidence with it to prevent penetrating charged particles from being recorded. It may also be arranged that pulses corresponding to energies above the endpoint of the spectrum of the source are rejected. To give an indication of the effectiveness of these techniques, the results obtained by Fergusson (1955) may be quoted. With a proportional counter having a volume of 7.7 litres, the background counting rate could be brought down to 14 counts/min when the filling was inactive carbon dioxide at a pressure of 3 atmospheres. The counting rate obtained with carbon dioxide made from fresh natural carbon was 117 counts/min.

A further increase of sensitivity would still be very valuable for a number of applications, and particularly for carbon dating of specimens of great age. Some improvement might be achieved by operating the proportional counter at higher pressure, but there are serious practical difficulties in doing this and the quantity of the gas source which can be obtained is often

limited. A more satisfactory approach might be to reduce the background counting rate still further, and it was with this in mind that the present work was carried out.

Preliminary Analysis of Background of a Proportional Counter.

The background counting rate exhibited by a proportional counter in the absence of any intentional source arises principally from the detection of charged particles in the cosmic rays, and of gamma radiation from cosmic rays and from radioactive material in the surroundings. In a normal unshielded counter the components of the background due to charged and uncharged radiations are approximately equal. A protecting ring of Geiger counters connected in anticoincidence readily removes the charged component of the background counting rate, and it has been shown that even without Geiger counters at the end of the main counter the residual background due to penetrating charged particles is then negligible (see e.g. De Vries, 1956).

The component of the background due to uncharged radiation is more difficult to eliminate. The usual shielding of lead or iron about 8 inches thick is sufficient

to make the counting rate from external gamma rays of energies below a few MeV small, but fresh gamma rays originate in the shield itself. These may be produced as bremstrahlung from high energy charged particles, and may also come from radioactive contaminants in the shield material. There is not yet sufficient experimental data to enable the residual background to be analysed completely. It has been suggested (De Vries, 1956) that (n, γ) reactions due to neutrons in the cosmic radiation sometimes make an appreciable contribution.

In the present work attention was directed to the possibility of reducing the sensitivity of the counter to the gamma radiation present within the shielding. Previous experience at Glasgow had suggested that part, at least, of the background of a low level counter comes from the counter walls, a peak corresponding to the K X-ray energy of the wall material being apparent in the background pulse spectrum. Moreover, Ferguson (1955) has reported that the background counting rate of his proportional counter varied linearly with the pressure of the filling gas, from 10 counts/min at 1 atmosphere to 14 counts/min at 3 atmospheres. This might be

interpreted to indicate that 8 counts/min of the background are due to particles coming from the walls of the counter, and at one atmosphere 2 counts/min are due to charged particles produced in the gas by uncharged radiations. It therefore seemed possible that the background of a low level counter might be considerably reduced if the effects of the walls were eliminated. In order to check this possibility, and obtain further information on the origins of the background counting rate in a proportional counter, a special counter system in which wall effects are avoided was developed.

Counting Arrangement.

The counter eventually constructed consists effectively of one proportional counter surrounded by another coaxial proportional counter connected in anti-coincidence with it, with no intermediate wall. A diagram of the arrangement is given in Fig. 6. The main counter has a central anode wire, and a cathode made up of 36 thicker wires mounted in a circle. The region between this array of cathode wires and the outer case is turned into a separate counter system by 18 anode wires, interspaced by a further 18 cathode wires. Each of these anode wires, with the five cathode wires adjacent

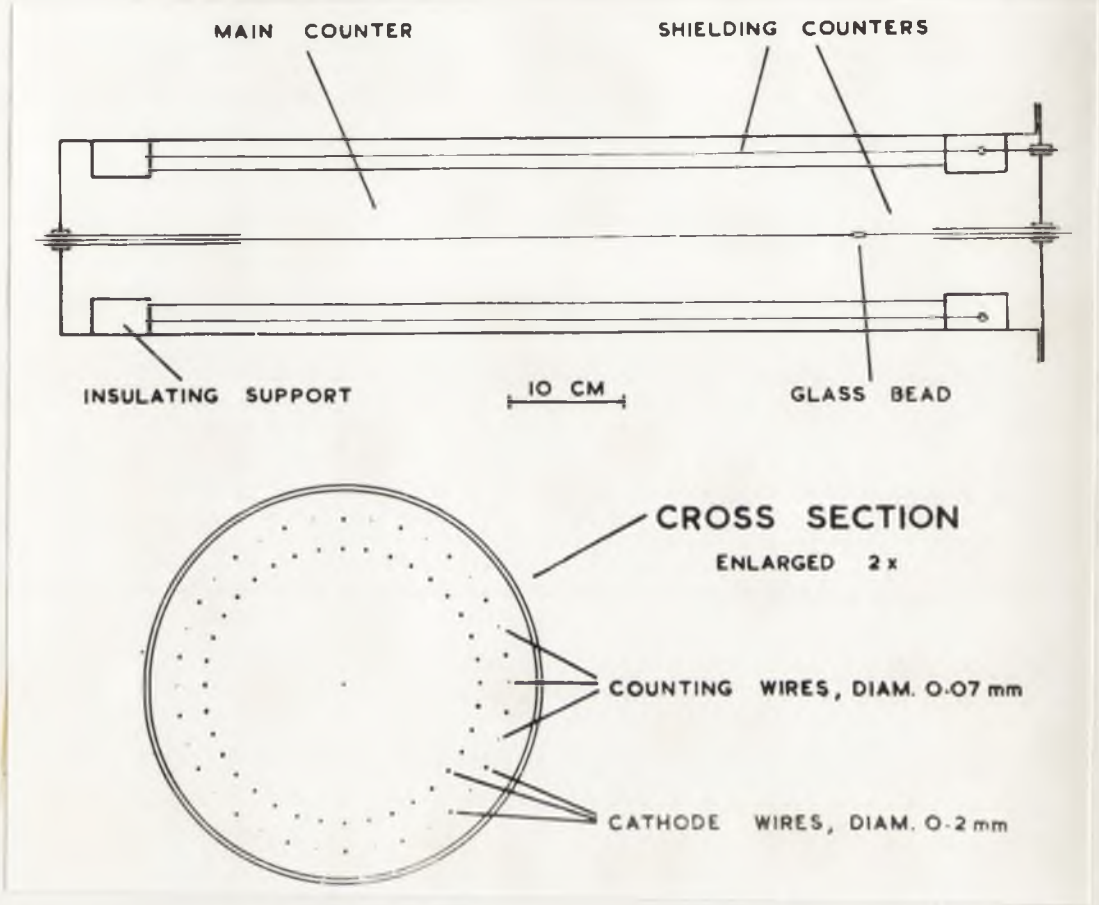


Fig. 6. General arrangement of the low-background counter.

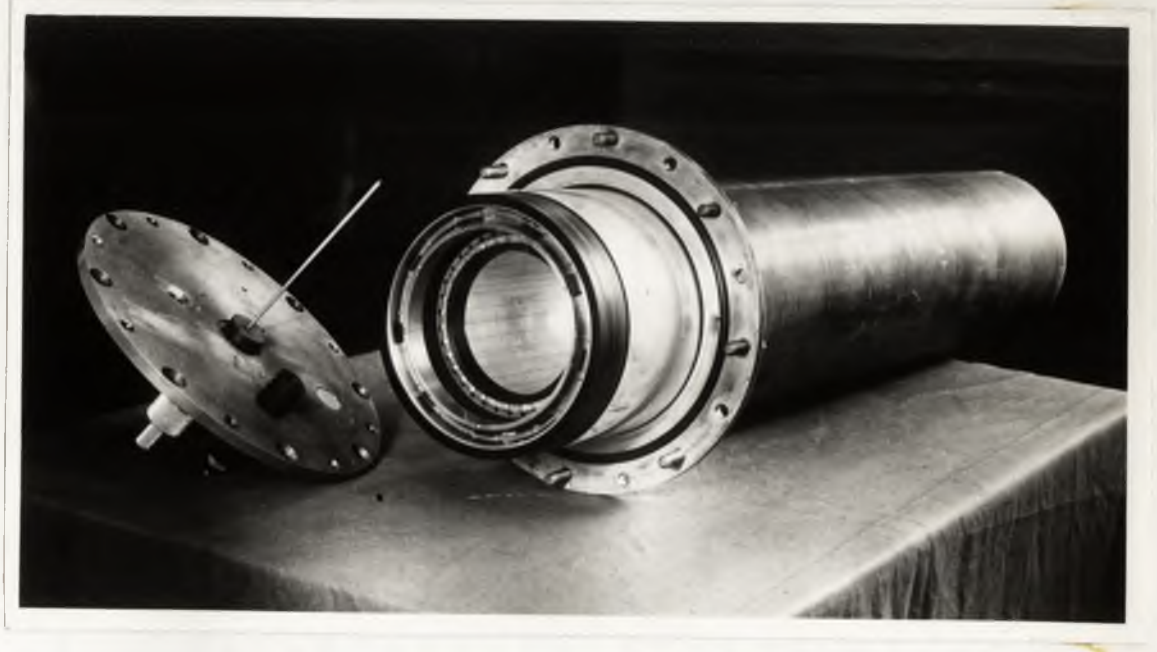


Fig. 7. The above counter, with the ring counting assembly partly withdrawn.

to it, and a portion of the case, forms an individual counter unit; and the 18 such units are connected together and operate as one multiwire proportional counter.

The electronic equipment is arranged so that a pulse from the central counter is recorded only if there is no simultaneous pulse from the ring counter system. Thus charged particles coming from the walls of the containing vessel are not registered, and the wall component of the background is effectively removed. There is still a small chance of particles coming from the ends of the vessel being detected in the central counter without passing into the ring system, and to reduce the probability for this the central anode wire is divided into two parts, connected by a glass bead at one end of the main counter. The shorter piece of wire is used as a separate end shielding counter, independent of the main central counter.

When a counter of this type is operated with the filling gas at a low pressure some loss of efficiency might result from beta particles passing out of the main sensitive volume into the ring system, and operating the anticoincidence circuit. To minimise this loss, the

counter is mounted inside a solenoid which can produce a magnetic field of 1500 gauss along the axis of the counter to curl up the tracks of the beta particles.

Operation and Characteristics of the Counter.

Both the ring and the central counter work as proportional counters so there is no appreciable coupling through photon production in the avalanche. It is necessary, however, to have a high gas multiplication in the ring counter since the energy spent by a fast electron passing through the ring is only of the order of 10 keV at one atmosphere. Moreover the total capacity of the 18 anode wires and their connections is 300 pF, so the output voltage pulse is smaller than would be given by a normal counter at the same gain. In the absence of any previous published work with high-gain multiwire counters there was some doubt whether the arrangement would operate satisfactorily, so every precaution was taken in the detail design to eliminate the possibility of difficulties from trivial sources such as insulation breakdown, or sparking. All the anode wires were fitted with guard tubes, and the anode wires of the outer counters were connected together by a steel ring enclosed by an annular screening box inside the pressure vessel. The screening box and the guard

tubes were kept at the same potential as the wires.

The wires in the ring counter are kept taut by individual springs, and to simplify the task of mounting the 72 wires the ring system was built as a unit which could be readily removed from the main vessel. The construction is illustrated in Fig. 7, where the ring system is shown partly withdrawn from the main counter. The cover for the circular metal box screening the connections of the ring counter has been removed, and the ring to which the anode wires are attached is visible.

The working of the counter was checked using argon-methane fillings and an external Ge^{71} calibration source, and both central and ring counters were found to operate satisfactorily. The gain of each of the 18 counting units in the ring system was found to be the same, as is shown by the calibration peak from the ring counter given in Fig. 8. This pulse spectrum was obtained when the whole ring system was irradiated by 9.2 keV X-rays entering through a window at one end of the counter.

The electronic equipment employed with this counter was planned with the later application to electron capture measurements in mind, and is slightly more complex

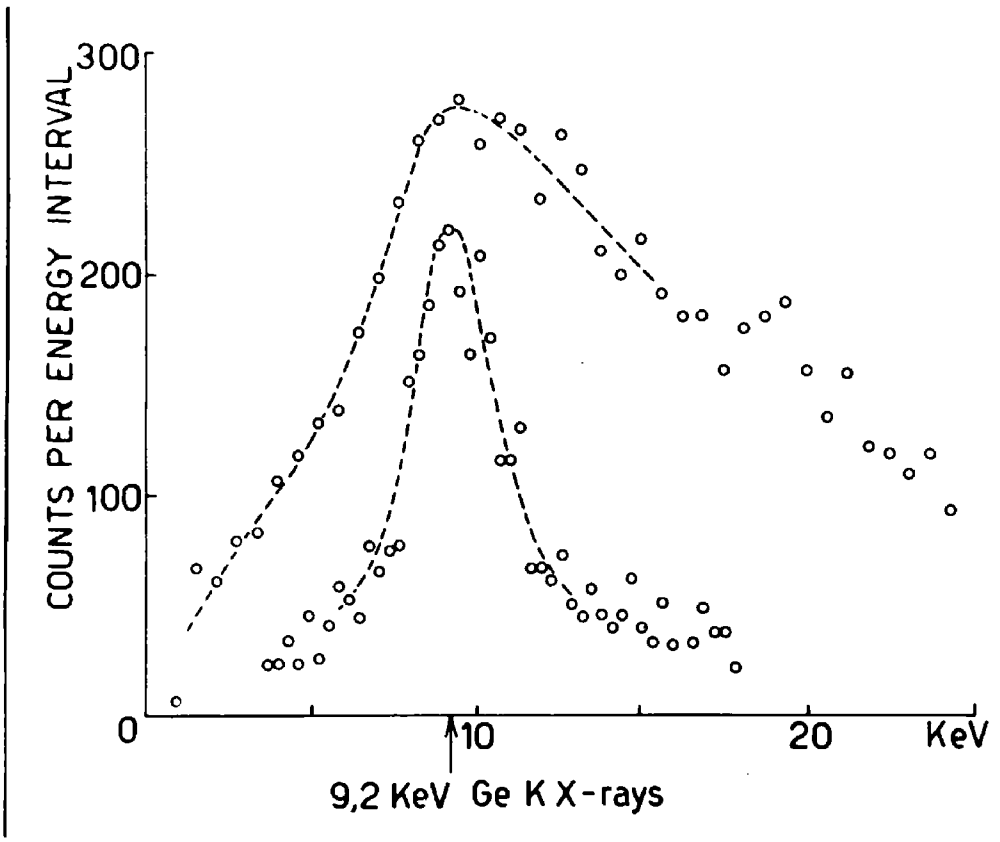


Fig. 8. Pulse spectra from the ring system of the low-background counter. The lower curve shows a calibration peak obtained when the counter is irradiated by X-rays from Ge^{71}. The upper curve (not on the same vertical scale) is the background due to cosmic rays, showing the scarcity of small pulses.

than would be required for low level counting alone. The requirements of this particular low level counter are themselves unusually stringent, since about 500 pulses are rejected by the anticoincidence gate for every one or two allowed to pass, while the signals controlling the gate include pulses with a wide range of amplitudes and lengths. Standard equipment available did not satisfy these requirements sufficiently and a special non-blocking amplifier and linear gate circuit had to be developed. These are described in an appendix to this thesis.

A schematic diagram of the whole apparatus is given in Fig. 9. Pulses from the central counter pass through a delay line to a gate controlled by the ring counter, and then to a pen recorder, scaler, and analyser as required. The upper limit to the energy region counted is defined by a discriminator connected to an early stage of the main amplifier, which closes the gate circuit for pulses corresponding to energies above the endpoint of the spectrum of the source being measured. To achieve the complete freedom from electrical interference which is essential in this work, a third

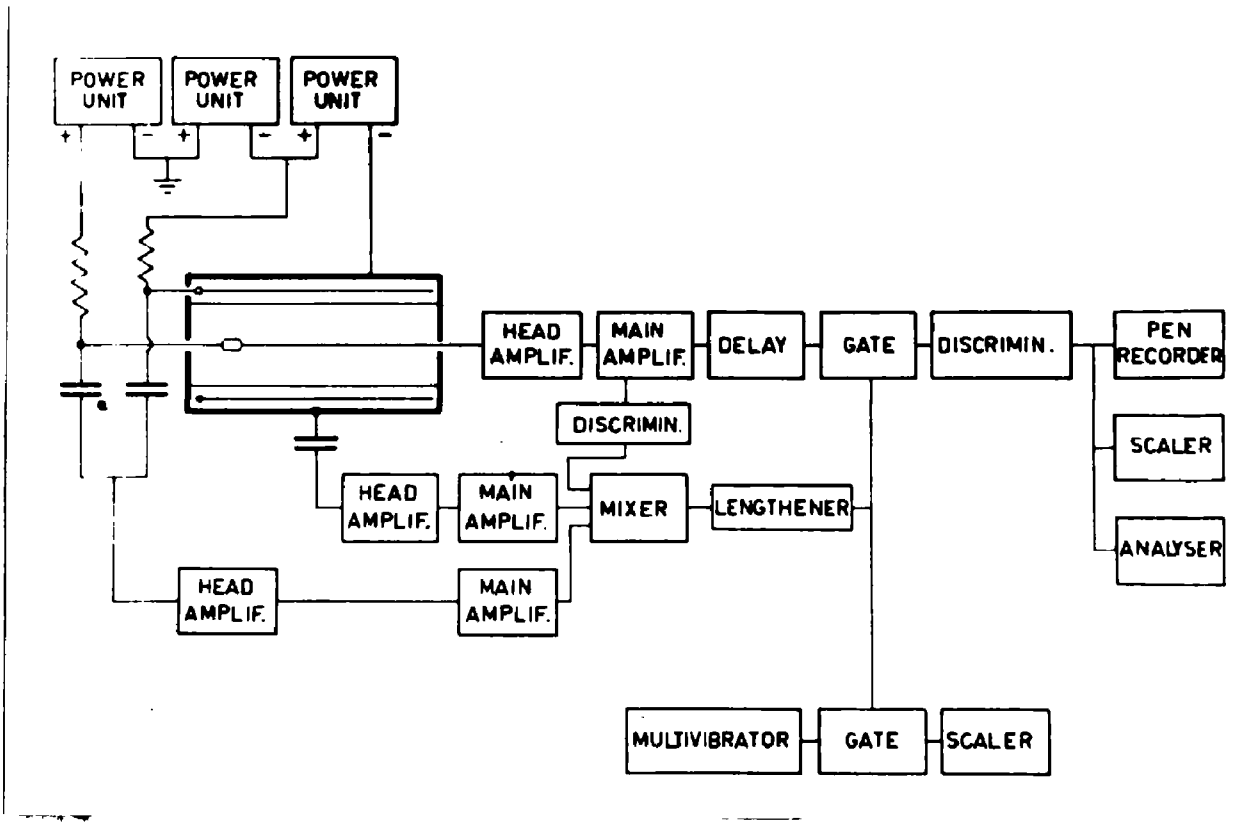


Fig. 9. Electronic arrangement for low-level counting.

amplifier is connected to the case of the counter and this closes the gate if interference or slight high-voltage breakdown occurs.

Background Measurements.

Preliminary experiments with the counter showed that a lower background counting rate could be obtained than with standard low level counting systems of the same sensitive volume, and a series of background measurements were carried out to make possible some analysis of the residual background.

For these measurements a normal argon-methane filling was used, the methane being at a partial pressure of 20 cm of mercury throughout. The solenoid in which the counter was mounted provided shielding equivalent to 3 inches of copper over the main part of the counter, and for further shielding 3 inches of lead around the solenoid, and 4 inches at the ends, was added. The upper limit discriminator was arranged so that pulses corresponding to energies higher than 150 keV, the endpoint of the beta spectrum of C^{14}, where not recorded; and the lower limit to the counting region was set at 4.0 and 10.0 keV in different runs.

The results of the measurements are given in Table 3.

TABLE 3.

BACKGROUND COUNTING RATES WITH ARGON-METHANE FILLING.

(Counts per minute)

Pressure (atmos.)	Lower energy limit (keV)	Without end counter		With end counter	
		Field off	Field on	Field off	Field on
1.0	4.0	2.4	3.7	2.3	3.0
	10.0	1.6	2.8	1.8	2.2
2.0	4.0	4.7	5.3	-	4.7
	10.0	3.3	4.2	-	3.3

The upper energy limit is 150 keV throughout.

The data obtained with a lower energy limit of 4.0 keV are the most interesting. In all cases application of the magnetic field causes the background counting rate to increase, due apparently to the increase in effective counting efficiency for Compton electrons produced in the gas, and for electrons entering the central counter from the ends. Application of the end shielding counter leads to an appreciable decrease of the background only when the magnetic field is present, and the reduction then is approximately 0.7 counts/min at pressures of both 1.0 and 2.0 atmospheres. This suggests that the component of the background counting rate which is removed by use of the end counter arises from low energy electrons which enter the main counter from the end, and are prevented from reaching the ring counter by the magnetic field. It might be expected that if a second end shielding counter were fitted at the unprotected end then a further reduction of the background counting rate by 0.7 counts/min would result.

With the magnetic field applied, increase of pressure from 1.0 to 2.0 atmospheres leads to an increase of background of almost 1.7 counts/min, both with and

without the end counter. This is evidently due to charged particles produced in the gas itself by gamma rays or neutrons, and if this part of the background counting rate is subtracted from the background found at a filling pressure of one atmosphere a residual pressure-independent counting rate of 1.3 counts/min is obtained. It would seem likely that 0.7 counts/min of this pressure-independent background comes from the unshielded end of the counter, and the remaining 0.6 counts/min may be due to charged particles originating in the cathode wires, or inefficiency in protection against penetrating charged particles by the ring system.

Some preliminary measurements with fillings of inactive carbon dioxide gave background counting rates approximately equal to those found with argon fillings at the same density, suggesting that Compton effect predominates in the production of electrons in the gas by gamma radiation. In a preliminary run with inactive methane slightly higher counting rates were obtained. This increase in background with a hydrogenous gas is similar to an effect reported by De Vries (1956) and, as he suggested, may arise from the detection of

recoil protons due to neutrons produced in the shielding by cosmic radiation.

The sensitive volume of the central counter is 5.5 litres, so the results indicate that the component of the background due to particles produced in the gas is of the order of 0.3 counts/min per litre-atmosphere for argon and carbon dioxide. This is close to the value which may be deduced from the data reported by Fergusson (1955). Of the remaining background, approximately 0.1 counts/min per litre of sensitive volume comes from the unshielded end of the counter, and not more than another 0.1 counts/min per litre is due to electrons from the cathode wires or inefficiency of the ring counter system.

Conclusions.

From the results obtained it may be concluded that the counting system developed provides an effective means of avoiding the major part of the wall component of the background in a proportional counter, and that even with a filling pressure of one atmosphere the main part of the residual background counting rate of the system is due to particles produced in the filling gas itself.

The low background obtained suggests that in conventional low level proportional counter arrangements the wall component of the background predominates. The reduction of background counting rate achieved in the present counter gives a considerable improvement in sensitivity, and in the case of carbon dating should enable the maximum age which can be measured to be extended by another half-life (5800 years) or more.

Further Applications.

With the counting system described above, charged particles which cross the boundaries of the main sensitive volume, entering or leaving, are not recorded. It is evident therefore that the wall effects that occur in spectrum measurements with normal proportional counters are modified in this arrangement.

In the Introduction (Chapter 2) it was shown how measurements of beta spectra, and of electron capture in the presence of beta emission, may be seriously affected by the presence of small pulses in the observed spectrum caused by beta particles which pass out of the counter before spending their whole energy. This type of wall effect is, in principle, eliminated in the present multiwire counting system, so more accurate

measurement of the low energy parts of beta spectra and of low energy electron capture radiations are possible. The high energy part of an observed beta spectrum might still be distorted, but this is usually of less interest in proportional counter studies since other instruments, such as magnetic spectrometers, are in any case more suitable in this region.

In measurements of electron capture where escape of X-rays from a normal proportional counter would be serious, the present type of counter can also be applied with advantage. If the ring counter system is made thick enough for efficient detection of the K X-rays, then L radiations accompanying K_{α} X-rays which escape from the central counter are not registered, and measurements of L-capture can be made without interference. It can also be arranged that K X-rays which enter from the part of the gas source in the ring counter compensate for those that leave the central counter, so that measurements of K-capture intensity can also be made more accurately than with a conventional counter of the same overall dimensions with the same filling gas. This will be discussed in more detail in Chapter 7, in connection with experiments on germanium 71.

This type of counting system thus enables the main wall effects which are important in low energy measurements, as well as those encountered in low level work, to be considerably reduced. These possibilities were realised early in the work described here, and construction of a second counter for electron capture measurements was begun as soon as it had been shown that the multiwire ring system was satisfactory. However, since much of the development of this experimental technique was carried out with the low level counter the present chapter has been devoted to it, and the application of these methods to electron capture measurements will be described more fully later.

It may be interesting to note that following the satisfactory results obtained by the writer in the original work on the two multiwire counters described in this thesis, a smaller counter of similar design was constructed by J. Scobie in this Department and was very successfully used in experiments on As^{76}, As^{79}, and C^{11} (Scobie, 1957; Scobie and Lewis, 1957).

EXPERIMENTAL STUDIES OF ELECTRON CAPTURE.

The two preceding chapters have dealt mainly with work related to the development of proportional counter techniques using gas sources. The remaining part of this thesis concerns measurements of orbital electron capture made with the aid of techniques of this kind.

Chapter 5.

DETECTION OF K-CAPTURE IN THE DECAY OF CHLORINE 36 AND MEASUREMENT OF THE K/β^- RATIO

Chlorine 36 lies between the two stable isobars A^{36} and S^{36}, and would therefore normally be expected to decay to both of them. The beta decay of Cl^{36} is well known, and the spectrum shape has been extensively studied by Wu and co-workers (see Wu, 1955; Longmire, Wu and Townes, 1949) since the decay is second forbidden unique and can give information about the interactions taking part. The endpoint energy is 714 keV (Feldman and Wu, 1952), while the half-life for beta emission is 3.08×10^5 years (Bartholomew et al, 1955). No gamma emission has been detected, the upper limit set to gamma intensity

being 5% of the beta intensity (Wu et al, 1949).

A search for positron decay was made by Wu et al, but no positrons were observed, and an upper limit of 10^{-4} was set for the fraction of Cl^{36} nuclei which decay by positron emission. Recent mass measurements (Wapstra, 1955) have, however, confirmed that K-capture to S^{36} should be possible. As this mode of decay might easily have escaped detection previously, a search for it was made in the present work.

It might perhaps be mentioned that the writer's attention was first drawn to the possibility of a K-capture decay in Cl^{36} by some unexpected results obtained in preliminary experiments on an activation technique for dating potassium minerals. This work has been described in a published paper (Moljk, Drever and Curran, 1955); but it will not be discussed in this thesis.

Preliminary Experiments.

In the absence of gamma emission in Cl^{36}, the atomic radiations of sulphur have to be detected to establish the existence of electron capture. The K absorption energy of sulphur is 2.47 keV, so a proportional counter is almost the only instrument suitable for this purpose, and a gas source has to be used.

Several gas compounds of chlorine were investigated in preliminary work before a completely suitable gas was found. Sources in the form of chlorine itself, and hydrogen chloride, rapidly became deposited on the walls of proportional counters made of brass, soda glass, and even polythene. (The properties of the polythene counter made for these tests are described in a published article (Moljk and Drever, 1957), and are interesting from more than one point of view). Chemical reaction with all these materials would not normally be expected, but in the case of the small quantities involved here deposition may have been the result of reaction with adsorbed water. Experiments with chloroform and carbon tetrachloride showed that these were more suitable gases chemically, but they exhibited considerable electron attachment which led to poor energy resolution. Eventually methyl chloride was found to be a satisfactory source gas in all respects, and this was used in the main experiments.

Source Preparation.

The Cl^{36} used was made by (n,γ) reaction in the high-flux pile at Chalk River, and was supplied in the form of hydrochloric acid of specific activity 120 μc/gm Cl,

with radioactive impurities removed. To prepare methyl chloride, chlorine gas was liberated from the acid by potassium permanganate, and was allowed to react with methane in diffused daylight for two hours. Hydrogen chloride produced in the photochlorination and any chlorine remaining were then removed by sodium bicarbonate and mercury, respectively, and the excess methane was left to act as carrier for the methyl chloride source.

To check the absence of radioactive impurities, the disintegration rate of a part of the source was observed for two weeks in a glass proportional counter. The possible impurities all have relatively short half-lives which would have become apparent during this period, but no changes in counting rate were in fact detected.

A counter made of brass was used in the main measurements, and the absence of appreciable chemical reaction, or adsorption, on the walls of this counter was confirmed by sharing the filling, containing some of the radioactive methyl chloride, with another vessel of equal volume. The counting rate due to the source was exactly halved in this process, showing that all the activity remained in gaseous form.

Measurements.

The proportional counter employed was of normal design, fitted with earth and field tubes, and had a volume of 6 litres. Argon was the main filling gas, with methane at a pressure of 10 cm Hg as quenching gas, together with a trace of the methyl chloride source. To cut down the rise of the pulse spectrum at low energies, which results from escape of beta particles, the total pressure of the filling was usually made equal to two atmospheres. The electronic equipment was standard, with a Hutchinson-Scarrott type of kicksorter.

A typical pulse height spectrum obtained with the source, in the neighbourhood of the K energy of sulphur, is shown in Fig. 10. A peak is evident, superimposed on the continuous pulse distribution due to the beta particles. Approximate energy calibration with the 9.2 keV X-rays from an external source of Ge^{71} indicated that the energy of the observed peak was close to 2.5 keV, but in order to check this more accurately some A^{37} was allowed to diffuse into the counter after the main spectrum measurements were complete. The 2.8 keV radiations due to K-capture in

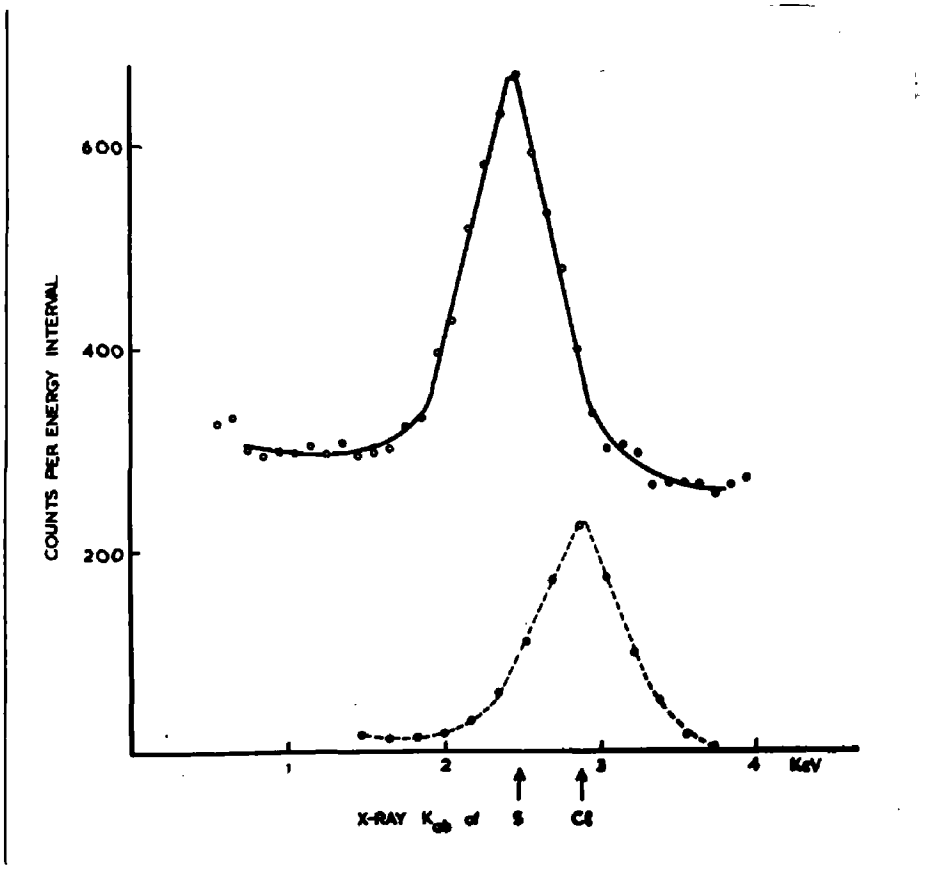


Fig. 10. K-capture in Cl^{36}. The upper curve is the pulse spectrum given by a Cl^{36} gas source. The lower curve is an A^{37} calibration peak, drawn with the vertical scale reduced by a factor of 100.

A^{37} were then observed, the spectrum obtained being shown dotted in Fig. 10, drawn with the vertical scale reduced by a factor of 100. Comparison of the mean pulse heights in the two peaks shows that the peak due to Cl^{36} has an energy equal to that of the K absorption energy of sulphur, and significantly different from the $K\alpha$ X-ray energies of chlorine, 2.62 keV, and argon, 2.96 keV. It may be noted that by this comparison technique the mean energy of the unknown peak can be determined with an error appreciably less than the half-width of the peak itself.

With the experimental arrangement used here a peak corresponding to the K absorption energy of sulphur can only be produced by K-capture in chlorine. In any other process by which X-rays might be produced from atoms in the sensitive volume of the counter, such as excitation of the argon by beta particles or the production of excited daughter atoms in beta decay, the X-rays are effectively simultaneous with beta pulses and would not be recorded as a peak, while even without this their energies would be significantly different from that of the observed peak. It may therefore be concluded that

the peak found is definite evidence for the existence of K-capture in the decay of Cl^{36}.

Branching Ratio.

The ratio of the decay constants for K-capture and beta emission was determined by comparing the counting rate in the observed K peak with the total counting rate due to K-capture plus beta decay as recorded by a scaler. Some small corrections have, however, to be applied in evaluating the branching ratio from the observed counting rates.

A certain number of the X-rays emitted by the source escape from the sensitive volume of the counter through the side wall and are not recorded. Rigorous calculation of the probability of this type of escape is difficult in general, but in the present case when escape is small approximations are sufficient. Such calculations give, for a counter diameter of 10 cm and an absorption half-thickness of 0.54 cm, a probability of escape of 0.050. Escape of X-rays at the ends of the counter is not included here, since the dead-space at each end of the sensitive volume is long compared with the half-thickness for absorption, and X-rays which enter the sensitive volume from the source at the ends compensate

for those that leave. The Auger effect in sulphur is so large that only 5.6% of the K-capture decays result in the emission of K X-rays, and the total correction for escape of X-rays therefore becomes 0.3% of the observed K-capture intensity.

In the case of the beta decay intensity, a correction for beta particles that enter the sensitive volume from the ends of the counter is necessary. The dead space at each end of the counter has a length of 6.5 cm, which is short compared with the average range of the beta particles, so absorption can be neglected in a first approximation and the number of beta particles entering found by considering solid angles alone. The sensitive volume of the counter has a length of 58 cm, and the required correction to the recorded beta intensity was calculated to be 4.5%.

Several separate measurements of methyl chloride sources were made, and the mean value obtained for the ratio of the counting rate in the K peak to the counting rate in the whole spectrum was 0.0165. After applying the corrections above, this gives a value for the branching ratio K/β^- of 0.017 ± 0.001.

Decay Scheme.

It was felt desirable to carry out a more detailed search for gamma emission than had been made before, since the upper limit set by Wu et al is not low enough to rule out the possibility that the K-capture transition now detected might lead to an excited state of S^{36}.

A scintillation counter with a NaI(Tl) crystal, 5 cm in diameter and 5 cm long, was used to look for gamma rays in the energy region above 20 keV. One microcurie of the Cl^{36} source, in the form of hydrochloric acid enclosed in a polythene beta absorber, was mounted in front of the crystal and the resulting pulse spectrum was analysed. No peaks were detected above the continuous spectrum due to bremstrahlung. The energy resolution of the counter, checked with gamma rays from RaD, Cs^{137}, and Na^{22}, was such that any gamma radiation from Cl^{36} with intensity greater than 1% of the total bremstrahlung intensity would have been detected, so the experiment sets an upper limit to gamma emission of 10^{-4} of the intensity of beta emission.

There is still a possibility that a strongly internally converted gamma ray, of low energy, might have

escaped detection by the scintillation counter. A further search of the energy region below 50 keV was therefore made with the proportional counter, using a methyl chloride source. No peaks other than that due to K-capture were observed. The counting rate of pulses due to the beta particles set a limit to the sensitivity, and a peak above 15 keV corresponding to an intensity of 1% of the beta intensity might not have been detected. It should be noted, though, that K-capture leading to the emission of a gamma ray in this region would not in any case have been recorded in the K-peak previously observed.

It may be concluded that the observed K-capture transition goes directly to the ground state of S^{36}. The decay scheme of Cl^{36} is therefore as shown in Fig. 11.

It may be interesting to remark that on the completion of this work Cl^{36} was the lightest nucleus in which the atomic radiations due to K-capture had been directly and quantitatively observed. It is unlikely that the K-capture could have been detected by any method other than that used here, and this relatively simple experiment demonstrates the power of the gas source technique.

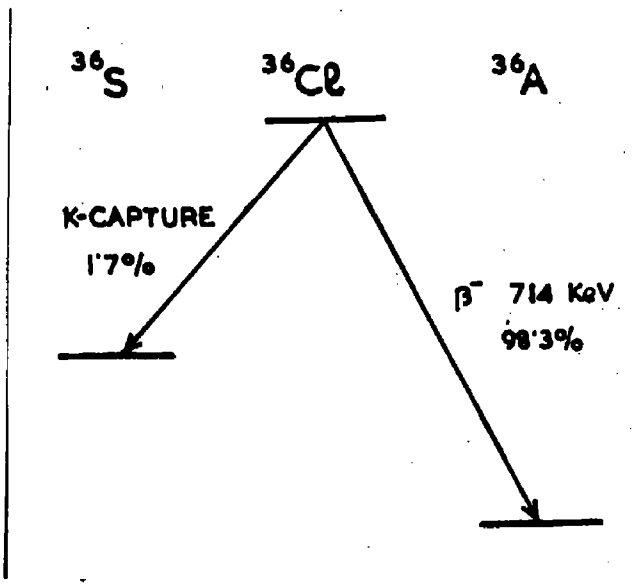


Fig. 11. Decay scheme indicated for Cl^{36}.

Chapter 6.

THE K/β^+ RATIO IN THE DECAY OF FLUORINE 18.

Introduction.

It was shown in the first chapter of this thesis how an experimental value for the K/β^+ ratio in an allowed decay can give some information about the combination of interactions in beta decay, in particular about the Fierz interference term b defined in equation (6). It was pointed out, though, that of the few nuclei for which accurate measurements of K/β^+ ratios have been made, none is known with certainty to decay by an allowed transition. A more reliable check on the value of b would be given by a direct measurement on a simple well-known allowed decay. The decay of fluorine 18 is very suitable for this purpose, so this nucleus was chosen for investigation in the present work.

Fluorine 18 is a positron emitter, having a simple spectrum with a maximum energy of 649 keV and an allowed shape (Ruby and Richardson, 1951). The half-life is 112 minutes (Blaser et al, 1949), and with log fT = 3.6, the transition is allowed and favoured. The spin of F^{18} is 1, as indicated by isotopic spin selection rules

(Moszkowski and Peaslee, 1954), and the decay to oxygen 18, of measured spin 0, involves a Gamow-Teller interaction. No gamma emission has been detected, and Knox (1948) set an upper limit to gamma intensity of 10% of the positron intensity.

Electron capture had not been observed in F^{18} before the present work, since the K radiations of oxygen emitted after K-capture have a maximum energy of only 530 eV and would not have been detectable in the counters used. Auger electrons are, moreover, emitted instead of K X-rays in 99.5% of the K-capture events.

The experimental technique employed in the present measurements was similar to that used for the work on Cl^{36}, in that the capture ratio was found from the pulse spectrum from a proportional counter containing a gas compound of the source; although differences of half-life and of source preparation led to some changes in detail.

Source Preparation.

Fluorine 18 cannot be made by (n,γ) reaction from a stable isotope, so the use of (γ,n) and (n,2n) reactions were considered. After estimating the specific activities obtainable with the fluxes of gamma rays and

neutrons available in this Department it was decided to prepare the source by (γ,n) reaction on fluorine using the beam from the 340 MeV electron synchrotron. The intensity of the bremsstrahlung spectrum in the neighbourhood of 20 MeV, where the (γ,n) cross-section is near its maximum, is just sufficient to give the required specific activity.

Following the satisfactory results obtained with methyl chloride in the work on Cl^{36}, preliminary experiments on F^{18} were made with the source in the form of methyl fluoride. It was found, though, that methyl fluoride prepared directly from irradiated potassium fluoride did not have sufficient activity; while when a Szilard-Chalmers process involving irradiation of a solution of potassium fluoride in acetic acid was used an appreciable C^{11} activity was obtained in addition to that from F^{18}. On account of these difficulties with methyl fluoride some experiments were made with boron trifluoride, and this gas was found more convenient.

The radioactive boron trifluoride was prepared by heating boric oxide and sulphuric acid, with calcium fluoride which had been irradiated for 4 hours close to

the vacuum chamber of the synchrotron. Water vapour and other condensible impurities were removed by cooling the gas to the temperature of solid carbon dioxide. Some A^{37} was detected in sources made in the preliminary work, and in the main experiments this was removed by freezing the boron trifluoride with liquid air and pumping off the argon. Boron trifluoride prepared by the method employed here may contain traces of silicon tetrafluoride, which has a high electron attachment coefficient, but the shortness of the half-life of the source made the procedure required to remove this gas impractible. The presence of some silicon tetrafluoride in the boron trifluoride used set a limit to the amount of source which could be put into the counter.

Measurements.

The counting equipment employed was the same as that used in the earlier work on Cl^{36}. However the lower energy of the K-capture Auger electrons and X-rays from F^{18} made it necessary to reduce further the effects of escaping beta particles, so the counter was operated at a pressure of three atmospheres. The difficulties associated with overloading of the electronic apparatus

became much more serious in this case, but it was found possible by modifying the amplifier (as outlined in the Appendix) to improve its overload performance sufficiently for the present experiment.

Several separate measurements of the K/β^+ ratio were made, and in each run the boron trifluoride was prepared immediately after the irradiation of the calcium fluoride, and spectrum measurements were begun as rapidly as possible.

In all cases a peak was observed in the pulse height spectrum, near 500 eV, as is shown in Fig. 12. The counting rate in the peak was found to decay with a half-life close to that of F^{18}. As in the work on Cl^{36}, X-rays from Ge^{71} were used for energy calibration during the main measurements, while an accurate determination of the energy of the observed peak was made by introducing A^{37} into the counter during one of the runs. Comparison of the pulse height in the F^{18} peak with the pulse height in the peak due to A^{37}, measured after the activity of the fluorine had become negligible, confirmed that the observed peak corresponds to the K absorption energy of oxygen, and thus represents K-capture in F^{18}.

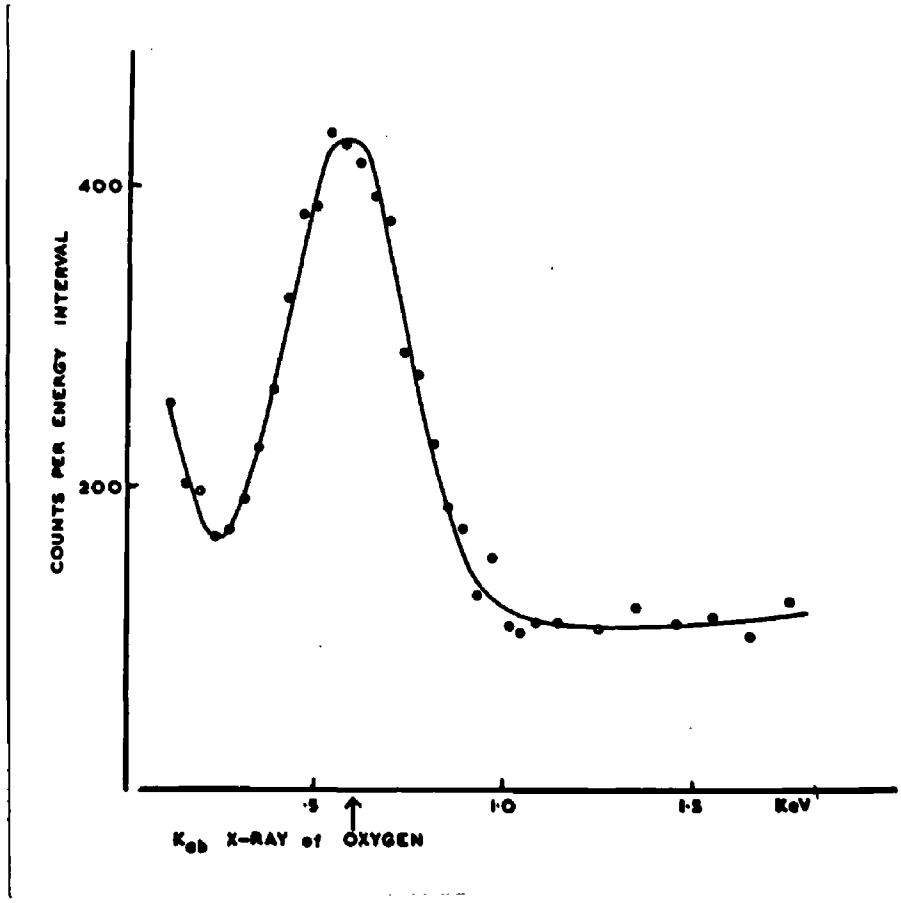


Fig. 12. Pulse spectrum from F^{18} in a proportional counter, showing K-capture peak.

To determine precisely the number of pulses due to K-capture in a spectrum like that in Fig. 12 it is necessary to establish accurately the position of the boundary between the continuous pulse distribution and the peak superimposed on it. The steep rise in the recorded positron pulse spectrum at low energies might introduce some uncertainty here, particularly as the K-capture peak itself is relatively broad due to the statistical fluctuations in ionisation, and possibly also to some electron attachment in the source and the effects of the relatively high counting rate of the large positron pulses. To increase the accuracy of the measurement a comparison method was therefore employed. When the A^{37} source used for energy calibration was in the counter, the pulse spectra in the neighbourhood of the argon K-capture peak (2.8 keV) and L-capture peak (230 eV) were measured. In the effective absence of a positron background the shapes of these two peaks could be accurately determined. The extent of the peak due to K-capture in F^{18} was then found by comparison with the relative areas and shapes of the A^{37} peaks.

Simultaneously with the measurements of K-capture, the total counting rates due to positron decay plus

K-capture were determined. The decay of the total activity was followed for several half-lives and was found to correspond closely with the reported half-life for F^{18} of 112 minutes. Towards the end of the measurements the presence of a weak long-lived activity became evident. The intensity of this impurity activity was, however, less than 5% of that of the initial F^{18} activity; so it merely led to a slight increase in the effective background of the counter, which could be determined from the decay curve. Allowance was made for this long-lived activity when evaluating the F^{18} counting rates.

Previous experience with Cl^{36}, and also with tritium, had shown that gas sources may become deposited on the walls of a counter even when chemical reaction would not be expected, so a separate run was made to check this possibility here. After following the decay of a boron trifluoride source for 1 hour, the gas was pumped out of the counter, and the residual counting rate of the counter was measured. This experiment showed that in fact 23% of the F^{18} activity had been deposited on the brass wall. The decay curves had already shown that no loss of source occurred during the measurements, and it

may be assumed that all the deposition took place immediately on filling. Experiments with Cl^{36} showed that a source on the wall of a proportional counter does not contribute appreciably to a low energy K-peak, so in the present work the deposited source would only have the effect of making the positron counting rate higher than that from the gas itself. The observed positron counting rate was therefore corrected accordingly when calculating the K/β^+ ratio. As in the experiment on Cl^{36}, a further correction had to be made for particles which enter the sensitive volume from the ends of the counter, and in this case the correction was found to be 4.6%.

The half-thickness for absorption of K X-rays of oxygen in the filling gas is less than 1 mm, and the fluorescence yield is only 0.5%, so escape of X-rays from the sensitive volume of the counter is completely negligible.

From the results obtained in several separate runs, the mean value found for the ratio of the decay constants for K-capture and positron emission in F^{18} is

$$\lambda_K/\lambda_+ = 0.030 \pm 0.002$$

where the probable error assigned includes estimated

systematic errors in the experiment, as well as statistical errors in counting.

Decay Scheme.

Although previous work on F^{18} did not indicate the presence of any gamma emission, the upper limit which was set then is not low enough to show whether the K-capture now detected goes entirely to the ground state of O^{18}, or partly to an excited state. A further search for gamma emission therefore had to be made before a direct comparison of the K/β^+ ratio found here with the theoretical value could be justified.

A scintillation spectrometer with a NaI(Tl) crystal was used to investigate the energy region above 30 keV. The presence of the positron annihilation radiation makes direct detection of any other low-intensity gamma radiation difficult, so a comparison between pulse spectra given by F^{18} and C^{11} sources was carried out to increase the sensitivity of the experiment. Carbon 11 is a positron emitter with a half-life of 20 minutes, in which no gamma emission has been observed (Siegbahn and Peterson, 1946). The source used here was made by (γ,n) reaction on spectrographically pure graphite, using the gamma beam from the synchrotron.

The pulse spectra given by the radiations of F^{18}, in the form of calcium fluoride powder, and of C^{11}, as graphite, where measured under identical conditions; each source being spread in a thin layer over a perspex beta absorber covering the face of the NaI(Tl) crystal. The two spectra obtained are shown in Fig. 13, normalised to the same total number of counts. The points found with F^{18} are indicated by dots, and those due to C^{11} by circles. It will be seen that the spectra lie closely together, indicating absence of nuclear gamma emission by either source. From these results it is estimated that no gamma ray above 30 keV with intensity greater than 0.5% of the positron intensity is emitted by F^{18} or C^{11}.

The region of the spectrum below 50 keV was examined with the proportional counter using a boron trifluoride source, but no peak apart from that due to the K radiations already observed was detected.

On the basis of these results it may be taken that the decay of F^{18} is simple, both positron emission and K-capture going directly to the ground state of O^{18}.

Comparison with Theory, and Conclusions.

Theoretical values for K/β^+ ratios have been tabulated by Zweifel (1954); but they do not extend to

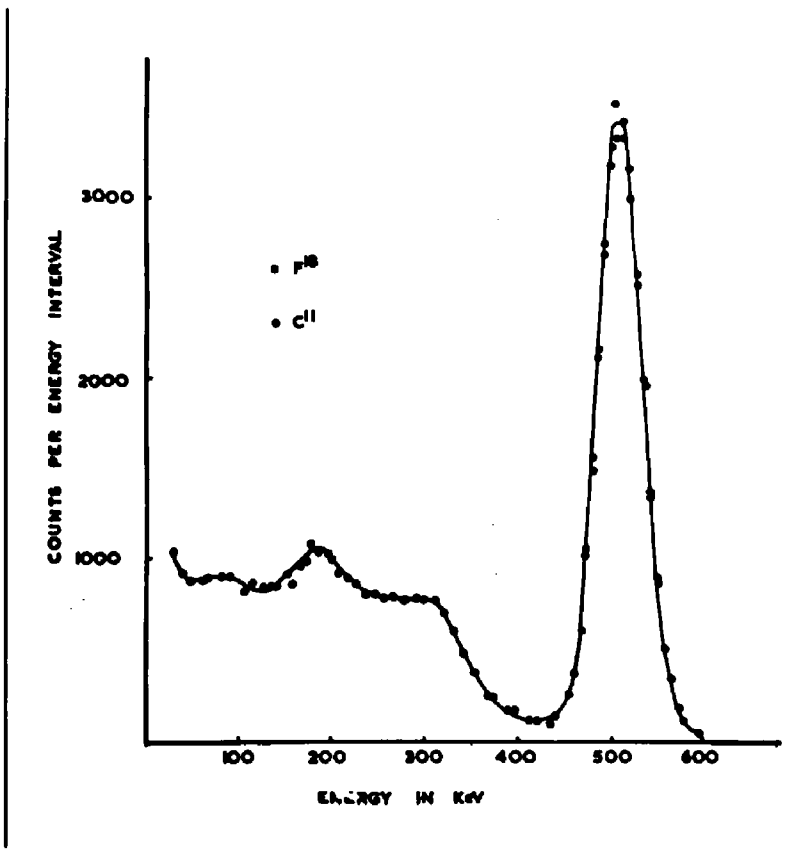


Fig. 13. Pulse spectra given by a scintillation counter with gamma radiations from sources of F^{18} and C^{11}. The F^{18} spectrum is indicated by dots; the C^{11} one by circles.

nuclei as light as F^{18}, so here the theoretical result is best found using equations (7) or (8) of Chapter 1. As the interference term b is known to be small it may be neglected in a first calculation, the K/β^+ ratio then being given by equation (8). The ratio evaluated in this way, using the tabulated Fermi functions of Rose (1955) and the screening corrections of Reitz (1950), is found to be $R_0 = 0.0295$.

The measured K/β^+ ratio, 0.030 ± 0.002, is in close agreement with this theoretical value for b=0. However, to obtain an upper limit on b from the results it is necessary to consider the more complete expression for the K/β^+ ratio given in equation (7). The relation between the branching ratio R for a finite value of b and the ratio R_0 found if b=0 may be written in the form

$$\frac{R}{R_0} = \frac{1 + b}{1 - b.\langle W^{-1} \rangle} \qquad \text{.....(11)}$$

where $\langle W^{-1} \rangle$ is the mean value of 1/W taken over the positron spectrum corresponding to b=0. Graphical integration of 1/W over the theoretical spectrum of F^{18} gives a value for $\langle W^{-1} \rangle$ of 0.69. On substituting for R the measured K/β^+ ratio, the value found for the Fierz

interference term is

$$b = 0.008 \pm 0.04,$$

where errors in the theoretical ratio R_0 are assumed to be small compared with the experimental errors.

This result on the Fierz interference term in a pure Gamow-Teller decay is very similar to that deduced by Sherr and Miller (1954) from their measurement of the K/β^+ ratio in Na^{22}. The present experiment is, however, free from the slight uncertainty about the nature of the beta decay which was present in the work on Na^{22}. The result thus gives satisfactory confirmation, with improved accuracy, of the conclusions about the smallness of the Fierz interference drawn from the shapes of allowed beta spectra.

The expression (6) previously given for the Fierz term b may be written, for a pure Gamow-Teller decay, in the form

$$b = 2.(C_T/C_A + C_A/C_T)^{-1} .$$

If $b \ll 1$, then it follows that either $C_A/C_T = b/2$ or $C_T/C_A = b/2$. The experiments on the recoil of He^6 show that $C_A < C_T$, so that the former alternative has to be taken as the correct one. On the basis of these data,

then, the result found in the present work may be taken to correspond to a ratio of axial vector to tensor interaction of

$$C_A/C_T = 0.004 \pm 0.02$$

It has already been pointed out in the Introduction that the recent discovery that parity is not conserved in beta decay makes this final conclusion less certain now than it was when the experiment on F^{18} was actually carried out. In the most general case now, when no relationships are assumed between the parity-conserving interactions and the parity-nonconserving ones, and the coupling constants may be either real or complex, the interpretation of the result for b can only be written

$$\frac{Re\,[C_A \cdot C_T^* + C_A' \cdot (C_T')^*]}{|C_A|^2 + |C_A'|^2 + |C_T|^2 + |C_T'|^2} = 0.004 \pm 0.02 \ .$$

This becomes the same as the previous result if the two-component neutrino theory is valid, and if the interactions are invariant to time reversal.

Note on the Experiments Described in the Last Two Chapters.

The results obtained in the work on Cl^{36} and F^{18} show the accuracy and usefulness of a simple gas source proportional counter technique. However, in both experiments the low-energy background due to escaping beta particles was troublesome, and in fact this caused the failure of an attempt to measure L-capture in Cl^{36}. These difficulties could have been avoided if a multiwire counter like that described in Chapter 4 had been available at the time, but the experiments on Cl^{36} and F^{18} were carried out before the completion of the work on the low-background counter which first demonstrated the practicability of this arrangement. The multiwire counter system was, however, employed with advantage in the experiments on Ge^{71} to be described in the following chapter.

Chapter 7.

THE L/K CAPTURE RATIO IN THE DECAY OF GERMANIUM 71.

Introduction.

The experimental difficulties of measuring L-capture and of determining transition energy in pure electron capture decays have very much restricted the checking of theoretical calculations of L/K capture ratios. In the few cases for which direct comparison between theory and experiment is possible considerable discrepancies are apparent, as was shown in the first chapter of this thesis. Attempts have been made to modify the theory, but these have not explained all the experimental data.

Some of the differences between measured and calculated L/K capture ratios may arise from errors in the decay schemes assumed, but a serious discrepancy in the case of germanium 71 cannot be explained in this way. The reported L/K ratio is between two and three times larger than the expected value; while the decay involves only a pure electron capture transition which has been well studied, and whose energy is accurately known. A remeasurement of the L/K capture ratio for this particular decay is therefore very desirable.

Germanium 71 decays by electron capture, with a half-life of 12.5 days (Bisi, Germagnoli, et al, 1955), directly to the ground state of gallium 71. The absence of nuclear gamma radiation, and of beta emission, makes Ge^{71} a convenient source in which to study the emission of internal bremsstrahlung in electron capture, and several investigations of the bremsstrahlung spectrum from this isotope have been carried out. Saraf et al (1953) and Saraf (1954) measured the spectrum shape, and found good agreement with theory from the endpoint down to 100 keV. Langevin (1954a) reported a low energy component in the bremsstrahlung spectrum, with a longer half-life than the main part; but subsequent work by Bisi, Germagnoli, et al (1955) has shown that this was due to the presence of a radioactive contaminant in the source. The spectrum obtained by the latter workers agreed closely with theory down to 50 keV when well-purified sources were used. From the measured endpoint of the bremsstrahlung spectrum the transition energy is found to be 231 keV, and the decay is allowed with log fT = 4.6. The theoretical L/K capture ratio corresponding to this decay energy, calculated with the electron wave functions of Brysk and Rose, is 0.106.

A measurement of the L/K capture ratio has been made by Langevin (1954b and 1956), using a proportional counter with the Ge^{71} in the form of germanium hydride gas. The main filling gas was propane, so that most of the K X-rays were not detected, and the L/K capture ratio was deduced from the peaks due to L and K Auger electrons. A value of 0.30 was obtained for the capture ratio. However, this result depends strongly on the K fluorescence yield of gallium, which Langevin took as 0.45, the value given by a semi-empirical formula of Burhop (1952). If, instead, the fluorescence yield of 0.50 indicated by the work of Broyles, Thomas and Haynes (1953) is used then the L/K capture ratio found becomes 0.19. Recently Laberrique-Frolow, Radvanyi and Langevin (1956) have suggested that a better value of the fluorescence yield of gallium is 0.477, which with the same experimental data on Ge^{71} gives an L/K capture ratio of 0.25. It is evident that the dependence on fluorescence yield of Langevin's measurement of Ge^{71} makes the result obtained for the capture ratio rather uncertain.

The present measurement has been carried out by a method which does not require any accurate knowledge

of fluorescence yield. The L/K capture ratio is, however, still obtained by a proportional counter technique using a gas source. It may therefore be useful at this point to discuss some general aspects of such L/K measurements.

General Considerations.

In a measurement of an electron capture source of low or medium atomic number it is usually not difficult to arrange that the dimensions of the counter and the density of the filling gas are such that effectively all the Auger electrons emitted by the source in the sensitive volume are detected. If this is the case, and if a fraction P of the K X-rays emitted escape detection, then the K-peak recorded corresponds to a fraction (1 - PF) of the K-capture events which occur in the sensitive volume, F being the mean K fluorescence yield of the daughter atom produced in the decay. Usually escape of L X-rays from the counter is negligible, so that all L-capture events taking place are recorded in the observed L-peak. This peak also contains pulses corresponding to a fraction PFk of the K-capture events, where k is the fraction of vacancies in the K shell of the daughter atom which lead to the emission of K_α X-rays. Thus the L/K capture

ratio, R, is given in terms of the counting rates N_L and N_K in the observed L and K peaks by the equation

$$R = \frac{N_L}{N_K} \left(1 - PF \right) - PFk \qquad \text{.....(12)}$$

In general, therefore, the L/K capture ratio obtained is a function of the fluorescence yield F.

In practice the value of P, the probability of escape of K X-rays, can be determined most accurately when P is either close to unity or close to zero, and it is preferable to arrange the measurement so that either one or other of these conditions is satisfied. The former alternative is experimentally the simpler, and is the one chosen by Langevin, but accurate knowledge of the fluorescence yield is then required. It may be shown by differentiating eqn. (12) and substituting the values of F, k, and the theoretical value for R, appropriate to Ge^{71} that any error in fluorescence yield leads to a relative error in the L/K capture ratio which is 9 times greater. It may be noted also that any error in the experimental determination of N_L/N_K causes a relative error in the final result 4.5 times larger.

These difficulties do not arise if it is arranged that all the X-rays are efficiently detected, so that P

is close to zero. Then the L/K capture ratio is given directly by N_L/N_K, and the fluorescence yield only has to be introduced as a small correction for any residual escape of X-rays. This is the technique which has been used for A^{37}, but with nuclei of higher atomic number the reduced absorption of the X-rays makes the method more difficult to apply. In the case of Ge^{71} the $K\alpha$ X-rays, of energy 9.2 keV, have a half-thickness for absorption in argon at one atmosphere of 5.0 cm, and a very large high-pressure counter would be required. To reduce the total escape correction in the final result to 5%, for example, a counter filled with argon to a pressure of 6 atmospheres would have to be at least 1 metre in diameter. A slightly smaller counter might suffice if krypton or xenon were used as filling gases; but the very high cost of these gases, along with the large quantities required, makes this hardly feasible.

In the present work a multiwire counter system was employed, and this enabled an argon-filled counter of practicable dimensions to be used.

Principle of the Measurement.

The multiwire proportional counter used is similar in general design to the one built for the work

on low level counting described in Chapter 4, and a cross-section is given in Fig. 14. For this application the central counter was of relatively small diameter, and the surrounding ring counter, made up of five counting units, was of greater thickness. Pulses from the central counter, in anticoincidence with pulses from the ring counter, were analysed; and the L/K capture ratio was determined from the counting rates in the L and K peaks in the pulse height distribution.

In analysing the method, it should be noted first that with the filling used, at a pressure of 6 atmospheres, the probability that a K X-ray originating in the central counter will pass through the ring counter without being absorbed is only about 0.1%. In this preliminary discussion this probability will be taken to be zero, and it will be assumed that every X-ray which leaves the central counter is detected in the ring counter.

Any K-capture event which takes place in the central counter giving rise to a K Auger electron, or a K X-ray which is detected in this counter, is recorded in the measured K peak. However, a considerable fraction

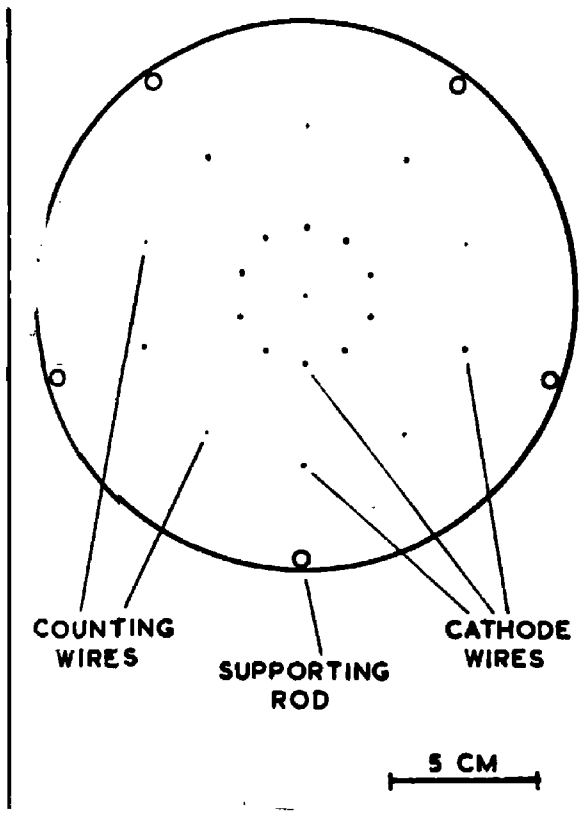


Fig. 14. Cross-section of counter for L/K ratio.

(about 30%) of the K X-rays originating in the central counter are not absorbed there, and these are detected instead in the surrounding ring counter. This operates the anticoincidence gating circuit, and prevents any L X-rays or Auger electrons produced simultaneously from being registered. Incorrect recording of an L-capture event when a K_{α} X-ray escapes, which is the major escape effect in a normal counter, is thus avoided.

Although the K-capture processes in the central counter giving K X-rays which escape from that counter are prevented from registering in the L peak, these events are not recorded in the K-peak either. However, some of the K X-rays produced by K-capture in the ring counter pass into the central counter and are detected there, and this compensates exactly for the K X-rays which escape from the central counter into the ring*. For this compensation to be complete it is necessary that the anticoincidence circuit is not triggered by pulses from the ring counter due to L X-rays emitted

* In a radioactive medium of uniform activity and uniform absorption coefficient, the mean rate of absorption in any region A of photons originating in any other region B is equal to the mean rate of absorption in B of photons originating in A. This is a particular case of a general theorem by Mayneord (1945).

simultaneously with K X-rays in the ring, so the circuits controlling the gate are biased to be insensitive to pulses corresponding to less than the K energy. With this arrangement the total counting rate in the recorded K peak, the two components of which are not resolved, is equal to the rate of occurrence of K-capture in the central counter.

In the case of the L peak, any escape of L X-rays from the central counter is similarly compensated for by L X-rays entering from the ring, and the counting rate in the L peak is equal to the rate of occurrence of L-capture in the central counter. It may be concluded that, provided every K X-ray leaving the central counter is detected in the ring, the L/K capture ratio is given directly by the ratio of the counting rates in the observe^{d} L and K peaks.

In practice any K X-rays from the central counter which penetrate the gas in the ring without being absorbed, or which are absorbed in the wires which make up the ring counter, escape detection. This leads to an increase in the counting rate of the L peak, and a decrease in that of the K peak since there are no

corresponding X-rays entering the central counter. These consequences are similar to the consequences of X-ray escape in a normal, single, proportional counter; but in the multiwire arrangement they can be made considerably smaller than in a normal counter of the same overall diameter, with the same filling. From this point of view the ring system can be regarded as effectively confining the measurement to a region of the source near the axis of the counter, where the probability of X-ray escape is a minimum. In the case of the present work on Ge^{71}, the probability of a K X-ray escaping through the ring without being absorbed was less by a factor of a hundred than the probability of X-ray escape in a normal counter of diameter equal to the outside diameter of the ring.

With this large reduction in the X-ray wall effect, escape of X-rays at the ends of the counter, which is usually considerably smaller, becomes relatively more important. $K\alpha$ X-rays passing out through the unguarded ends of the central counter may lead to the recording of spurious L events, though the K-peak itself is not affected since K X-rays enter also from the dead space at the ends. The probability of X-ray escape at

the ends is almost as large in this multiwire counter as in a normal counter of the same length, and in the present experiment accounted for the major correction required to the observed data. However, even when all the possibilities of escape of X-rays are taken into account, the total correction required amounted to less than 5% of the final result.

Design of the Counter.

In this application of a multiwire counter the dimensions of the centre and ring systems are determined almost entirely by considerations of X-ray escape, since the maximum range of a K Auger electron or a photoelectron is almost negligible (less than 0.5 mm in the present experiment). The major factor is the probability of escape of K X-rays from the central counter right through the ring counter. In a system of given outside diameter this probability decreases as the diameter of the central counter is reduced, though as the relative source counting rate in the central counter falls also, a compromise has to be made. In the present experiment the diameters of the ring and central counters were made 18 cm and 4.5 cm respectively.

The source counting rate in the ring counter was then about 15 times the counting rate in the central counter, and a measurement of the Ge^{71} L peak to a statistical accuracy of 1% could be made in 1 hour, without an excessive counting rate in the ring.

A diagram of the counter is given in Fig. 15, and its construction is illustrated by the photographs in Figs. 16 to 19.

The general arrangement and construction of the counter are similar to that of the preceding low level counter, but with the thicker ring system, five counting units in the ring were sufficient. As before, the ring system was designed so that it could be removed from the outer vessel for wiring. In this case the 20 wires in the ring are mounted in a cage-like frame consisting of brass end plates connected by five supporting rods arranged in positions, near the outer wall, which give minimum field distortion at the anode wires. The ring system is shown removed from the pressure vessel in Fig. 18 where the guard tubes fitted to the anode wires can be seen. At one end of the counter the guard tubes are connected to the annular screening box which encloses the anode connections. In Fig. 19 the circular

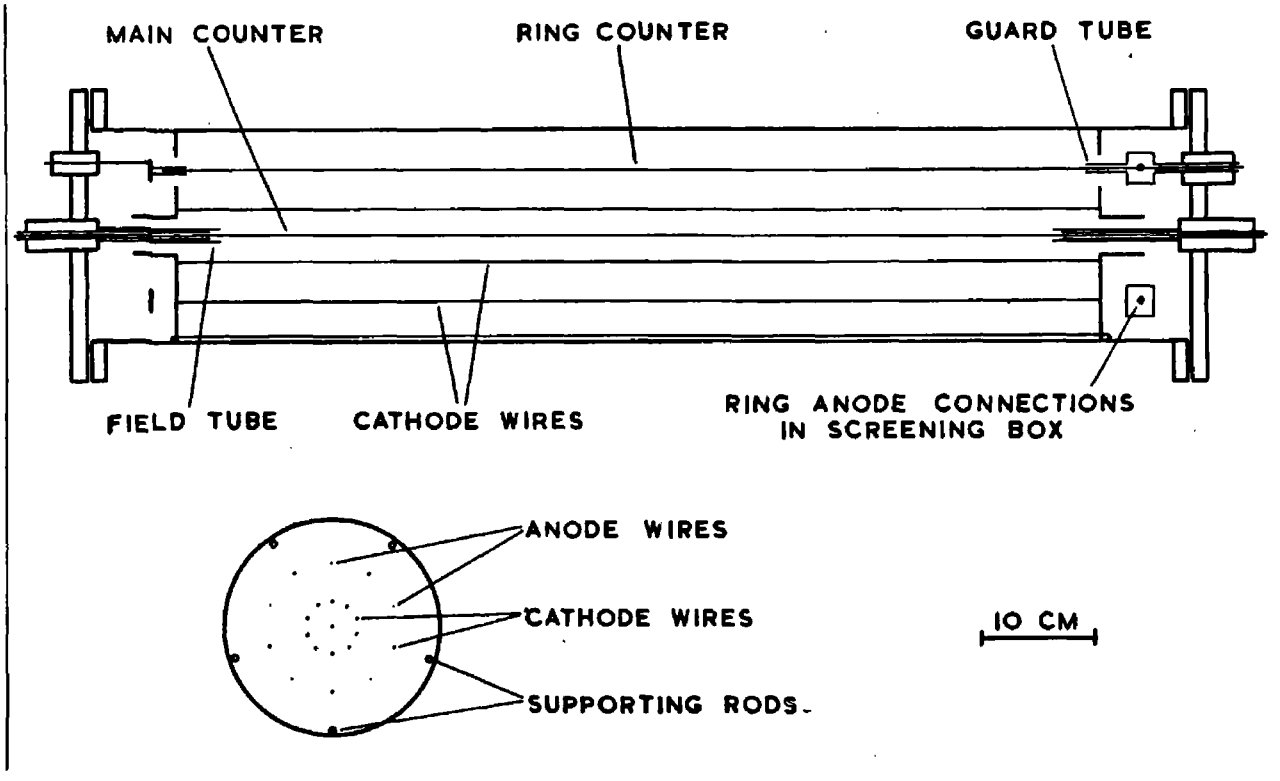


Fig. 15. General arrangement of counter for L/K ratio.

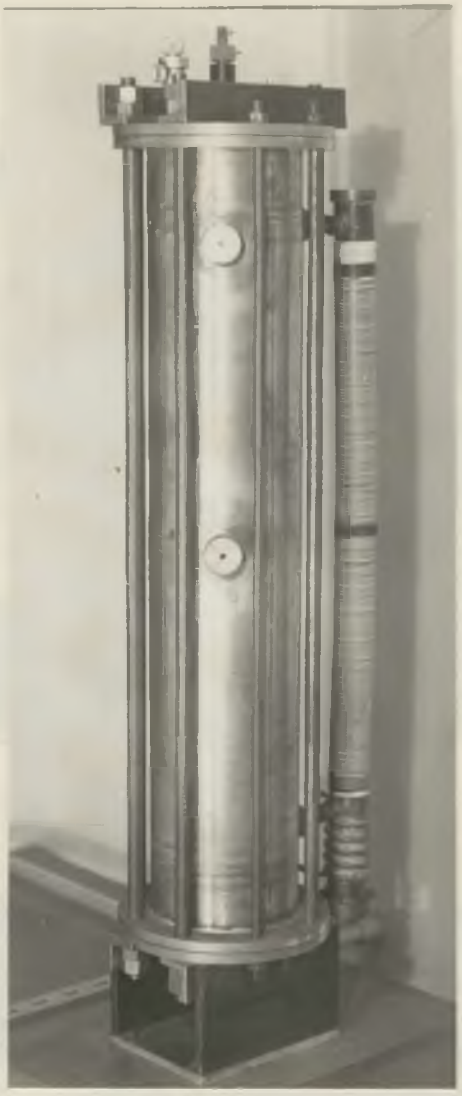
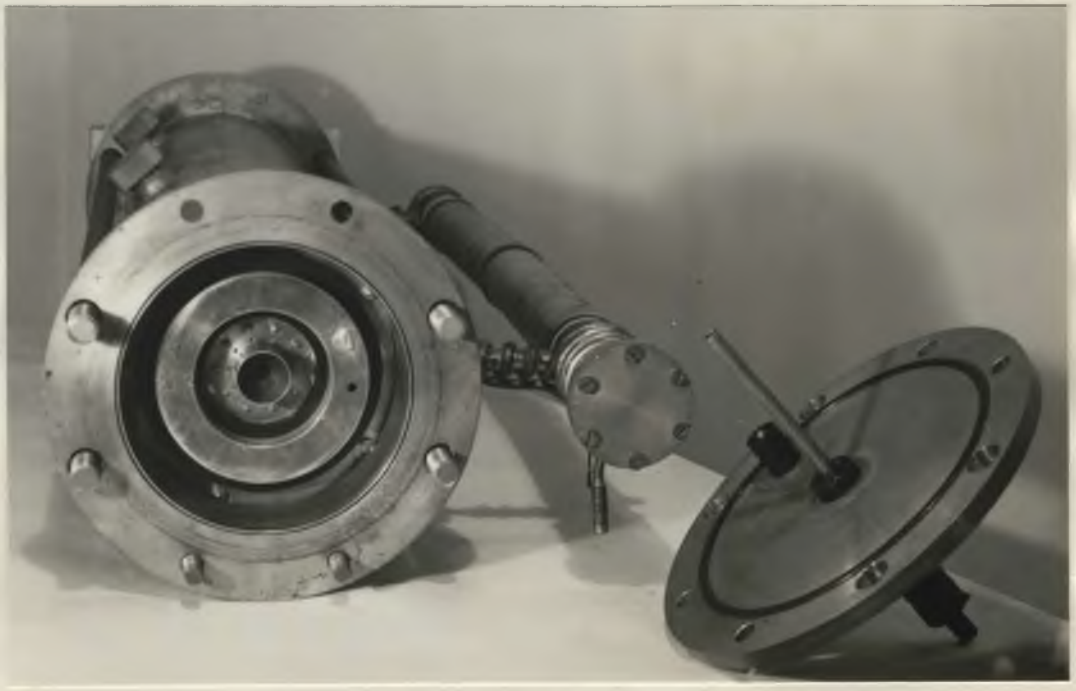


Fig. 16.

L/K counter:
general view.

Fig. 17.
(below)

L/K counter:
End removed to show screening of ring connections.



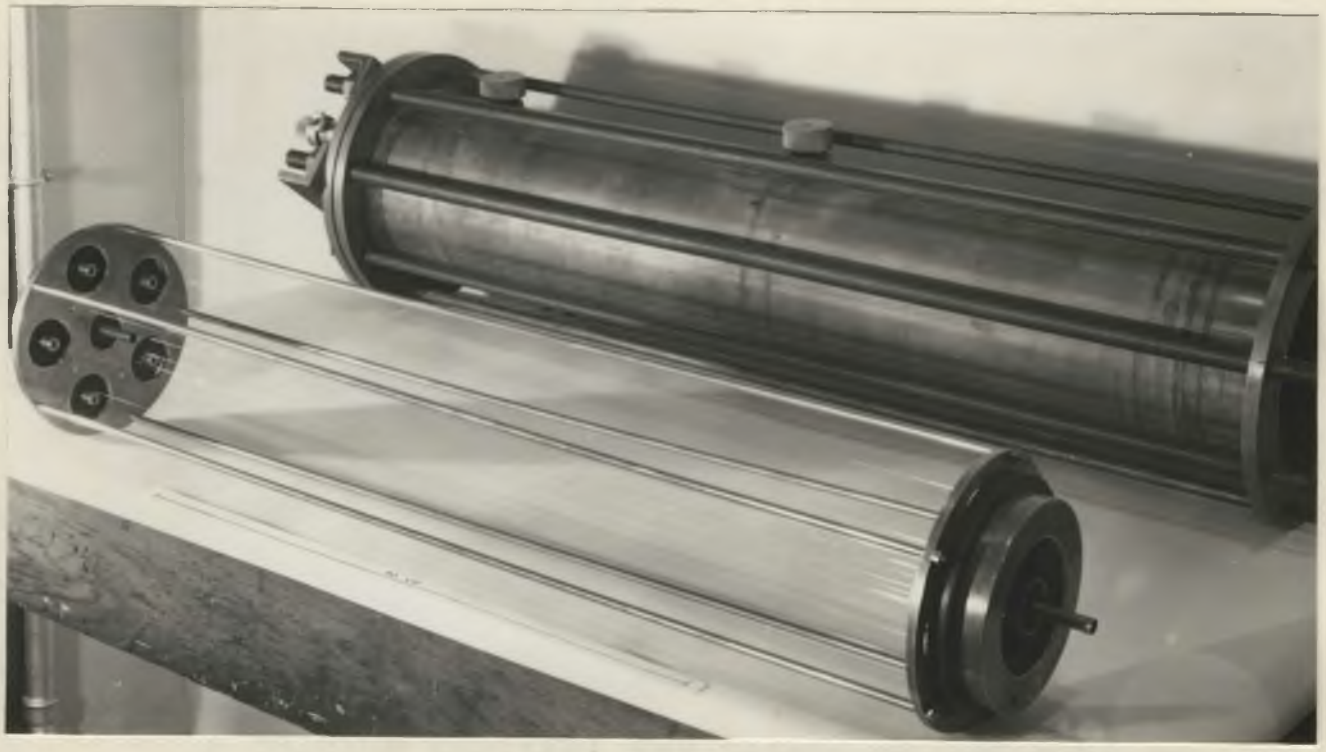


Fig. 18. L/K counter: ring assembly removed from case.

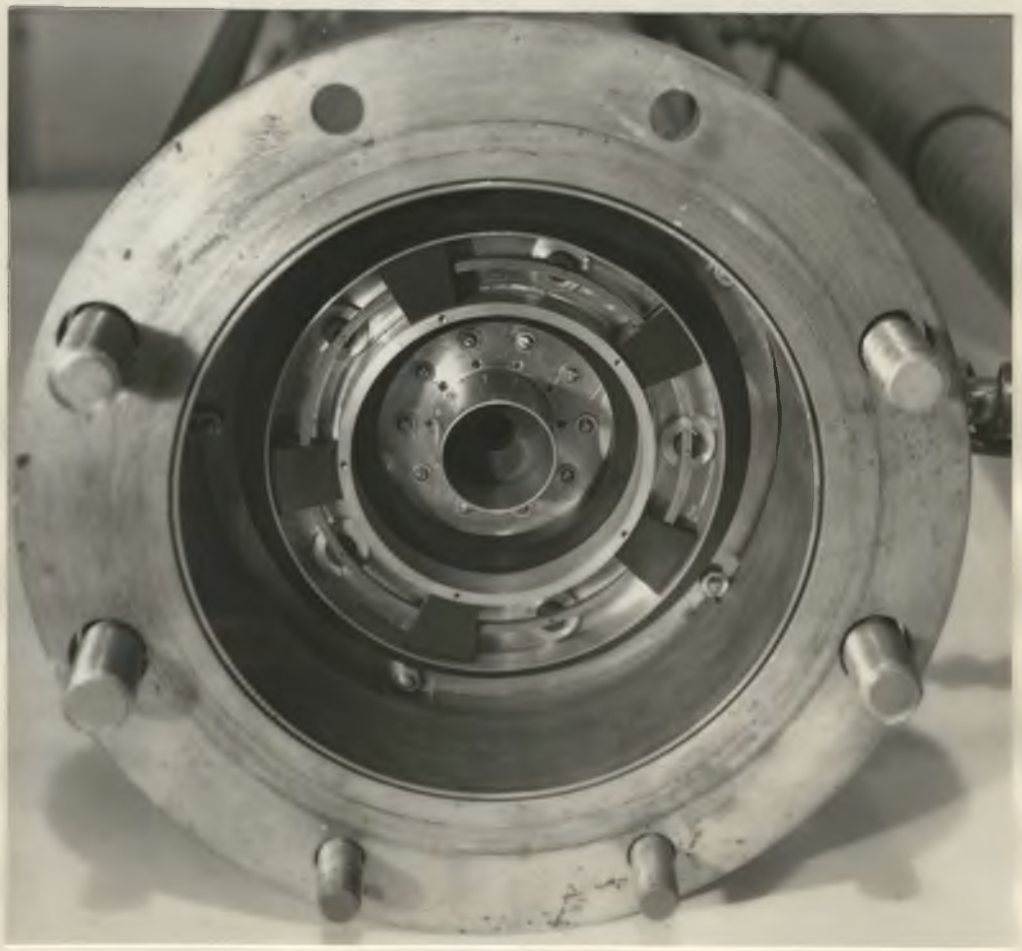


Fig. 19. L/K counter: ring anode connections exposed.

cover for this box has been removed, to show the method of attaching the five anode wires to a common steel ring, using watchmaker's taper pins. Electrical connections are brought out through ebonite insulators in the outer case, fitted with guard rings where necessary, and sealed with O-rings. The central counter has field tubes as well as guard tubes, and has a sensitive length of 76 cm.

A calcium purifier was fitted at one side of the counter, as can be seen in the photographs. This contains trays of calcium chips, and the counter is normally operated with its axis vertical so that adequate circulation of the gas over the heated calcium is achieved by convection. The purifier was used in most of the preliminary experiments, but during the main runs it was sealed off from the counter to avoid any possibility of loss of source through chemical reaction with the calcium.

When this counter was designed, experience of multiwire proportional counters for low energy measurements was still very limited, and, as for the low-background counter, it was decided to take every possible precaution against internal sparking and insulation breakdown at the

high voltages required. The resulting extensive use of guard rings, together with complete screening of all anode connections, makes the detailed design rather complex, but the photographs reproduced here are probably sufficient to show the principal features of the construction.

Operation and Performance of the Counter.

The working voltages of both central and ring counters were in the neighbourhood of 6.8 keV with the argon-methane filling at a pressure of 6 atmospheres, and it was arranged that the outer case was kept at the high potential so that the anode of the central counter could be connected directly to input of its pre-amplifier. Any small difference in the voltages required for the two counters was accommodated by applying a suitable positive or negative potential to the anode of the ring counter.

The electronic equipment employed was almost the same as that used with the low-background counter, as it had been designed for both types of work. A schematic diagram of the arrangement in the present experiment is given in Fig. 20, and some of the circuits are described in the Appendix to this thesis.

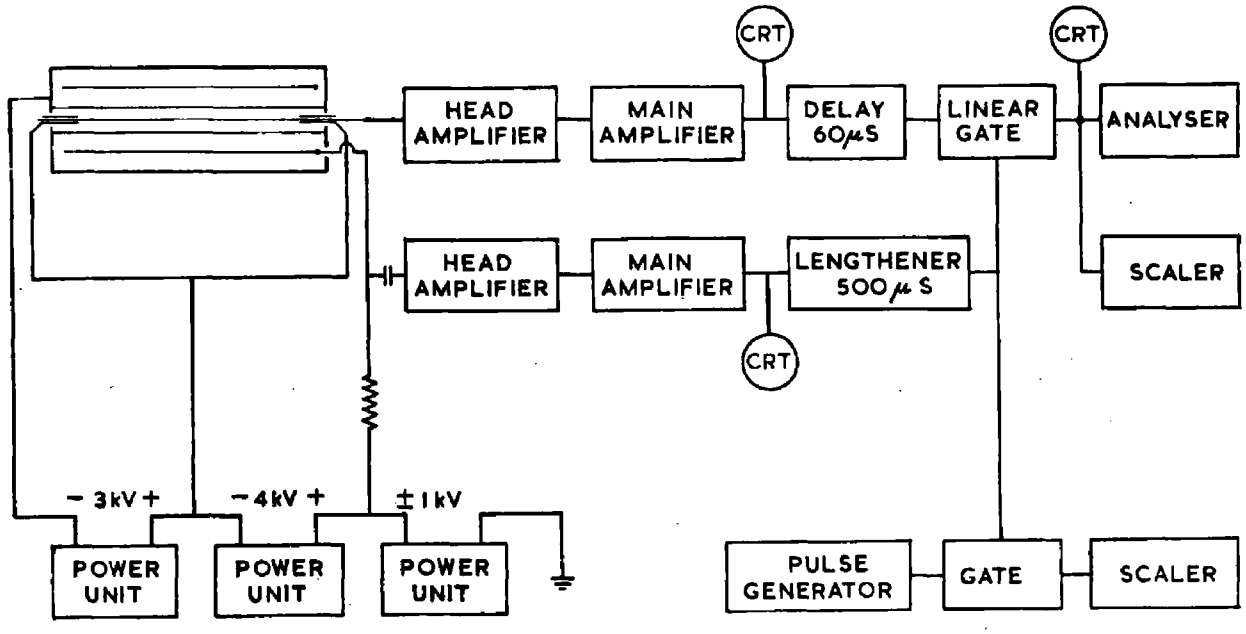


Fig. 20. Electronic arrangement for L/K measurement.

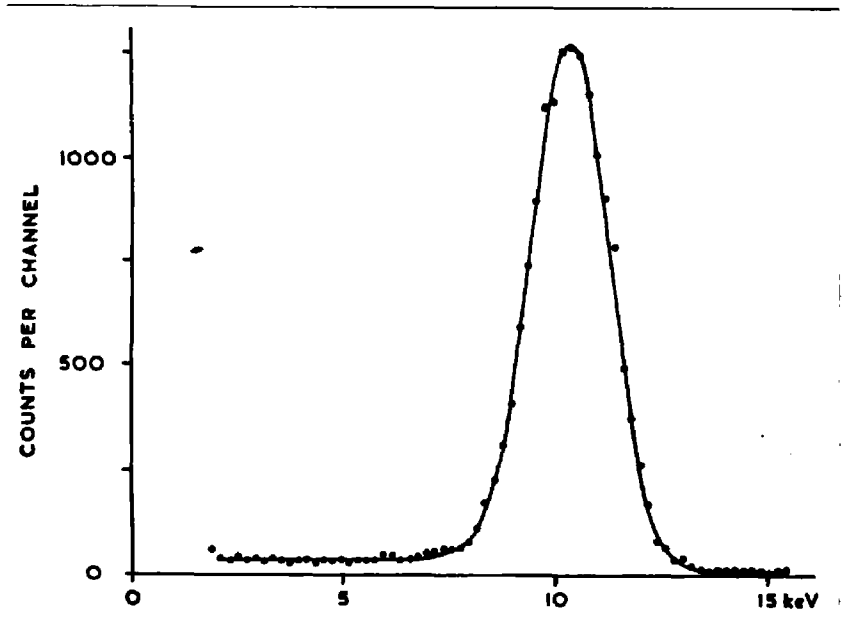


Fig. 21. K-peak obtained from the ring system of the counter with a gas source of Ge^{71}.

The relatively high counting rate in the ring counter causes the anticoincidence gate to be closed for an appreciable fraction of the total time, though the recorded L/K ratio is not affected provided the effective dead time remains constant. The timing unit included in the anticoincidence system provided a useful check of the latter condition. By counting pulses from a gated pulse generator the effective counting time could be determined directly.

The previous work with the low-background counter showed that the efficiency of the type of multiwire ring counter system used here can be high, and its characteristics may be almost like those of a normal single proportional counter. It was, however, felt desirable to check this again with the present arrangement, where the pressure of the filling gas is higher and the individual counting units are larger. The counter was therefore filled with the argon-methane mixture used in the main measurements, at a pressure of 6 atmospheres, together with some Ge^{71} in the form of germanium hydride gas; and the pulse spectrum from the ring counter was analysed. The recorded spectrum in the neighbourhood of the K-capture peak is shown in

Fig 21. The width of the peak indicates that the energy resolution, though slightly poorer than in a normal counter, is quite satisfactory; and also that variations of gain between the five counting units are negligible. The low amplitude of the continuous part of the pulse spectrum at energies below that of the K-peak, suggests that any loss of efficiency due to processes such as electron attachment in the low-field regions between adjacent counting units is very small. The observed low-energy "tail" can be accounted for entirely by cosmic ray background and by end effects resulting from the absence of field tubes in the ring counter. A tail of similar relative amplitude is obtained with proportional counters of normal design which are not end-corrected.

The triggering level of the circuits controlling the anticoincidence gate in the main measurements was set to correspond to an energy of 3 keV, and it may be concluded that then all but a negligible fraction of the K X-rays absorbed in the gas of the ring counter operate the gate. It is found, in fact, that the only appreciable loss of counting efficiency is that arising from absorption of X-rays in the cathode wires between

the two counters, and this is an effect which is calculable.

In both the previous multiwire counter, and the present one, it was noticed that pulses from the central counter are always accompanied by smaller pulses of opposite sign from the ring system. This is due to charges induced on the anode wires of the ring counter by the movement of positive ions produced in the avalanche of the central counter. The ratio of the charges induced on the anodes of the ring, and central, counter respectively, is approximately equal to the ratio of the mutual capacity between the anode wires to the capacity between the exposed length of the central wire and the cathode. In the present system the mutual anode capacity is 0.3 pF, and when capacities to earth are taken into account the ratio of the voltage pulses expected is close to the value of 1/40 which is found experimentally. Since the induced pulses on the ring system have such small amplitudes the gains of the two counters can readily be chosen in such a way that the slight coupling has no effect on the experiments.

Measurement of L/K Ratio.

The Ge^{71} used was made at Harwell by (n,γ) reaction, and was supplied in the form of irradiated germanium oxide. Arsenic 77 and Ge^{67}, which have half-lives of 38 and 40 hours respectively, are also produced in the pile; so before making measurements sufficient time was allowed to elapse for these activities to become negligible. The gaseous compound of germanium employed in the experiments was germanium hydride, GeH_4, which was prepared by reaction of sodium amalgam with a water solution of the germanium oxide. Water vapour was removed from the gas by cooling with solid carbon dioxide, and the excess hydrogen liberated in the reaction was pumped off with the germanium hydride frozen at liquid air temperature.

It was found that the presence of the germanium hydride in the counter, at the partial pressure of less than 1 mm Hg used, had no detectable effect on energy resolution or general performance. By checking background counting rates after the source had been in the counter for several days it was established also that there was no deposition of source on the walls.

For an L/K measurement, a suitable quantity of the germanium hydride was passed into the evacuated counter, together with methane to a partial pressure of 15 cm Hg, and argon was added to a total pressure in the range from 2 to 6 atmospheres. The maximum working pressure was set principally by the mechanical strength of the counter. After filling, a period of three hours or more was always allowed for mixing of the gases before beginning the main counting. Subsequent measurements with any one filling showed no change of counting rate over several days, apart from that expected due to the decay of the Ge^{71}.

Pulse spectra from the central counter, in anticoincidence with the ring counter, were measured at several different filling pressures. In each set of measurements the spectra in the neighbourhood of the 10.2 keV K-capture peak and the 1.3 keV L-capture peak were recorded with the equipment operating under identical conditions, apart from the change in amplifier gain required to cover the two energy regions.

Typical pulse spectra obtained are shown in Figs. 22 and 23, these particular curves having been

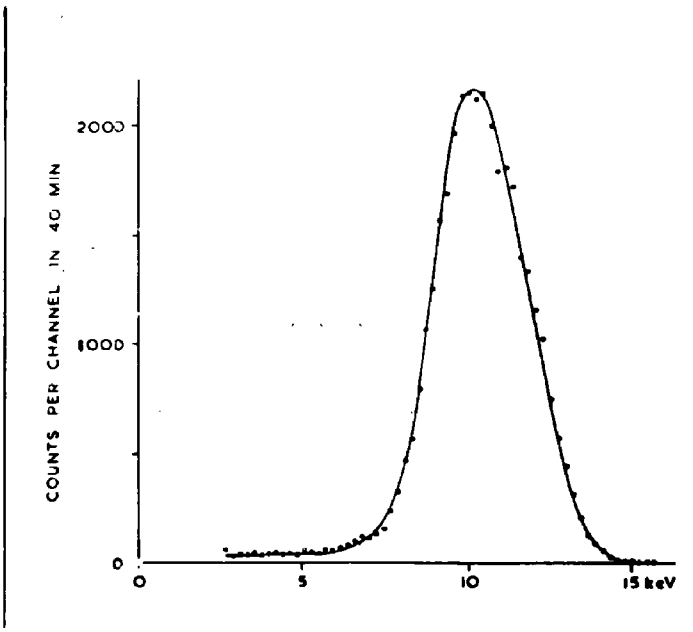


Fig. 22. K-peak of Ge^{71} from the central counter, in anticoincidence with the ring counter.

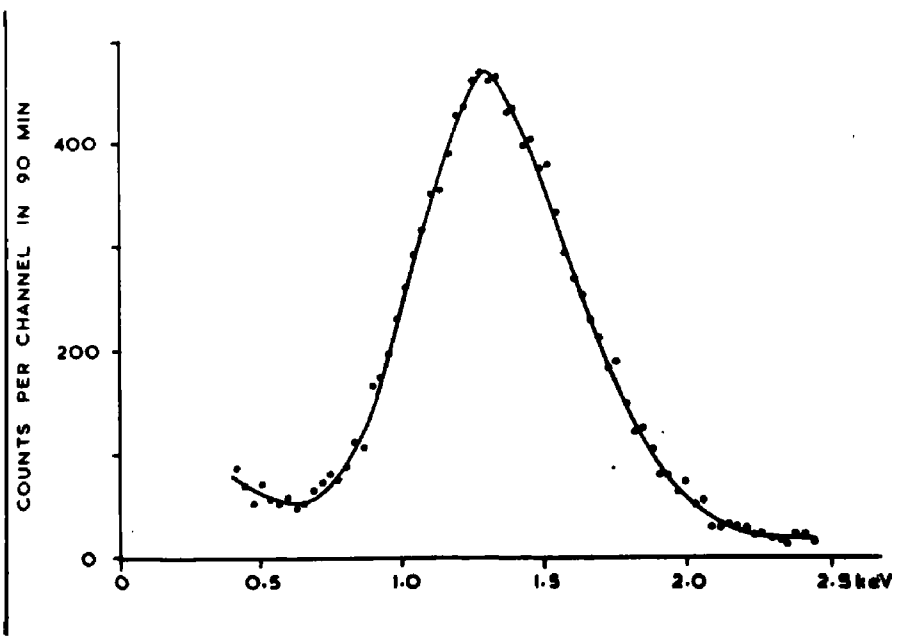


Fig. 23. L-peak of Ge^{71}, measured under the same conditions as the K-peak above.

taken at the maximum pressure of 6 atmospheres. The background spectra were measured separately, and have not been subtracted in these diagrams.

The K-peak shown is slightly broader than a normal calibration peak obtained at this energy. This is due mainly to the presence of the two components, one corresponding to the K absorption energy of gallium and the other to the K_{α} X-ray energy. In the present case, though, there is a further small increase in width due to a practical difficulty in the construction of the counter which resulted in the anode wire being slightly off-centre. This has no appreciable effect on the accuracy of measurement of the counting rate in the peak, which remains well-defined.

Results.

The ratio of counting rates in observed L and K peaks at five different pressures are given in Table 4. No corrections for escape effects have been made here, and only some small corrections for kicksorter deadtime (0.6 ms) have been applied.

The smallness of the change of observed L/K ratio with pressure, at the higher pressures, confirms that escape

TABLE 4.

RATIOS OF COUNTING RATES IN L AND K PEAKS FROM Ge^{71}.

Pressure (atmospheres)	2	3	4	5	6
Observed L/K ratio	0.165	0.144	0.136	0.135	0.135

of X-rays through the ring counter is almost negligible at 6 atmospheres, since errors due to this type of escape would alter rapidly with pressure. However, the probabilities of escape at the ends of the counter, and of absorption of X-rays in the cathode wires, change only linearly with pressure. The magnitudes of the latter effects may therefore be obtained more easily by calculation than from the observed L/K ratios.

The probabilities for the various modes of escape of X-rays from the central counter can be evaluated by methods used in gamma-ray shielding problems, such as discussed by Glasstone (1956), Blizard (1955) and more recently by Price et al (1957). Rigorous calculations for the rather complex geometry of the present counting system are difficult, but since the corrections required are all small sufficient accuracy can be obtained by approximate methods. Such calculations, based on results quoted in the first two articles just referred to, indicate that at a pressure of 6 atmospheres the probability P_1 that a K X-ray emitted in the sensitive volume of the central counter will escape through the ends without being detected in the ring counter is $P_1 = 7 \times 10^{-3}$. It

is found also that the probability P_2 that a K X-ray from the central counter will be absorbed by the cathode wires is $P_2 = 7 \times 10^{-3}$. The remaining mode of escape, by penetration right through the gas in the ring without being absorbed, is, as expected, very small; its calculated probability P_3 being $P_3 = 6 \times 10^{-4}$.

When these three corrections for residual escape effects are taken into account, the L/K capture ratio is given in terms of the ratio of the observed counting rates in the L and K peaks by the modified form of equation (12):-

$$R = \frac{N_L}{N_K} \left[1 - (P_2 + P_3).F\right] - (P_1 + P_2 + P_3).Fk \qquad \ldots\ldots\ldots\ldots(13)$$

The experimental result for N_L/N_K at 6 atmospheres is 0.135, this being a mean value obtained from three complete runs with different samples of germanium hydride. On substituting this result, and the calculated escape probabilities, in equation (13) the value found for the L/K capture ratio is R = 0.128.

Here the values taken for the K fluorescence yield of gallium and for the fraction of K_α X-rays in the K series are 0.50 and 0.86 respectively. It

should be emphasised, however, that the result obtained is very insensitive to these factors, a change of 10% in either of them leading to a change in the value obtained for the L/K capture ratio of only 0.5%.

The probable error in the final result due to statistical errors in counting is 1%, but in assigning overall errors to the measurement other sources of inaccuracy have to be considered also. Some uncertainty might be introduced, for example, in defining the boundary of the low energy part of the L peak, and, to a lesser extent, of the K peak. It is difficult to determine the magnitude of these errors precisely, but they can be estimated. When such estimates are taken into account, the overall error of the final result becomes approximately $(^{+4}_{-2})\%$. The value obtained for the L/K capture ratio of Ge^{71} may thus be stated

$$R = 0.128 \quad (^{+0.005}_{-0.003})$$

Comparison with Theory.

The value for the L/K capture ratio given by the present work is considerably lower than that obtained by Langevin, and a possible explanation for

this difference will be discussed later. The present result is, however, closer to the theoretical value.

Calculation of the L/K capture ratio depends on knowledge of the wave functions of the L and K electrons at the nucleus, and of the transition energies involved, as has already been indicated. In the original work of Marshak single-electron wave functions in the field of a nucleus of effective charge given by Slater screening rules were used, and were evaluated at the nuclear radius. Other approximations have subsequently been employed; and Rose and Jackson (1949) used Hartree wave functions for atoms of atomic number less than 30, and relativistic wave functions for heavier atoms. These calculations have been extended by Brysk and Rose (1955) who have added further corrections for effects of finite nuclear size, variation of wave functions over the nuclear volume, and screening. In the case of Ge^{71}, however, the values for the L/K capture ratios given by the various methods do not differ by more than a few percent. Taking the results of Brysk and Rose, and a transition

energy of 231 keV, the ratio of the probabilities of L_1 capture and K capture comes to 0.105, while the ratio of the probabilities of L_2 capture and K capture is 0.0006. The corresponding total L/K capture ratio is thus 0.106, the result already quoted.

Although the present measured ratio is in better agreement with this than the previous experimental values, there remains a difference of 20% between theory and experiment.

The possible sources of error in the measurement have been carefully examined, and it seems very unlikely that there is any systematic, or accidental, error large enough to account for this discrepancy. It would appear that part, at least, of the difference between the theoretical and experimental results is real, and further investigation of the theoretical calculation seems desirable.

It has recently been suggested by Odiot and Daudel (1956) that there may sometimes be appreciable error in the usual theory of electron capture due to neglect of correlations between the positions of the electrons in the atom, and of effects of the change in

nuclear charge. These authors emphasise that it is incorrect in principle to consider a K-capture process as the removal of a K electron from the atom, since all electrons are indistinguishable, and that the event should be regarded rather as a transition from a complete atom of atomic number Z, to another atom of atomic number Z+1 with a vacancy in its K shell. Unfortunately, complete calculations are difficult, and only practicable for the lightest atoms. It is found, though, that the effect of the change of nuclear charge is negligible for atoms heavier than Be^7, and that the correlation effects are more important. When a part of the electron correlation, that due to the exclusion principle, is considered, the values for L/K capture ratios of helium (a hypothetical case), Be^7, and A^{37} are increased by factors of 5, 3, and 1.25 respectively. A full calculation taking into account all the correlations has only been made for helium, and this gives an L/K capture ratio which is increased by a factor of 10.

From these results it would appear that this development of the theory of electron capture leads always to higher values for L/K capture ratios, though

the increase over the previous results is greatest for atoms of low atomic number. There are insufficient data to make definite conclusions about Ge^{71}, but it seems possible that more complete calculations may bring the theoretical L/K capture ratio into closer agreement with the experimental result found in the present work.

The K Fluorescence Yield of Gallium.

The difference between the present value for the L/K capture ratio of Ge^{71} and the value reported by Langevin is so large that it was felt desirable to investigate possible reasons for the discrepancy. The simplest explanation would be provided by an error in the fluorescence yield of gallium assumed by Langevin, so, to check this, an independent measurement of the fluorescence yield has been made. The fluorescence yield of gallium had not, in fact, been previously measured, and the result used by Langevin was obtained by interpolation from data on neighbouring atoms.

The multiwire counter employed in the measurement of the L/K capture ratio was used in this work on fluorescence yield also, but here the anti-

coincidence circuit was disconnected so that all pulses from the central counter were recorded.

The counter was filled first with methane to a pressure of one atmosphere, together with a trace of the Ge^{71} gas source. The pulse spectrum then obtained from the central counter was analysed, and the counting rate in the K peak determined. This gave a measure of the rate of emission of K Auger electrons in the sensitive volume, since the chance of absorption of K X-rays in the methane is small. Argon was then added to the counter to bring the total pressure to 5 atmospheres, and, after allowing time for thorough mixing, the K peak was measured again. In this case both K X-rays and Auger electrons were detected, so, by comparison with the previous counting rate, the rate of emission of K X-rays could be determined.

A subsidiary experiment was performed to check that no loss of source occurred during the addition of the argon to the methane filling already in the counter. Here the argon was introduced in two stages, to give pressures of 3 and 6 atmospheres successively, and the absence of any change in counting rate at the second addition of argon was verified. In all these measurements

the ring system was not operated as a counter, but it was still effective in reducing K X-ray escape effects. The ratio of the voltages on the ring and central counters was kept constant throughout to prevent any changes in effective sensitive volume of the central counter.

The ratio of the counting rates obtained with the methane filling at 1 atmosphere, and with the argon-methane filling at 5 atmospheres is 0.490. To determine the fluorescence yield from this ratio it is necessary to take into account absorption of X-rays in the methane filling, and also escape of X-rays when the argon filling is used. Calculations showed that absorption in the methane led to the detection of one K X-ray in the central counter for every 40 emitted; and that with the argon-methane filling the effective probability of X-ray escape (equivalent to $P_2 + P_3$ in the previous work) was 8×10^{-3}. When these corrections are applied, the experimental value obtained for the K fluorescence yield of gallium is 0.53.

The accuracy of this measurement is not as great as that of the L/K capture ratio, and, taking into

account possible uncertainties in defining the boundaries of the peaks, the estimated probable error in the result for the fluorescence yield is ± 6%.

During this experiment on fluorescence yield the fraction of K_α X-rays in the K series, the factor k in equation (12), was also determined. With the methane filling at one atmosphere, the counting rate in the L peak, due mainly to escaping K_α X-rays, was measured. By subtracting the component due to L-capture itself, found from the known L/K capture ratio, the rate of emission of K_α X-rays in the central counter was found, and could be compared with the total rate of emission of K X-rays. Corrections similar to those mentioned above were applied, and the final result obtained for the fraction of K_α X-rays in the K series of gallium is k = 0.83. The accuracy of this particular result is, however, not high, and it is considered that the difference between this value and the one used by Langevin (k = 0.865) is not significant.

Discussion.

When the experimental data of Langevin's measurement of Ge^{71} are recalculated with the fluorescence yield of 0.53 ± 0.03 found now, then the result obtained

for the L/K capture ratio becomes 0.14 ± 0.07. (Here the indicated error includes the estimated error in fluorescence yield). This result, although still not precise, is in much better agreement with the theoretical value, and with the value found in the present measurement, than the ratios obtained previously. It therefore seems probable that the earlier apparent discrepancies have been due largely to incorrect values of fluorescence yield. The experimental data are unfortunately not sufficiently accurate to establish this completely, and it is not impossible that other factors assumed, such as the fraction of K_α X-rays in the K series, may also be in error.

It should be pointed out that the fluorescence yield of gallium found now is higher than those suggested by most empirical or semi-empirical relations between fluorescence yield and atomic number. The curves obtained by Burhop (1955), Bergstrom (1955), and Laberrique-Frolow et al (1956) give values ranging from 0.473 to 0.50 for gallium. However, there is considerable scatter in the experimental data on which

these curves are based, and recent measurements on argon and krypton by Heintze (1955) and detailed theoretical calculations by Rubenstein and Snyder (1955) also give higher values of fluorescence yield than those previously accepted.

Conclusion.

The results of the present work on Ge^{71} remove the most serious of the apparent discrepancies between theoretical and experimental L/K capture ratios. It can now be stated that in the two cases, A^{37} and Ge^{71}, of electron-capture decay of known transition energy and accurately measured L/K capture ratio, theoretical and experimental ratios agree to within 20%. It is probable also that in the less certain case of Kr^{79} a more accurate fluorescence yield will give better agreement. There is therefore no evidence now for the gross errors in the theory of electron capture which the previous results suggested, and it seems likely that the relatively small discrepancies remaining may be reduced when more complete calculations are made.

Appendix.

A NON-BLOCKING AMPLIFIER AND ANTICOINCIDENCE GATE.

The electronic equipment employed in the work described in this thesis was of standard type as far as possible. However in several of the experiments the performance demanded of the apparatus was unusual in certain respects, and it was found necessary to develop special circuits. Problems connected with the overloading of the amplifiers by excessively large input pulses became important in many of the low energy measurements; and had even greater significance in the work with the low-background multiwire counter, which required a very high performance from the equipment controlling the anticoincidence gate. Methods of improving the overload properties of standard amplifiers were therefore developed, and a special type of non-blocking amplifier was eventually constructed for use with the multiwire counters. For these counters it was also necessary to design a linear anticoincidence gate which would operate efficiently with input pulses varying widely in amplitude and

duration. The electronic circuits developed by the writer in solving these problems are largely original, and as they have been found very satisfactory and suitable for other applications it may be useful to describe them here.

Overloading in Linear Amplifiers.

The majority of applications of linear amplifiers in counting experiments involves pulses varying in amplitude by not much more than a factor of 20:1, and the behaviour of the amplifier for pulses outside this range is unimportant. However, in experiments such as those described in this thesis, where counting extends down to low energies, the ratio of the maximum amplitude of the background pulses to that of the pulses being studied may be much greater, and the overload properties of the electronic equipment are of major importance.

When an excessively large pulse is applied to the input of a normal amplifier one or more of the stages may saturate, but it can usually be arranged that the resulting distorted output pulse is still large enough to be outside the part of the spectrum

being studied. The charging of coupling condensers through flow of grid current is more serious, since this leads to alterations of gain and displacement of the zero level of the output which may persist for a time much longer than the duration of the overloading pulse itself. Following pulses may therefore be incorrectly recorded, or may even be missed altogether.

There are several ways of avoiding this type of grid current blocking in condenser-coupled circuits. Chase and Higinbotham (1951) designed an amplifier based on cathode-coupled stages arranged so that the maximum output of any one stage is insufficient to cause grid current in the following stage. Magee, Bell and Jordan (1951) achieved a similar result by adding diode limiters to normal pentode amplifier stages. An alternative method (Scarrott, 1950) of reducing charging of a coupling condenser is to include a large resistor in series with the grid of the following valve.

Grid current blocking is due basically to nonlinear effects, but another type of overload disturbance can arise purely through limitations in

in the low frequency performance of the amplifier system. The output voltage from a counter operated with a large load resistance is almost a step waveform; and to obtain short pulses a differentiating circuit, of time constant T_1 say, is normally included in the amplifier. With a single differentiating circuit the pulses are of the same sign throughout but if there is any other restriction to the low frequency response of the system, such as might be imposed by an A.C. coupling of time constant T_2 (where $T_2 >> T_1$), then the pulses overshoot the baseline. The overshoots are of long duration, decaying almost exponentionally with time constant T_2; and the ratio of the amplitude of an overshoot to that of the corresponding main pulse is approximately T_1/T_2.

It is usual to arrange that the time constants of all the coupling circuits, and of the input circuit, are so large compared with the single short differentiating time constant T_1 that the amplitudes of overshoots are negligible. However, when overload pulses of very large amplitude are present this is difficult to achieve. An upper limit to the time

constant of the input circuit, for example, is determined by the need to restrict piling-up of separate pulses; while if the input circuit itself were used to define the time constant T_1 then the large bandwidth of the remainder of the amplifier would lead to a high noise level. In a normal amplifier these restrictions to frequency response may give rise to appreciable overshoots after large input pulses, causing errors in the recording of following smaller pulses. This is illustrated in Fig. 24(a), which is drawn for the case $T_2 = 150.T_1$ and for an overloading pulse 200 times the amplitude of the smallest pulse shown.

Difficulties of this nature became prominent during the work on Cl^{36}, and a method of avoiding them by the use of two differentiating circuits of equal time constant was developed then, and was later much used.

Double Differentiation.

If the normal differentiating circuit used to shape the pulses is replaced by two separate differentiating circuits of equal time constant T_1,

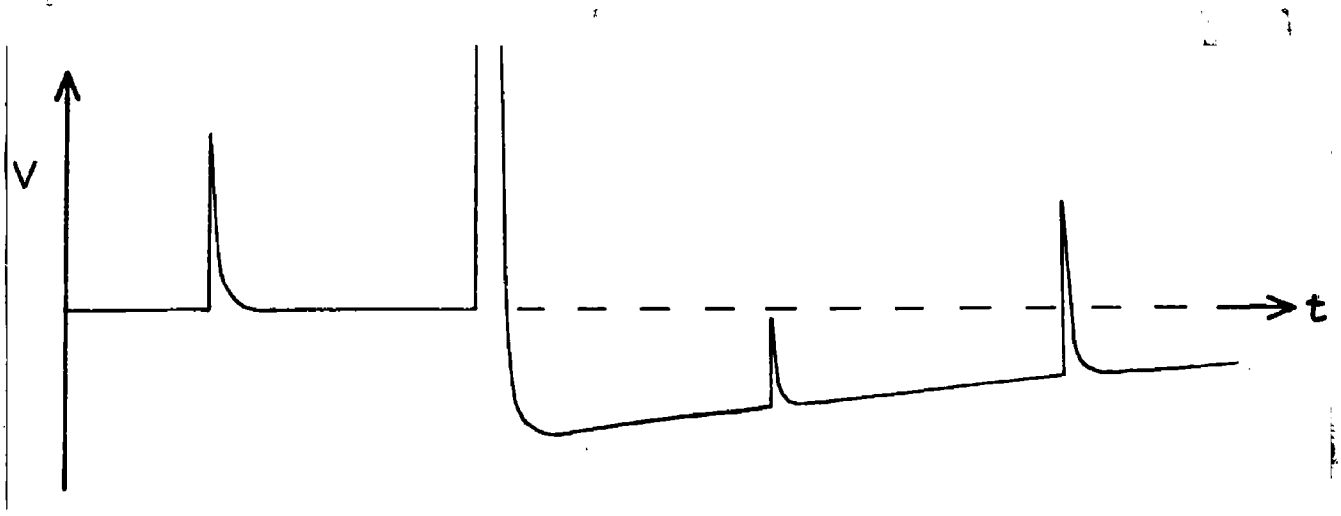
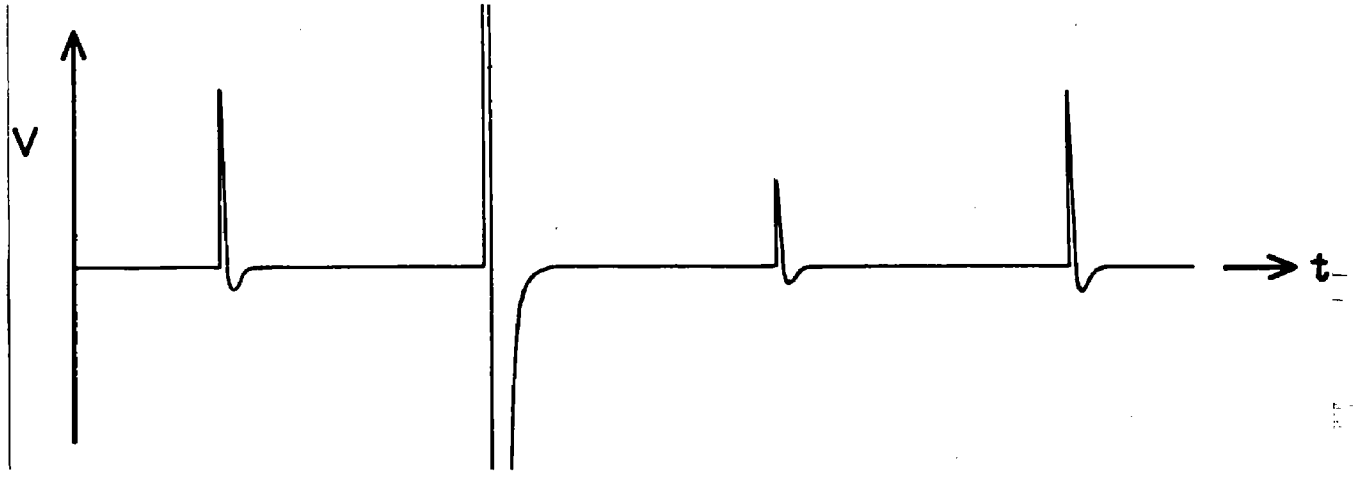


Fig. 24. (a) Typical waveform obtained when pulses shaped by a single differentiating circuit are passed through an A.C. coupling circuit. Here $T_2 = 150\ T_1$, and the amplitude of the overloading pulse is 200 times that of the smallest pulse shown.



(b) Waveform obtained if the pulses are shaped by two differentiating circuits of equal time constant T_1, all other conditions remaining the same as in (a).

than pulses having negative overshoots of relatively large amplitude are produced. The output voltage corresponding to an input step of unit amplitude has the form

$$V = (1 - \frac{t}{T_1}).e^{-t/T_1},$$

the overshoot having a maximum amplitude of $1/e^2$ of the amplitude of the main pulse. This overshoot is, however, of short duration, the output voltage being -1% of the maximum value at a time approximately $6T_1$ from the beginning of the pulse. The integral of the voltage over the pulse and the overshoot also comes close to zero in a time of the same order, so subsequent coupling condensers are left with less charge than in the case of a normal singly-differentiated pulse. With double differentiation a coupling circuit of time constant T_2 has the effect of producing a small positive overshoot after the main negative one, but the amplitude of this is approximately $(T_1/T_2)^2$ times that of the initial pulse so it is easily made negligible even for a large input pulse. The output signal obtained in the presence of overloading is like that illustrated

in Fig. 24(b), instead of that in Fig. 24(a), and the probability of incorrect recording of pulses is very much reduced.

The two differentiating circuits for shaping the pulses may be placed at different stages of the amplifier, and it is convenient to use a small load resistance on the proportional counter itself to make the time constant of the input circuit equal to T_1, while placing the second differentiating circuit later in the amplifier where it is effective in reducing noise. This eliminates piling-up of pulses in the first stage.

The full advantage of this type of pulse shaping is only obtained when the amplifier has already been designed so that grid current blocking cannot occur, but it has been found that even in standard amplifiers the overload properties can be considerably improved by the addition of the second differentiating circuit.

It should be mentioned that a non-blocking amplifier using two delay lines to produce a negative pulse after each normal positive pulse has been described

by Fairstein (1956). This method of pulse shaping is similar in principle to the technique described here, which was developed independently. The present amplifier is simpler, and more suitable for the higher gain required in low energy measurements with proportional counters.

A Non-blocking Amplifier using Double Differentiation.

A circuit diagram of the amplifier built for the work with the multiwire counters is given in Fig. 25. Four cathode-coupled push-pull stages are employed, which give satisfactory linearity and gain stability without external negative feedback.

Grid current blocking is avoided in the first three stages by using direct coupling. Condenser coupling is still used between the third and fourth stages, but this is arranged to act as one of the pulse shaping circuits and has a time constant of only 15 μsec, so recovery from any blocking here is rapid. The other differentiating circuit is formed by the stray capacity of the counter, and the input resistance in the first stage.

The first three stages form themselves a

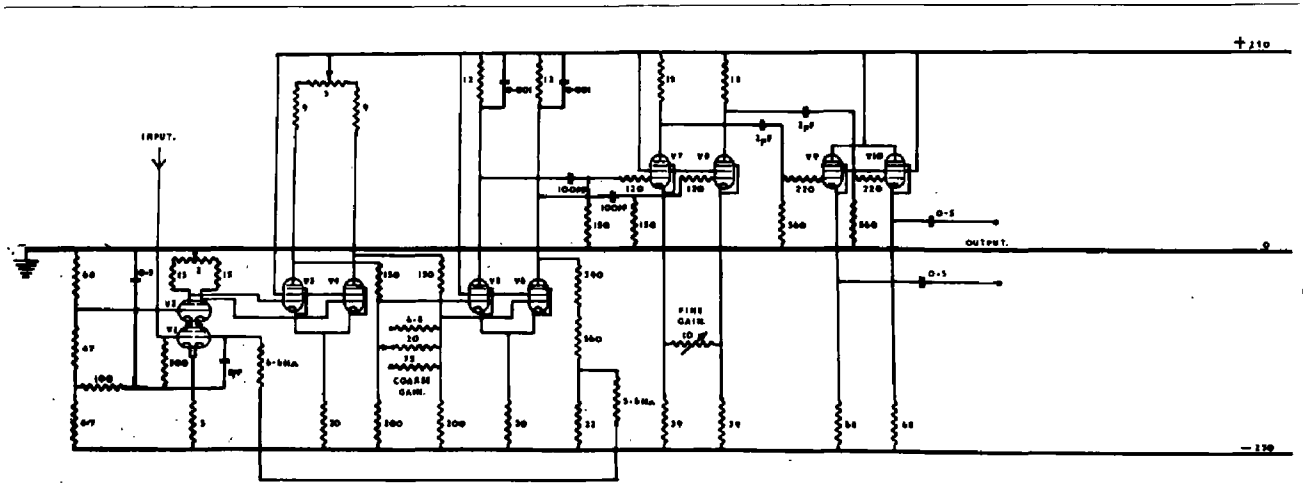


Fig. 25. Circuit of non-blocking amplifier.

In this diagram and in Fig. 26 all triode valves are type CV 455, pentodes are CV I38, and diodes are CV I40 or CV I092. Resistances are in kilohms and capacities in microfarads unless stated otherwise.

D.C. amplifier of considerable gain, and in preliminary tests the effects of slow drift in the input valves were troublesome. This difficulty was overcome by introducing negative feedback, for direct current only, round the three stages, thus making the amplifier self balancing. An integrating circuit of time constant 100 seconds makes feedback negligible at signal frequencies.

The effective integrating time constant of the whole amplifier is defined by the anode circuits of the third stage, and is made comparable with the differentiating time constant to give the maximum ratio of signal amplitude to noise. With the relatively low input resistance needed to give a suitable input time constant, a large part of the frequency spectrum of the thermal noise comes into the region passed by the amplifier, so the noise level is slightly higher than that of a normal amplifier of the same gain. In the type of work described in this thesis this disadvantage is amply compensated by the very much better overload properties of the present amplifier.

It should be noted finally that the circuit given is that of the amplifier as it was actually used

in the experiments, and with the experience gained it would now be possible to make some minor modifications and simplifications. However, such improvements would have little effect on the performance of the amplifier, which was found in practice to be very satisfactory.

A Linear Anticoincidence Gating Unit.

The work with the multiwire counters required a gating unit which would enable a multichannel kicksorter, and scalers biased at different voltages, to be used simultaneously. The low background counting rate of the first of these counters demanded an anticoincidence system of very high efficiency, and a special anticoincidence gate had eventually to be developed.

The basic circuit employed is a simple cathode-coupled gate, but the triggering system used to operate it had to be more complex. A simple univibrator would not be satisfactory since it takes a finite time to recover after each pulse and is insensitive during this period. A form of lengthening circuit was used instead, which closes the gate at the beginning of a pulse and keeps it closed until a fixed time has elapsed

after the end of the pulse or any following overshoot. To ensure reliable closing of the gate by pulses due to interference it is arranged also that the system is sensitive to signals of either sign.

A circuit of the complete anticoincidence unit is given in Fig. 26.

Pulses from the amplifier connected to the ring counter ("veto pulses") are passed first to a phase splitter (valve V2) which produces two signals of opposite polarity. A mixing circuit (V3, V4a) selects whichever signal is positive and passes it, together with pulses from the discriminator defining the upper energy limit, to the pulse shaping system.

The pulse shaper comprises two Schmitt discriminators and a diode lengthener. The first Schmitt circuit produces output pulses of constant amplitude and of the same duration as the input pulses. These are then lengthened by a fixed amount in the diode circuit, and trigger the second Schmitt discriminator which generates the rectangular pulses required to operate the gate itself (V11).

The pulses from the central counter pass through

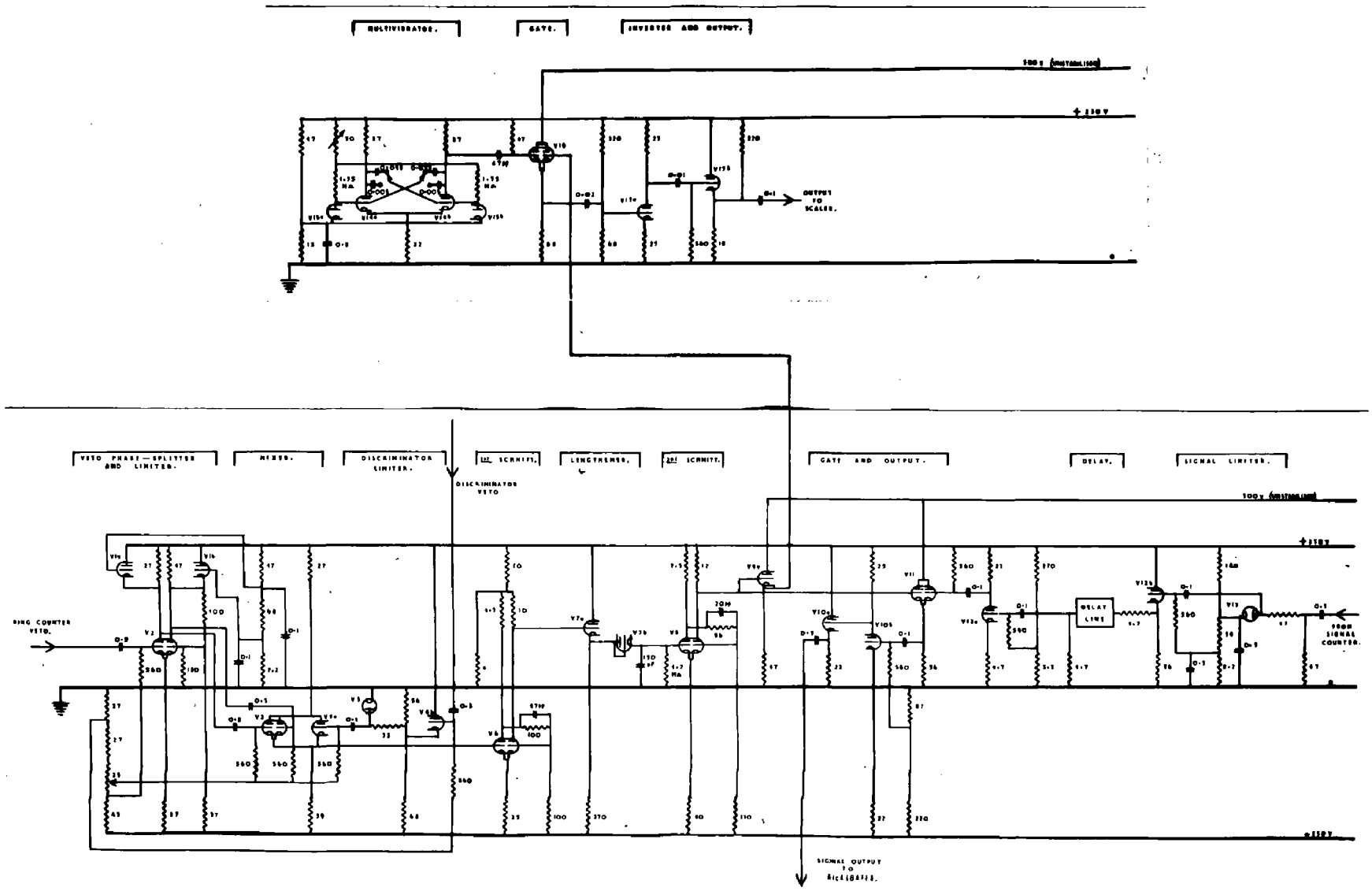


Fig. 26. Circuit of complete anticoincidence gate.

a limiter, a delay line, and an inverter before reaching the gate valve V11. The output signals from the gate are negative, and are inverted again and passed to a final cathode follower feeding the cable to the kicksorter and scalers.

The variable duration of the gate closing time in this anticoincidence unit makes it difficult to calculate the correction required for the fraction of the counting time for which the gate is closed. Although the correction is usually small it was felt desirable to make provision for determining it experimentally, and this facility was found useful in the work on Ge^{71}.

To measure the dead-time correction an auxiliary gate is operated in parallel with the main gate. A multivibrator (V14a, V14b, V15a, V15b) generates a square waveform of constant repetition frequency, and this is differentiated to give short pulses, which are passed to the auxiliary gate (V16). The pulses transmitted by the gate are inverted and counted by a scaler. The counting rate recorded by the scaler thus represents the effective counting time,

and the scaler can in fact be used as a clock for timing the experiments. The repetition frequency of the timing pulses is made much higher than the anticoincidence counting rate to be measured, so statistical errors in the determination of effective counting time are always small compared with the statistical errors of the main measurement.

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