EXPERIMENTAL STUDIES OF ORBITAL ELECTRON CAPTURE.

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PREFACE.

This thesis contains an account of research conducted at the University of Glasgow between September 1955 and April 1958.

Chapter 1 consists of a general review of beta-decay theory, a statement of those results of the theory which are relevant to the work described later, and a survey of the experimental data on orbital electron capture and the Fierz interference term.

Chapter 2 contains a discussion on the limits which the wall effect imposes on the usefulness of conventional proportional counters, and a description of two counters which have been designed to eliminate this effect. The multi-wire counter was designed and operated by the author after Drever et al. had shown that such a counter would work satisfactorily, but it was felt that the design was sufficiently original to warrant a full description here. The plastic counter was designed and operated jointly by the author and Dr. G.M. Lewis .

In chapter 3 results are presented of measurements on the ratio of K-capture to positron emission in F^{18}. This measurement sets an upper limit on the contribution of the Fierz interference term to the decay probability in Gamow-Teller interactions. The author assisted Mr. R.W.P. Drever and Dr. A. Moljk in the prosecution of these experiments and was also responsible for the initial draft of the published paper describing them.

The results of measurements on the K/β^+ ratio in C^{11} are presented in chapter 4. This measurement sets a new upper limit on the contribution of the Fierz interference term to the decay probability in Fermi interactions. These experiments, and the analysis and publication of the results, were carried out jointly by the author and Dr.G.M.Lewis, but the apparatus was designed, constructed and operated mainly by the author.

A direct measurement has been made of the L/K-capture ratio in I^{126}, using an internal source scintillation spectrometer technique, and this work is described in chapter 5. The author was assisted in this work by Mr.E.Gabathuler.

Chapter 6 contains a description of measurements carried out on the electron capture decay of As^{74}, resulting in values for the ratio of electron capture to electron emission, and the ratio of L-capture to K-capture.

In chapter 7 a description is given of some experiments on the decay of As^{76}, leading to a very low upper limit on the probability of electron capture in this isotope. The author was solely responsible for the work described in chapters 6 and 7.

The significance of the present work on K/β^+ and L/K ratios is briefly stated in chapter 8 , and the relative importance of the various outstanding problems in this field is discussed. A section of this chapter is devoted to a discussion on the conclusions which can be drawn from the Fierz condition, having regard to recent theoretical and experimental work on parity non-conservation.

The appendix contains a description of preliminary experiments to measure the beta-spectrum of He^6 by the internal source technique, using a LiI crystal. This work was conducted jointly by the author and Dr. G.M. Lewis.

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CHAPTER ONE

INTRODUCTION.

1. GENERAL THEORY OF BETA-DECAY.

This section, the purpose of which is to present some results which will be used throughout the remainder of the thesis, is based largely on the numerous review articles on beta-decay which have appeared since about 1945. (Refs. 1-5). No attempt has been made here to survey the large amount of material which has been published since the end of 1956 dealing with the non-conservation of parity and related topics, since little of this work is directly relevant to the study of electron-capture transition probabilities, which is the matter of primary importance here.

A. INTRODUCTION.

At the end of 1956 it appeared that the character of the weak interaction in beta-decay was becoming clearer. As noted below several experiments had shown that the interaction Hamiltonian contained the scalar and tensor terms, with perhaps a certain amount of pseudoscalar coupling, and the remaining questions concerned the relative magnitudes and signs of these three terms. However, at this time discussions on the possible solution to the $\tau - \theta$ puzzle led Lee and Yang (6) to point out that no experimental evidence exists for the hypothesis of conservation of parity in the weak interactions in meson and hyperon decay and in beta-decay. It was found that the addition to the interaction Hamiltonian of the five couplings which do not conserve parity has no effect on the results of any of the experiments which had previously been carried out, such as those on the allowed and forbidden spectra, beta-gamma angular correlations, and beta-neutrino angular correlations. In the calculations relating to these experiments the coupling constants C and C' (cf. section B) always appear in terms proportional to $|C|^2$ plus terms proportional to $|C'|^2$.

The determination of the amount of mixing of the C and C' interactions requires the detection of an interference term of the type CC', and it turns out that such a term can only be associated with a pseudoscalar formed out of the experimentally measured quantities, such as $CC'\underline{p}_1\cdot(\underline{p}_2\times\underline{p}_3)$ or $CC'\underline{p}\cdot\underline{\sigma}$, where $\underline{p}_1, \underline{p}_2, \underline{p}_3,$ $\underline{\sigma}$ are three momenta and a spin. It is seen that in the experiments mentioned above no such terms could be formed from the measured quantities. Lee and Yang proposed several experiments in which such terms could be detected if parity conservation is violated. These experiments have been carried out, and show clearly that parity non-conserving terms are present in the weak interactions in beta-decay (7-9), and in π and μ -meson decay (10-13). In addition the experiment of Wu et al. (7), in which a large asymmetry was observed in the electron intensity with respect to the nuclear polarization direction, also indicates that charge conjugation invariance is violated. No experimental results have been reported as yet on the problem of time reversal invariance.

During 1957 a very large amount of work, both theoretical and experimental, has been done on the problem of the validity of the conservation laws in the weak interactions, and on the related problem of the exact nature of the interactions. Throughout the remainder of this chapter the theoretical results will be presented with the interaction Hamiltonian as given by Lee and Yang (6), but, since most of the theoretical results discussed here are not affected by considerations of this type, no further reference to this work will be made here. The discussion on the Fierz interference terms in chapter 8 is developed in the light of the more recent results.

B. THE ALLOWED BETA-TRANSITION PROBABILITY.

The probability of a beta-transition is calculated by assuming an interaction between the nucleons and leptons analogous to the interaction between charges and the electro-magnetic field. Since the decay rates of beta-transitions are much slower than those of comparable gamma-transitions, it is clear that the beta-interaction is comparatively weak. It turns out that five independent relativistically invariant forms can be chosen for the interaction Hamiltonian, and these are usually called the scalar, vector, tensor, pseudovector and pseudoscalar interactions (S,V,T,A,P). As an example, the vector type of interaction, which was the one used by Fermi (14) in his original formulation, is now given, for negatrons, by

$$\mathcal{H}_V = H_V + H_V' = g(\psi_p^\dagger \gamma_4 \psi_n)(C_V \psi_e^\dagger \gamma_4 \gamma_\mu \psi_\nu + C_V' \psi_e^\dagger \gamma_4 \gamma_\mu \gamma_5 \psi_\nu) \quad \text{------} \; (1)$$

where ψ_p, ψ_n, ψ_e and ψ_ν are the wave functions of the proton, neutron, electron and neutrino respectively. C_V and C_V' are the coupling constants for the parity conserving and parity non-conserving parts of the vector interaction and are a measure of the contribution of the vector coupling to the total interaction. The γ's are combinations of the Dirac matrices, and the Fermi constant g determines the strength of the interaction.

The complete beta-interaction is then a linear combination of the above five:

$$\mathcal{H} = \sum_k H_k + H_k' \qquad k = S, V, T, A, P \quad \text{----} \; (2)$$

The C_k, C'_k are so chosen that $\sum_k (|C_k|^2 + |C'_k|^2) = 1$

Neglecting the effect of the Coulomb field of the nucleus on the electrons, both electrons and neutrinos can be represented by plane waves $\propto e^{i\underline{k}.\underline{r}}$, where k is the wave vector of the particle. Since r must be of nuclear dimensions, and since kR is normally $\ll 1$ (R=nuclear radius), it is clear that in a power series expansion of the plane waves the successive terms get progressively smaller. Clearly transitions in which the lepton wave functions contain the first terms in such expansions will take place at a faster rate than those in which these terms vanish due to selection rules. The former are referred to as "allowed" transitions. On examination of the matrix elements of the allowed transitions the following selection rules are obtained:

S , V	$\Delta I = 0$,	no parity change.
T , A	$\Delta I = 0 , \pm 1$, no $0 \rightarrow 0$,	no parity change.
P	$\Delta I = 0$,	parity change.

ΔI is the nuclear spin change in the transition.

It is clear that the P interaction cannot co-exist with the other interactions because of the different parity selection rule, and, since there are several allowed transitions with $\Delta I = 1$, it must be assumed that there is no P interaction in allowed transitions.

The selection rules split the remaining four interactions up into the group S and V (Fermi), and the group T and A (Gamow-Teller).

Starting from simple statistical considerations of the distribution of the electron and neutrino momenta, the probability per unit time that a beta-particle is emitted in an allowed transition, with energy between W and W + dW, is obtained as

$$P_{\pm}(W)\,dW = \frac{g^2}{2\pi^3} F(\mp Z, W)\, p W (W_0 - W)^2 \times$$

$$\times \left(|C_F|^2 |M_F|^2 + |C_{G-T}|^2 |M_{G-T}|^2\right)\left(1 \mp \frac{b}{W}\right) dW \quad \text{-----} \; (3)$$

where $|C_F|^2 = |C_S|^2 + |C_S'|^2 + |C_V|^2 + |C_V'|^2$

and $|C_{G-T}|^2 = |C_T|^2 + |C_T'|^2 + |C_A|^2 + |C_A'|^2$

The system of units in which $\hbar = m = c = 1$ is used throughout, and the neutrino rest mass is assumed to be zero. The upper sign in eqn. 3 refers to positron emission and the lower to negatron emission. F is a function correcting the energy spectrum for the effect of the nuclear Coulomb field on the electrons. p is the electron momentum and W_0 is the total energy available for the transition. M_F and M_{G-T} are respectively the Fermi and Gamow-Teller nuclear matrix elements.

The transition probability for electron-capture can be calculated in an analogous way and will be presented below.

The term b, which arises from interference between the S and V interactions and between the T and A interactions, is known as the Fierz interference term (15), and is given by

$$b = 2\gamma \,\mathrm{Re}\left\{ \frac{(C_S C_V^* + C_S' C_V'^*)|M_F|^2 + (C_T C_A^* + C_T' C_A'^*)|M_{G-T}|^2}{|C_F|^2 |M_F|^2 + |C_{G-T}|^2 |M_{G-T}|^2} \right\} \qquad \text{------ } (4)$$

where $\gamma = \sqrt{1-\alpha^2 Z^2}$ and $\alpha = \frac{e^2}{\hbar c}$ the fine structure constant.

Clearly the determination of the relative magnitudes of the coupling constants C_k, C_k' is of great importance in the theory of weak interactions, and therefore the constant b should be known as accurately as possible. A detailed discussion of the experimental data on this constant will be given below.

C. THE FORBIDDEN BETA-TRANSITION PROBABILITY.

It has been seen above that allowed transitions can take place only between nuclear states with the same parity and where the spin changes by no more than one unit. In the derivation of the transition probability for allowed transitions two important approximations are usually made:

a. additional terms in the V,T and A forms of the interaction are neglected due to their being of order v/c , where v is the nucleon velocity.

b. all terms other than the first in the expansion of the lepton plane wave functions are neglected, i.e. $e^{i\underline{k}.\underline{r}}$ is replaced by unity.

Clearly when the above selection rules for allowed transitions are violated these neglected terms will determine the rate of the transition which, in view of the relatively small size of the terms, will be a slower or "forbidden" transition. An examination of the new matrix elements obtained leads to the conclusion that there are n-times forbidden transitions only for (2)

$$|\Delta I| = n\ ,\ n + 1 \quad ; \quad \Delta\pi = (-1)^n \qquad (n \neq 1)$$

$$|\Delta I| = 0\ ,\ 1\ ,\ 2 \quad ; \quad \Delta\pi = -1 \qquad (n = 1)$$

where $\Delta\pi$ is the change in parity in the transition.

In the transitions with $|\Delta I| = n + 1$ only the Gamow-Teller interaction is effective and these transitions are usually designated "unique" forbidden.

It turns out that the theoretical expression for the transition probability of an nth-forbidden transition can be obtained by multiplying eqn. 3 by a so-called "shape factor" $S_n(W)$, which depends on the degree of forbiddenness. It is possible in certain cases to determine the type of a transition from observations on the deviation of the beta-spectrum from the allowed shape. S_n is in general a complicated function in which the energy dependence is determined by the relative magnitudes of two or more generally unknown nuclear matrix elements, and the interpretation of observed forbidden spectrum shapes is in most cases not unique. For the unique forbidden transitions with $|\Delta I| = n + 1$ and Gamow-Teller coupling only, the theoretical energy dependence is unambiguous and has been experimentally verified for n = 1, 2, 3 (16-18), thus providing strong evidence for the existence of the Gamow-Teller coupling.

In the case of the first-forbidden non-unique transitions it has been found that the beta-spectra in general have allowed shapes (19,20). However, more accurate work on high energy spectra might show small deviations from the allowed shape.

D. ft VALUES.

The half-life t is given by $t = \ln 2/\lambda$, where λ is the decay constant of the beta-transition and is obtained by integrating eqn. 3 i.e.

$$\lambda = \int_1^{W_0} P(W)\,dW = \frac{g^2}{2\pi^3}\left(|C_F|^2|M_F|^2 + |C_{G-T}|^2|M_{G-T}|^2\right) f(Z W_0) \qquad \text{-----} (5)$$

where $$f(Z,W_0) = \int_1^{W_0} pW(W_0-W)^2 F(Z,W)\,dW$$

and the Fierz interference term is neglected.

Then, for an allowed transition, the product

$$ft = \frac{B}{(1-x)|M_F|^2 + x|M_{G-T}|^2} \qquad \text{-------} (6)$$

where $x = |C_{G-T}|^2 = 1 - |C_F|^2$, and $B = \frac{2\pi^3 \ln 2}{g^2}$ are universal constants, is a function only of the nuclear matrix elements, and, since these would be expected to be of the same order of magnitude for all allowed transitions, so also would the ft values.

It has been found (21,22,23) that the observed ft values of beta-transitions fall into roughly defined groups corresponding to the degree of forbiddenness of the transitions, indicating that for a given order of forbiddenness there is no great variation in the magnitudes of the nuclear matrix elements. In addition it is found that the ft values of allowed transitions break up into two groups, with log ft from 4.0 to 6.0 and about 3.5 , which are referred to as normal allowed and super-allowed respectively. The latter include the "mirror" transitions in which $(N-Z) = \pm 1$ for both initial and final nuclei, and also some transitions for which $(N-Z) = \pm 2$ for one nucleus in the transition, and $N = Z$ for the other. It is assumed that in the super-allowed transitions the initial and final nuclear wave functions are very much alike, and overlap almost completely, leading to a very large matrix element.

The observed ft value has become a useful addition to the criteria to be adopted in the identification of the type of a previously unclassified transition.

E. THE BETA-INTERACTION.

As regards the character of the beta-decay interaction, strong evidence for the presence of the Fermi interaction is obtained from the existence of the group of ($0 \rightarrow 0$, no) beta-transitions (O^{14}, C^{10}, Cl^{34}, Al^{26}), which are strictly forbidden by the Gamow-Teller selection rules, and allowed only by the Fermi rules.

Similarly, convincing evidence for the presence of the Gamow-Teller interaction is provided by the decay of $He^6 \rightarrow Li^6$ ($\Delta I = 1$, no), which is not allowed by the Fermi selection rules. The existence of the unique forbidden transitions, which take place through pure Gamow-Teller interaction, is an additional argument for this interaction.

Now that both the Fermi and the Gamow-Teller interactions have been shown to contribute to the general beta-decay interaction, it becomes of considerable importance to determine the relative contribution of each of these two. Several attempts have been made to evaluate $|C_F|^2 / |C_{G-T}|^2$ (24-30), and the more recent work fairly consistently supports the conclusion that $|C_F|^2$ is approximately equal to $|C_{G-T}|^2$, with the latter probably slightly larger.

Recently Kofoed-Hansen and Winther (27) have repeated their 1953 analysis, using new experimental data on mirror beta-transitions and on those transitions of type ($0 \rightarrow 0$, no). For each transition they plot a B,x line defined by (cf. eqn. 6)

$$B = ft\left\{(1-x)|M_F|^2 + x|M_{G-T}|^2\right\}$$

The Fermi matrix element M_F can be evaluated for these transitions on the assumption of charge independence of nuclear forces only, while a semi-empirical value of M_{G-T} is deduced from the experimental nuclear magnetic moments, according to the Bohr- Mottelson collective model. Using the method of least squares to obtain the common intersection point of the B,x lines, they obtain the values

$$B = 2787 \pm 70$$

$$x = 0.560 \pm 0.012$$

Having established that $|C_F|^2 \simeq |C_{G-T}|^2$ it remains to determine the relative importance of the S and V terms in the Fermi interaction and of the T and A terms in the Gamow-Teller interaction. This has been done by effectively measuring the angular correlation between the directions of emission of the electron and the neutrino in the beta-decay of the rare gas isotopes He^6, Ne^{19}, Ne^{23}, and A^{35}, and in the decay of the neutron.

From their measurements on He^6, which is a pure Gamow-Teller transition, Rustad and Ruby (31,32) have concluded that the tensor interaction is dominant in this decay, and this result has been confirmed by Allen and Jentschke (33).

The results obtained by several groups for the mixed Fermi and Gamow-Teller positron transition in Ne^{19} could be explained by either the S,T or A,V combination of interactions (34-36), and similar conclusions can be drawn from Robson's results on the neutron decay (37).

More recently Hermannsfeldt et al. (38) have measured the angular correlation coefficient for the positron emitter A^{35}. This is expected to be predominantly a Fermi decay, and the results obtained indicate that in the Fermi interaction the vector term is at least twice as large as the scalar. This result, if combined with those of the Ne^{19} experiments, requires that the A,V combination be dominant in the beta-decay interaction, and is difficult to reconcile with the apparently unambiguous He^{6} result.

In an attempt to resolve this difficulty Ridley (39) has determined the electron-neutrino angular correlation coefficient for the negatron transitions in Ne^{23}, which are expected to occur mainly through the Gamow-Teller matrix element. His results are consistent with the presence of about equal amounts of T and A in the Gamow-Teller interaction.

In chapter 8 these rather confusing results are compared with those obtained in the recent experimental and theoretical studies of parity non-conservation.

2. THE RATIO OF K-CAPTURE TO POSITRON EMISSION.

A. THEORETICAL.

Yukawa and Sakata (40) pointed out in 1935 the possibility that an unstable nucleus could undergo a radioactive transition to a stable isobaric nucleus by capturing one of its orbital electrons, and the phenomenon was first observed by Alvarez (41) in 1937.

The decay constant can be calculated in a similar way to that for beta-emission, and, for electron-capture from the K-shell in an allowed transition, is given by

$$\lambda_K = \frac{g^2}{2\pi^3} |M|^2 \frac{\pi}{2} (W_0 + W_K)^2 g_K^2(R) (1 + b) \qquad \text{------} \; (7)$$

where $|M|^2 = |C_F|^2 |M_F|^2 + |C_{G-T}|^2 |M_{G-T}|^2$ and b is the Fierz interference term. W_K is the K-shell electron energy, including rest mass, i.e. $W_K = 1 - \mathcal{E}_K$, where $\mathcal{E}_K$ is the K-shell binding energy. $g_K^2(R)$ represents the probability of finding a K-shell electron at the surface of the nucleus, g_K being the radial part of the "large" component of the K-electron wave function, evaluated at the nuclear radius R, and obtained by solving the Dirac relativistic wave equation for the K-electron in the Coulomb field of the nucleus. g_K^2 is given by

$$g_K^2(R) = \frac{1+\gamma}{2\Gamma(2\gamma+1)} R^{2\gamma-2} (2\alpha Z')^{2\gamma+1} \qquad \text{------} \; (8)$$

where $\gamma = W_K$, and $Z' = Z - \sigma$, σ being a measure of the screening effect of the other orbital electrons. For the K-shell σ is approximately equal to 0.3 (42), but more accurate values have been calculated for several values of Z (43). Z in the above formulae represents the atomic number of the initial nucleus. Γ is the gamma function and α is the fine structure constant.

The ratio of K-capture to positron emission for allowed transitions is of particular importance from the point of view of beta-decay theory, since the quantities which are at present the least well known, namely the Fermi constant g and the nuclear matrix element M, are eliminated when this ratio is formed. Graphs of this ratio for allowed transitions have been given, for several Z values, by Feenberg and Trigg (21), who used Coulomb wave functions for the orbital electrons. These results require to be corrected for the screening effect of the orbital electrons on the probability of positron emission. Unfortunately, because of the very rapid variation of the K/β^+ ratio with energy and with Z, it is difficult to obtain values from these curves by interpolation, and for accurate work it is advisable to use the above formulae (eqns. 3,7,8), applying the corrections for screening. (Tables of the Fermi function F are given in ref. 1).

Recently Zweifel (44) has calculated values of the allowed K/β^+ ratio for five Z values, including corrections for screening, but, again, the interpolation is unreliable for accurate work. These values have since been recalculated (45) including a correction for the effect of the finite nuclear size on the bound electrons. This correction, which is negligible at low Z, reduces the branching ratio by about 10 % at Z = 84.

Little theoretical work has been done so far on the K/β^+ ratio for forbidden transitions, mainly because a rigorous comparison with experimental results is in general not possible, owing to the lack of accurate information on the relative magnitudes of nuclear matrix elements. However, expressions have been presented by several workers (44,46-48), and some conclusions can be drawn from these. For first forbidden ($|\Delta I| = 0, 1$, yes) transitions the K/β^+ ratio is expected to be about the same as for allowed transitions, on the basis that the beta-spectra of such transitions have allowed shapes.

In the case of the unique forbidden transitions the presence of only the Gamow-Teller interaction allows an unambiguous theoretical value to be calculated. For the positrons in this case there is the energy dependent shape factor mentioned earlier to be taken into account, and this can be approximated by a simple polynomial in the positron and neutrino momenta. The K-capture decay constant has also to be multiplied by a correction factor, which takes the form of an even power of the neutrino momentum. The factor by which the allowed value of the K/β^+ ratio has to be multiplied turns out to be $\frac{2(W_o+1)}{W_o-1}$ in the case of a first forbidden unique transition, and the ratio is clearly considerably larger in this case. For higher unique forbidden transitions this increase is considerably intensified with increasing forbiddenness.

B. EXPERIMENTAL DATA.

Two methods have been used in general for the measurement of the K/β^+ ratio. In one of these the total number of positrons emitted in a given time interval is subtracted from the total number of transitions in this interval to give the number of transitions taking place by electron-capture. In practice this usually involves comparing the number of positrons going to an excited state of the product nucleus with the number of gamma-rays emitted from this state. In this type of experiment the total probability of electron-capture from all possible shells is obtained, and the comparison with theory is complicated by the present uncertainty regarding the validity of the theoretical results for the relative probabilities of capture from the various shells (cf. section 4). The total capture-positron branching ratio will be denoted by ϵ/β^+.

The other method of determining the K/β^+ ratio can be used for simple transitions where no gamma-ray is available for the above technique, and consists of measuring directly the number of K-capture events and the total number of transitions. Since this can only be done by measuring the K x-radiations emitted when the vacancy in the K-shell of the final atom is filled, this method has been used less often than the other. However, the proportional counter, with gaseous source, can be used for such measurements and, provided that the energy of the x-radiations is less than about 5 keV, corrections for absorption, scattering, geometry and efficiency can be made very small. The K/β^+ ratio is obtained directly from the observed pulse spectrum.

The ratio of K-capture to positron emission has been determined for a number of nuclei and the results have been reviewed by several authors (44,49,50). For the majority of the transitions surveyed the experimental K/β^+ ratios are fairly close to the theoretical values. However, the comparison with theory is not entirely satisfactory since the data concern nuclei with complex decay schemes, not always well established. For example, the nucleus which has excited the most interest as far as the K/β^+ ratio is concerned, namely Na^{22}, decays mainly to a 1.3 MeV excited state of the product nucleus, and in addition the transition may not be of the ordinary allowed type (49,51). However, the results of recent measurements of the ϵ/β^+ ratio for this transition are in good agreement with each other and with the theoretical allowed value, and a review of these experiments illustrates the more usual methods for the determination of this ratio.

Sherr and Miller (51) employed a coincidence arrangement to compare the intensities of positron emission and of gamma radiation, with a 4π beta counter to detect the positrons and a scintillation counter for the gamma-rays. Their result was $\epsilon/\beta^+ = 0.110 \pm 0.006$, which is in good agreement with the theoretical value of 0.1135 .

By measuring the relative numbers of 1.3 MeV gamma-rays and 0.51 MeV annihilation quanta in a 4 in. by 4 in. NaI scintillation spectrometer, Kreger (52) obtained a value of 0.123 ± 0.010 .

Allen et al. (53) compared the rate of evolution of Ne^{22} from a source of Na^{22} with the rate of emission of positrons, and obtained a value of 0.122 ± 0.010 . The rate of evolution of the neon was measured in a gas analysis apparatus, and the rate of emission of positrons was determined by absolute counting in a 4π Geiger counter.

Using thin solid sources of Na^{22} in 2π Geiger counters, Charpak (54) measured the intensity of the 0.85 keV Auger electrons emitted in the K-capture decay, and obtained a value of 0.065 ± 0.009 for the K/β^+ ratio. However, in view of the considerable scattering and absorption of such low energy electrons in even very thin sources, it seems likely that this result is lower than the correct value.

Since Radvanyi's review (50) little experimental work has been done on the capture-positron branching ratio which is accurate enough to justify comparison with the theoretical predictions, and only three experiments are worth mentioning in this respect.

The decay of Sc^{44} has been thoroughly investigated by Blue and Bleuler (55), using a magnetic lens beta-spectrometer and a NaI scintillation spectrometer in coincidence. The ϵ/β^+ ratio for the transition to the first excited state of Ca^{44} was obtained as 0.073 ± 0.017, which is in good agreement with the theoretical value of 0.077, and contradicts the earlier results of Bruner and Langer (56) and of Hibdon et al. (57).

Cook and Tomnovec (58), using a 4 in. by 4 in. NaI scintillation spectrometer, have studied the gamma-rays following the decay of Co^{58}, and, from the observed relative intensities of the nuclear gamma-rays and the annihilation radiation, have obtained a value of 5.9 ± 0.2 for the ϵ/β^+ ratio for the transition to the excited state of Fe^{58}. This is in agreement with earlier measurement of Good et al. (59), and is close to the theoretical value of 5.4 .

Since the present work on F^{18} and C^{11} was carried out, Van Nooijen et al. (60) have determined the ϵ/β^+ ratio in V^{48}, using beta-gamma coincidence measurements with a long lens beta-spectrometer and a NaI scintillation counter. Their result is that $\epsilon/\beta^+ = 0.75 \pm 0.03$, in good agreement with the theoretical value of 0.76 . Hagedoorn and Konijn (61) have also measured this ratio, using a proportional counter in coincidence with a NaI scintillation counter, and find that $\epsilon/\beta^+ = 0.74 \pm 0.02$. However, the decay scheme of V^{48} is complex, and the above results are based on the assumption that no gamma transitions occur between the 3.24 MeV and the 2.3 MeV levels in Ti^{48}. In fact such transitions could occur in up to 5 % of the disintegrations, according to the measurements of Van Nooijen et al.

In view of the complex decay schemes of those nuclei for which the K/β^+ ratio has been measured, and taking into account the indirect methods usually employed, it seemed of some importance that direct measurements should be made on simple allowed transitions, and the experiments on F^{18} and C^{11} were carried out with this in mind.

Very few measurements of the K/β^+ ratio for forbidden transitions have been made, and accurate results are available only for I^{126} and Rb^{84}.

The decay scheme of I^{126} has been thoroughly investigated by Koerts et al. (62), using magnetic beta-spectrometers and NaI scintillation spectrometers with coincidence techniques. The K/β^+ ratios for the various transitions were determined by comparing the intensity of the K x-rays with that of the nuclear gamma-rays. For the first forbidden unique transition to the ground state of Te^{126} the experimental value is 20.2 ± 2.0 , in good agreement with the theoretical value of 21.9 . The value for the ($\Delta I = 0$, yes) transition to the first excited state of Te^{126} is given as 95 ± 10 , which is considerably lower than the theoretical allowed value of 152 .

A very similar investigation has been made of the decay scheme of Rb^{84} by Welker and Perlman (63), using beta and gamma-ray scintillation spectrometers with coincidence measurements. Their result for the K/β^{+} ratio for the first forbidden unique transition to the ground state of Kr^{84} is 2.06 ± 0.36 , which does not agree with the theoretical value of 1.0 . For the ($\Delta I = 0$, yes) transition to the first excited state the value obtained is 5.15 ± 0.38 , which is considerably larger than the theoretical allowed value of 3.1 . This is in contrast with the result for I^{126}. Part of the interest of the present experiment on As^{74} lay in a similar comparison of the observed K/β^{+} ratio for a ($\Delta I = 0$, yes) transition with the theoretical allowed value.

The discrepancy observed above for the unique forbidden transition in Rb^{84} is particularly serious since for this type of transition only one matrix element is involved, and this is cancelled out when the K/β^{+} ratio is formed. The theoretical results for this type of transition are obtained by simple extensions of the allowed theory, and, in view of the success of this theory, it would seem that they are valid, and that the discrepancy is due to errors in the experiments of Welker and Perlman.

An accurate measurement of the K/β^{+} ratio for a unique forbidden transition would thus be of considerable importance, but these transitions generally occur in very complex decay schemes, and such a measurement would be rather difficult. The author has attempted this measurement for the second forbidden unique transition in Al^{26}, using gaseous sources in proportional counters, but so far has not found a suitable volatile compound of aluminium. Aluminium borohydride and aluminium chloride have been tried, the latter with the counter operating at 200-300°C, but with no success.

3. THE FIERZ INTERFERENCE TERM.

A consideration of eqns. 3 and 7 shows that estimates of the magnitude of the Fierz interference term can in principle be obtained from measurements either of beta-spectrum shapes or of K/β^+ ratios, and several such measurements have been made.

Mahmoud and Konopinski (64) adopted the published spectra of Cu^{64}, S^{35} and N^{13} as the most accurate measurements up to that date and, for each case, drew the conventional Kurie plot $K = C(W_0-W)$ and the corresponding "Fierz" plots $K = C(W_0-W)\sqrt{1 \pm b/W}$, using several different values of b. The upper limit on b was obtained from that Fierz plot in which the systematic mean deviation was just greater than the random deviation in the conventional Kurie plot. These authors concluded that the upper limit for b obtained in this way is about 0.2 for both the pure Gamow-Teller transitions and the mixed Fermi and Gamow-Teller transitions.

A statistical analysis of several allowed and first forbidden beta-spectra has been made by Davidson and Peaslee (65), and the results of this analysis for the allowed spectra have been studied in order to determine the magnitude of the Fierz term. The result for three pure Gamow-Teller transitions is that $b_{G-T} = 0.00 \pm 0.08$, where

$$b_{G-T} = 2\gamma\,\mathcal{R}e\left\{\frac{(C_T C_A^* + C'_T C'^*_A)|M_{G-T}|^2}{|C_{G-T}|^2|M_{G-T}|^2}\right\}$$

Of those transitions in which the Gamow-Teller contribution is considerably smaller than the Fermi part only N^{13} was considered to have had its spectrum measured with the necessary accuracy, and from this spectrum b_F (cf. b_{G-T}) was found to be less than 0.2 .

Pohm, Waddell and Jensen (66) have examined the beta-spectrum of P^{32} for any deviation due to the Fierz term, using both an intermediate image spectrometer and a thin lens spectrometer. The value of b_{G-T} was found to be 0.00 ± 0.03 .

More recently this spectrum has been remeasured, in a double lens spectrometer, by Porter, Wagner and Freedman (67), who found that $0.05 < b_{G-T} < 0.093$. This result, which does not include a zero value for b_{G-T}, is in disagreement with that of Pohm et al., and might indicate a non-zero value for the Fierz term. However, these authors consider it more likely that the non-linearity of the P^{32} Kurie plot can be explained by the presence of second-order effects in the spectrum, and point to the high ft value for this decay to support this view. These workers have also obtained the result $-0.02 < b < 0.02$ from measurements on the beta-spectrum of Na^{24}, which has both Fermi and Gamow-Teller terms.

It is seen that the results of spectrum measurements lead consistently to the limits for the Fierz interference terms $|b_{G-T}| \lesssim 0.05$ and $|b_F| \lesssim 0.20$. However, limits on these terms can be more accurately estimated by comparing the measured K/β^+ ratio for certain transitions with the theoretical value, and part of the interest in the present work lay in this comparison.

From eqns. 3 and 7 the K/β^+ ratio is given by:

$$\frac{\lambda_K}{\lambda_{\beta^+}} = \frac{\frac{g^2}{4\pi^2}(W_0+W_K)^2 g_K^2 |M|^2 (1+b)}{\frac{g^2}{2\pi^3}\int_1^{W_0} F(-Z,W)\, pW(W_0-W)^2 |M|^2 \left(1-\frac{b}{W}\right) dW} \quad \text{------ (8)}$$

If this ratio is denoted by R and the ratio calculated for b = 0 by R_0 then

$$R = \frac{R_0(1+b)}{1-b\langle W^{-1}\rangle} \quad \text{------ (9)}$$

where

$$\langle W^{-1}\rangle = \frac{\int_1^{W_0} F(-Z,W)\, pW(W_0-W)^2 \frac{1}{W}\, dW}{\int_1^{W_0} F(-Z,W)\, pW(W_0-W)^2\, dW} \quad \text{------ (10)}$$

b is then given by

$$b \doteqdot \frac{R-R_0}{R_0(1+\langle W^{-1}\rangle)} \quad \text{---- (11)}$$

using $R \doteqdot R_0$

The magnitude of b is then determined by substituting in eqn. 11 the experimental and theoretical values for R and R_0 respectively.

The first study of this type was carried out by Sherr and Miller (51), who, by analysing the results of their measurements on Na^{22}, found that $b_{G-T} = -0.02 \pm 0.04$. However, as these workers point out, the lack of knowledge of the exact character of the Na^{22} transition weakens the validity of their result. Recent work by Zweifel (44) has shown that, when certain correction factors are applied to the theoretical value of the K/β^+ ratio for this decay, the small difference observed by Sherr and Miller between the experimental and theoretical results becomes much smaller, and the amended result is $b_{G-T} = 0.00 \pm 0.04$.

Since the completion of the work described in this thesis, an estimate of the limits on b_{G-T} has been obtained by Hagedoorn and Konijn (61) from their measurements on V^{48}. These workers found that $b_{G-T} = -0.02 \pm 0.02$, but the significance of this result is somewhat weakened by the assumption on which it is based.

Another method by which limits can be placed on the magnitudes of the Fierz terms involves the consideration of the B,x lines (cf. p.12), with various values of b_{G-T} and b_F i.e. - - - - -- (12)

$$B = ft\left\{(1-x)(1 \pm b_F)\langle W^{-1}\rangle |M_F|^2 + x(1 \pm b_{G-T})\langle W^{-1}\rangle |M_{G-T}|^2\right\}$$

If it is assumed that the B,x lines should pass through a common point, limits can be placed on b_{G-T} and b_F. This has been done by Kofoed-Hansen and Winther (27), in their survey of the most recent experimental data, and, from considerations of the internal consistency of their B,x diagram, these authors find that the best fit of the data is obtained if they assume $b_F = 0.29 \pm 0.21$.

Unfortunately, it is more difficult to measure b_F than b_{G-T}, since very few pure Fermi transitions are available for such a measurement, and their short half-lives make accurate work on them very difficult. However, subsequent to the present work on C^{11}, Gerhart (68) has obtained an estimate of b_F by plotting $\dfrac{2\pi^3 \ln 2}{ft\,|M|^2}$ as a function of $\gamma\left\langle\dfrac{1}{W}\right\rangle$ for several ($0 \rightarrow 0$, no) transitions (O^{14}, Al^{26}, Cl^{34}). The matrix elements can be computed with quite high accuracy, and the slope of this plot gives the Fierz term b_F. Gerhart concludes that $b_F = 0.00 \pm 0.24$.

Part of the interest in the present measurements on F^{18} and C^{11} lay in the possibility of obtaining sufficiently accurate values of the K/β^+ ratios to enable an estimate to be made of the magnitudes of b_{G-T} and b_F, the latter being the more important in view of the large limits previously set.

At the time when the present experiments were started the expression for b was $$b = 2\gamma \frac{C_S C_V^* |M_F|^2 + C_T C_A^* |M_{G-T}|^2}{(C_S^2 + C_V^2)|M_F|^2 + (C_T^2 + C_A^2)|M_{G-T}|^2} \quad \text{-----} \quad (13)$$

and it is seen that a small value of b implied a small value for $C_S C_V$ and $C_T C_A$. The results of the electron-neutrino angular correlation experiments (pp. 12,13), together with the data on the Fierz term, led to the conclusion that the beta-decay interaction was mainly T and S, and that the limits on the contributions from V and A were $C_V/C_S = 0.00 \pm 0.15$, $C_A/C_T = 0.00 \pm 0.02$. However, the presence of the parity non-conserving terms in the expression for b (eqn. 4) now means that a small value for b no longer necessarily implies small values for any of the coupling constants, and at the present time several different conclusions could be drawn. A brief discussion on these conclusions, and on the theoretical and experimental results on which they are based, is given in chap. 8.

4. THE L/K-CAPTURE RATIO.

A. THEORETICAL.

For allowed transitions capture is possible from the L_I and L_{II} sub-shells, and of these the L_I sub-shell normally makes much the larger contribution. The decay constant for this process is similar to that for K-capture, being given by

$$\lambda_L = \frac{g^2}{2\pi^3} |M|^2 \frac{\pi}{2} (W_0 + W_L)^2 (g_{L_I}^2 + f_{L_{II}}^2)(1 + b) \quad \text{----} \quad (14)$$

where $W_L = 1 - \varepsilon_L$, and the difference between the L_I and L_{II} binding energies has been neglected. $f_{L_{II}}$ is the "small" component of the Dirac radial wave function for the L_{II} electrons.

Increasing interest in the L/K ratio has been shown in recent years, and many measurements have been reported. The earliest calculations were carried out by Marshak (46), using relativistic wave functions with Slater screening factors. More recently Rose and Jackson (69) have constructed a graph of $g_{L_I}^2 / g_K^2$ as a function of Z, using Hartree self-consistent field wave functions for low values of Z, and relativistic wave functions for a Thomas-Fermi atom for other values. This can be used to calculate L_I/K ratios for allowed transitions.

The most comprehensive theoretical work on this subject has been carried out, with the aid of an electronic computer, by Brysk and Rose (48), who have calculated wave function ratios using relativistic wave functions, corrected for screening, for variations in the electron wave functions over the nuclear volume, and for the finite size of the nucleus. These results can be used to calculate capture probabilities from the K-shell and from the three L sub-shells.

Results are also presented in this paper for forbidden electron-capture transitions. It turns out that, provided the energy available for the transition is well above the K-shell binding energy, the L_I/K ratio is independent of the order of forbiddenness, and is just the allowed value. Near the K-capture threshold however, the ratio depends strongly on the neutrino momenta for the two shells, and on the relative magnitudes of nuclear matrix elements, where there are more than one of these. With the same restrictions on the energy as above it is also shown that the L_{II}/L_I ratio is insensitive to the order of forbiddenness. For the L_{III} sub-shell it is found that, for $|\Delta I| = 0,1$ transitions the contribution is negligible, while for $|\Delta I| \geq 2$ the contribution can be comparable to that of L_{II}.

It is seen then that, for first forbidden (ΔI = 0,yes) transitions such as those in As^{74} and I^{126}, where several different matrix elements are involved, the theoretical L/K ratio is a complicated function of these matrix elements, of the components of the Dirac radial wave functions for L and K electrons, and of the momenta of the neutrinos emitted in L and K capture. However, provided that the transition energy is much greater than the K-shell binding energy, the L/K ratio associated with each matrix element has the same value as the theoretical allowed L/K ratio within a few per cent (for I^{126} and As^{74} about 2 %), as a consequence of the equality of the ratios of the "large" and "small" components of the Dirac wave functions, and the actual magnitudes of the nuclear matrix elements have little effect on the total L/K ratio.

B. EXPERIMENTAL DATA.

A comprehensive review of experimental work on L/K ratios, up to 1955, has been given by Robinson and Fink (70, cf. also 50,71-73). Two general methods have been used for the measurement of the L/K ratio. In the first of these the radioactive source is situated outside the sensitive volume of the detector, which is in general a scintillation or proportional counter, and the relative intensities of K and L x-rays are measured. Large corrections have to be applied for K and L-fluorescence yields, for absorption and scattering of the x-rays in the source itself and in the path from source to detector, and for the efficiency of the detector. Other external source methods have been used when the electron-capture transition is followed by a gamma-ray. In this case coincidence techniques of various kinds can be used to determine the L/K ratio. However, because of the large corrections required, this type of work requires great accuracy if the experimental limits on the final result are to be kept reasonably low.

The majority of the experiments which have been carried out using the external source method are on transitions in which there is no competing positron emission, and for which , in consequence, the transition energy is uncertain. The measured value of the L/K ratio is inserted in the theoretical expression for the ratio, and in this way the transition energy is deduced. Work of this type has been confined to fairly heavy nuclei, good examples being the studies of the Milan group on Cd^{109}(74),Gd^{153}(75,cf.also76),Ta^{179}(77),W^{181} (78),Os^{185}(79),Au^{195}(80). Similar work has been done recently on Np^{235}(81),Bk^{245} (82),Pd^{103}(71),Tl^{202}(83) and Au^{196}(84). This technique has become a valuable alternative to closed cycle calculations of transition energies in many cases where the more accurate methods (e.g. nuclear reaction threshold measurements (85) and inner bremsstrahlung spectra end-point determinations (86)).have not been applied.

Alternatively, where the transition energy is known the observed intensities of L and K x-rays, together with the theoretical L/K ratio, can be used to estimate the value of the L-fluorescence yield, and several such experiments have been carried out (70).

The other method which has been used for the measurement of L/K ratios involves the distribution of the radioactive source throughout the sensitive volume of the detector, which may be either a scintillation counter or a proportional counter. By this means the corrections for geometry, absorption and scattering become small provided that the ranges of the L and K radiations in the counter are small compared with the dimensions.

Der Mateosian (87) has measured the L/K ratio in Cd^{109} and I^{125} by growing NaI crystals to which had been added traces of the radioactive sources. Each of these isotopes decays by electron-capture to a low-lying excited state of the product nucleus, which then decays by gamma-emission to the ground state. The values of the L/K ratios obtained by Der Mateosian, by measuring the areas of the peaks in the pulse spectra, were used to determine the transition energies for the two decays, assuming the validity of the theory.

Very few measurements have been made of the L/K ratio for transitions in which the decay energies are known, and, until recently, the only case where the measured ratio was in agreement with the theoretical value was that of A^{37} (88), which was the first nucleus in which L-capture was observed, and also the lightest nucleus for which this measurement has been made. This experiment was performed by adding a trace of A^{37} to the filling of a proportional counter, and observing the peaks in the pulse spectrum due to the L and K x-radiations of chlorine. The correction for the escape of these low energy radiations is small and can be calculated quite accurately. The result obtained was that the L/K ratio is 0.085 ± 0.005, which is close to the theoretical value of 0.082 .

Using an external source coincidence method with scintillation counters, Welker and Perlman (63) have measured the L/K ratio for a first forbidden ($\Delta I = 0$,yes) transition in Rb^{84}. Their result of 0.12 ± 0.05 is in agreement with the theoretical value of 0.10, but is strongly dependent on the value assumed for the K-fluorescence yield.

Much of the recent work on electron-capture has been carried out by the Paris group, who have measured the L/K ratio in Ge^{71}(89,72) and Kr^{79}(50,90,91). These workers used small proportional counters with propane filling, and introduced the sources into the counters in gaseous form. In these conditions the K_{α} x-rays, which have energies of 9.2 keV and 11.7 keV respectively, have half-distances large compared with the counter dimensions, and the probability of K_α x-ray escape approaches 100%. Accordingly, the observed L-peak is largely due to L x-radiations emitted following K-capture events in which a K_α x-ray has escaped from the counter. The L/K ratio is then obtained from the relative areas of the L and K peaks, using the known value of the K-fluorescence yield. However, this method has the drawback that the final result for the L/K ratio depends very sensitively on the value assumed for the fluorescence yield, which is a quantity on which experimental limits are still quite high (92,93). The results obtained for the L/K ratios in Ge^{71} and Kr^{79} were 0.30 ± 0.02 and 0.26 ± 0.03 respectively, which are in marked disagreement with the theoretical values of 0.106 and 0.101 . These results were later recalculated to be 0.19 and 0.096 (70), using fluorescence yield values given by Broyles et al. (93). However, more recently still, Laberrigue-Frolow et al.(94) have carried out measurements of the K-fluorescence yield in Tc and In, and, taking into account recent determinations of this quantity in heavy elements, come to the conclusion that the L/K ratios have the values 0.25 ± 0.02 and 0.22 ± 0.03 respectively.

The L/K ratio in Ge^{71} has been remeasured recently by Drever and Moljk (95) using a gaseous source in a multi-wire counter (cf. chap. 2)(96). With an argon filling to a pressure of six atmospheres, the ratio of the areas of the L and K peaks in the pulse spectrum from this counter is close to the true L/K ratio, the corrections to be applied being very small. The observed value of the L/K ratio was $0.128 {}^{+.005}_{-.003}$ and, in view of the simplicity and accuracy of this work, it seems likely that the theoretical value of 0.106 is in error.

Recently Odiot and Daudel (97,98) have carried out calculations of correction factors to be applied to the theoretical L/K ratio to take account of the correlations between the positions of the orbital electrons, and this might bring the theoretical value for Ge^{71} into better agreement with experiment. However, at present the calculations have been done only for He, Be and A. For A^{37} the theoretical ratio is increased from 0.082 to 0.10. The A^{37} experiment has been repeated by Langevin and Radvanyi (99), following reports (100-103) that the observed charge distribution on Cl^{37} recoil ions from A^{37} did not appear to be consistent with the reported L/K ratio (88). This repeat experiment resulted in an L/K ratio of $0.092 \begin{smallmatrix}+.01\\-.005\end{smallmatrix}$, which is in agreement with the earlier result, but which does not definitely confirm the validity of the corrected theoretical value.

At present comparatively good agreement between theory and experiment is found only for A^{37}, Ge^{71} and Rb^{84}, the result in the latter case being slightly suspect because of the strong dependence on the fluorescence yield, and more work is required for transitions in which the decay energy is known. In particular, it would be of considerable importance to make accurate measurements for heavier elements, since considerable reliance is placed on the validity of the theory in this region to deduce transition energies and L-fluorescence yields. The present work on I^{126} was carried out with this in mind.

The experiment on As^{74} was undertaken in order to measure the L/K ratio and incidentally to show that, by a simple modification of their technique, the French workers (50,72) could have obtained results which would not have been so strongly dependent on the values of K-fluorescence yields.

CHAPTER TWO.

APPARATUS.

1. THE WALL EFFECT IN PROPORTIONAL COUNTERS.

The introduction of the internal gaseous source technique has led to the extensive use of proportional counters in very low energy beta and gamma-ray spectroscopy (104,105). In particular, for accurate work involving low energy Auger electrons and x-rays, this is the most suitable technique since it eliminates the need for the large corrections for scattering and absorption which make accurate work with solid sources extremely difficult at energies less than about 10 keV. In addition, by virtue of its essentially 4π geometry, the proportional counter with gaseous source has a high counting efficiency which leads to the integration of simultaneous radiations. This is of primary importance in the study of orbital capture, where x-radiations which could accompany beta emission or internal conversion of gamma-rays would be difficult to resolve from the x-radiations associated with orbital capture. With the gaseous source technique this problem does not arise since, in any such multiple event, the ionisation due to the x-radiation would be added to that due to the beta-particle or conversion electron, and the resultant pulse would be larger than that due to the x-radiations alone.

Although the proportional counter, with gaseous source, has proved to be the most powerful spectrometer for investigations at very low energies, the method is not without limitations. In the case where the source is a beta emitter the true pulse spectrum will only be obtained when each beta-particle spends the whole of its energy in the sensitive volume of the counter. In general, however, this will be true only at quite low energies. At higher energies a proportion of the particles will escape from the counter gas without dissipating their total energy, and will give rise to pulses corresponding to electrons of lower energy. Consequently the observed pulse spectrum will not be an accurate representation of the true beta-spectrum unless the dimensions of the counter are large compared with the range of the highest energy electrons emitted, or recourse is had to axial magnetic fields (106). Since the continuous beta-particle pulse spectrum rises steeply at low energies due to this "wall effect", the resulting distortion is particularly serious when other low energy phenomena, such as orbital capture radiations, are being studied at the same time. If the latter are of low intensity relative to the beta emission it is quite likely that they will escape detection because of this effect.

Clearly, reductions in wall effect could be obtained by using a sufficiently large counter and high pressure, but limitations are set by the high voltage necessary to give a sufficiently high electrical field strength near the counter wall, and by impurities in the filling

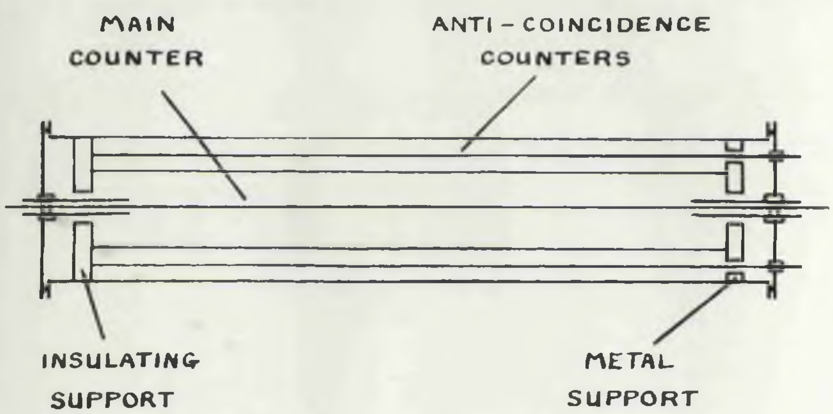


Fig. 1. General arrangement of the multi-wire counter.

gas.

Recently Drever et al. (96), in this laboratory, have constructed counters in which the above-mentioned wall effect is almost completely eliminated. The main counter is surrounded by a second proportional counter system in anti-coincidence; all the counters being enclosed in one metal case. The counting volume of the main counter is defined by a ring of wires joined to the case, and further wires divide the layer of gas surrounding the central counter into separate proportional counters of approximately square cross-section. Beta-particles escaping from the central counter are detected in the anti-coincidence ring counters, the pulses from which are used to gate the pulses from the central counter. The author has constructed a counter of this type and its design and operation are described below.

Another type of counter has also been constructed here, by the author and Dr. G.M. Lewis, which employs scintillation techniques to reduce wall effects. In this counter the gas envelope consists of scintillating material optically coupled to a photomultiplier. Particles which leave the counter gas lose the remainder of their energy in the plastic scintillator and the resulting photomultiplier pulses are used to eliminate, by means of an anti-coincidence gate, the corresponding proportional counter pulses.

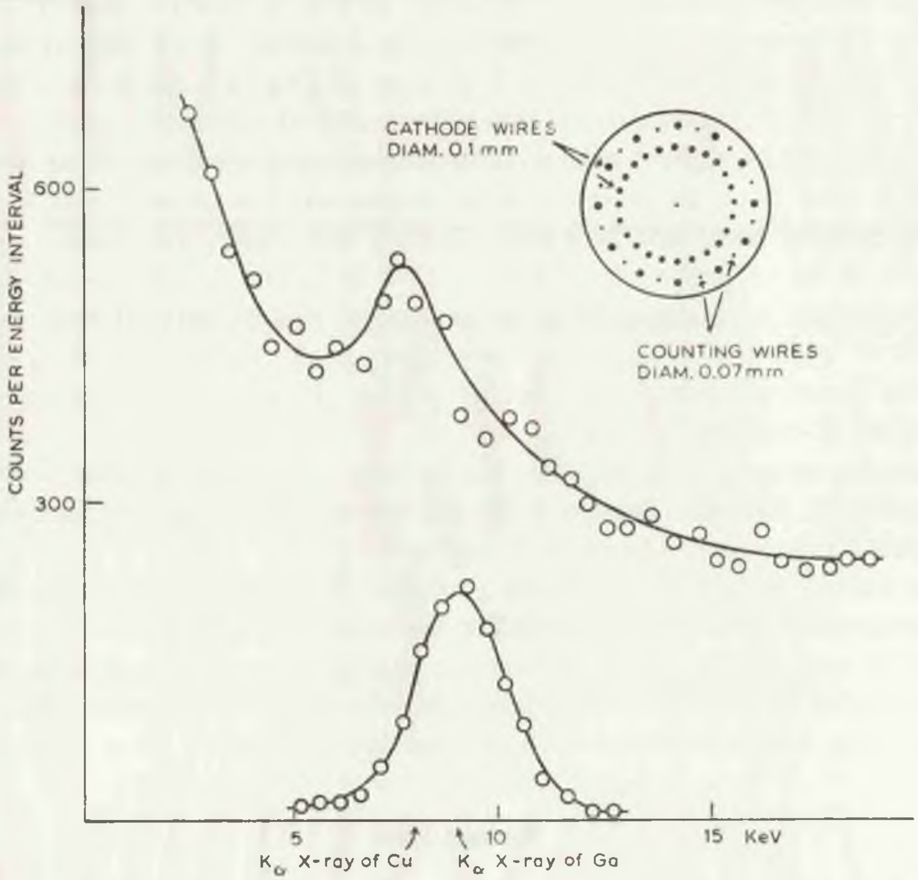


Fig. 2. Part of the proportional counter pulse spectrum of As^{76}. The lower curve is the peak due to the 9.2 keV X-rays from an external Ge^{71} source. Inset is a cross-section of the proportional counter used.

Fig. 2.

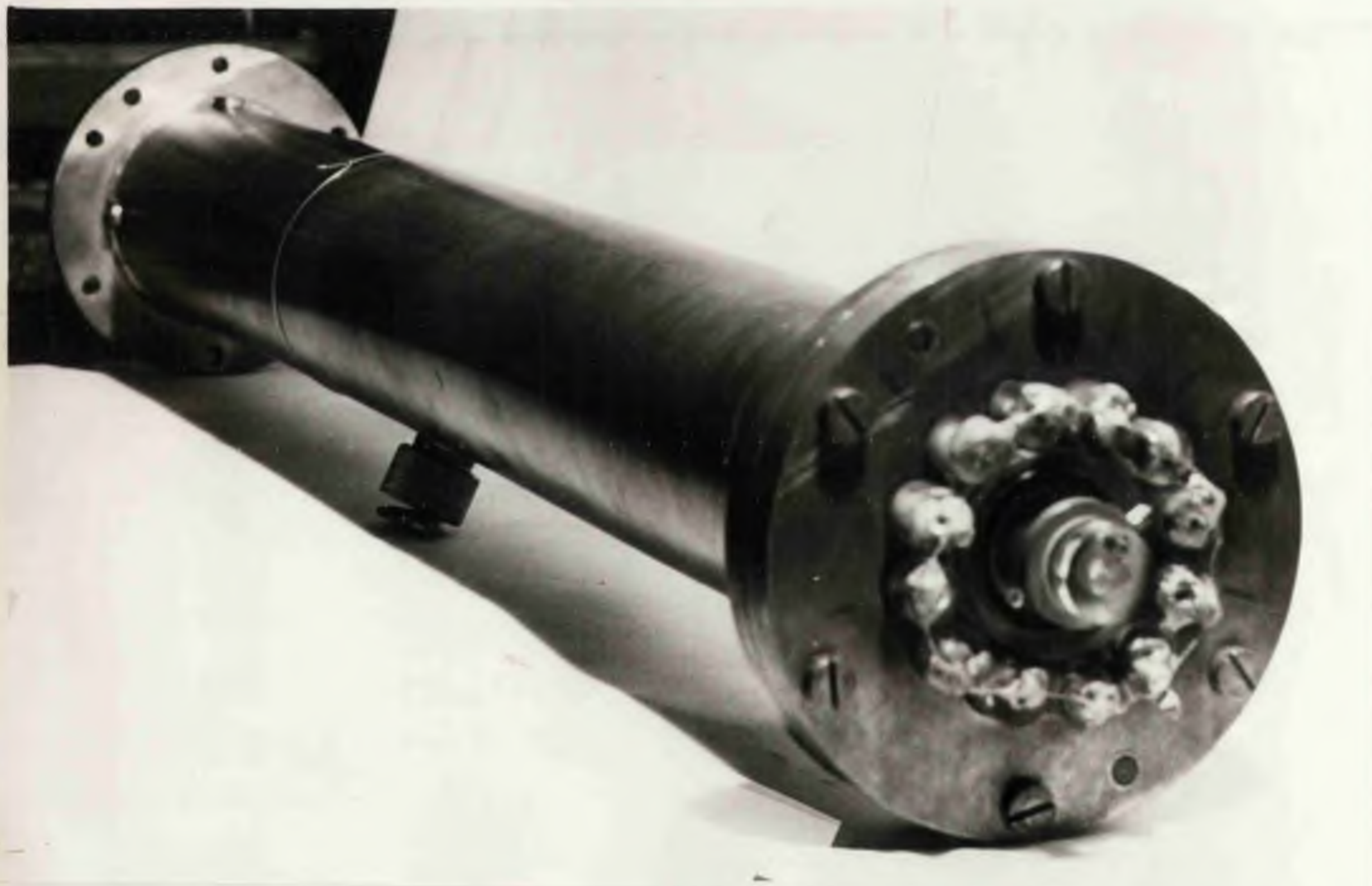
2. THE MULTI-WIRE COUNTER.

(a) Construction.

The general construction of the counter is illustrated in fig. 1 and a cross-section is shown in fig. 2 (inset). The total diameter is 3 in., and the sensitive length is 21 in. The cathode of the central counter consists of 24 tungsten wires of diameter 0.1 mm. arranged in a circle of diameter $1\frac{3}{4}$ in., and connected electrically to the case. The volume between this circle of wires and the case is divided up into twelve separate proportional counters by a further 12 tungsten wires of diameter 0.1 mm., which, together with the inner circle of wires, form the cathodes of the anti-coincidence counters; the anodes of these counters, as well as of the central counter, being tungsten wires of diameter 0.07 mm.

The cathode wires are supported at one end of the counter by a brass ring screwed to the counter wall, and at the other are kept taut by individual springs set into a Perspex ring. At this end the anode wires are similarly mounted, and at the other run through holes in the metal ring, and then through individual Perspex plugs, which are screwed into the end-plate of the counter. Each plug is provided with a separate neoprene 'O'-ring, and the anode wires run through 0.02 in. diameter holes in these plugs. The wires are then attached to screws set into the plugs and the holes sealed with wax.

The anodes of the twelve ring counters are connected together



Multi-wire counter.

externally so that the anti-coincidence ring operates as a whole. However, it might possibly be useful to have them operating separately, and with the above arrangement this is possible.

The anode wire of the central counter is at one end spring-connected to a Perspex insulator fitted with an 'O'-ring, and at the other end brought out through a 0.02 in. hole in a similar insulator and fixed to a screw on the latter. Brass guard tubes, maintained at earth potential, are set into the Perspex insulators and prevent the generation of any spurious pulses due to electrical breakdown of the insulators.

The 9.2 keV x-rays from an external Ge^{71} electron capture source are used for energy calibration, and a Perspex window is provided for this purpose.

A photograph of the multi-wire counter, showing the Perspex insulating plugs for the ring anodes, is attached opposite.

Since the anti-coincidence counters overlap the central counter at both ends by a few inches the sensitive volume of the latter is almost entirely surrounded by the anti-coincidence system, and very few ionising particles will be able to escape from or enter into the central counter without being detected in the ring counters. Gamma-rays of course can pass through the ring without being detected, but these are counted with very low efficiency. It is evident that with such an arrangement very low backgrounds should be expected, and, in fact, the

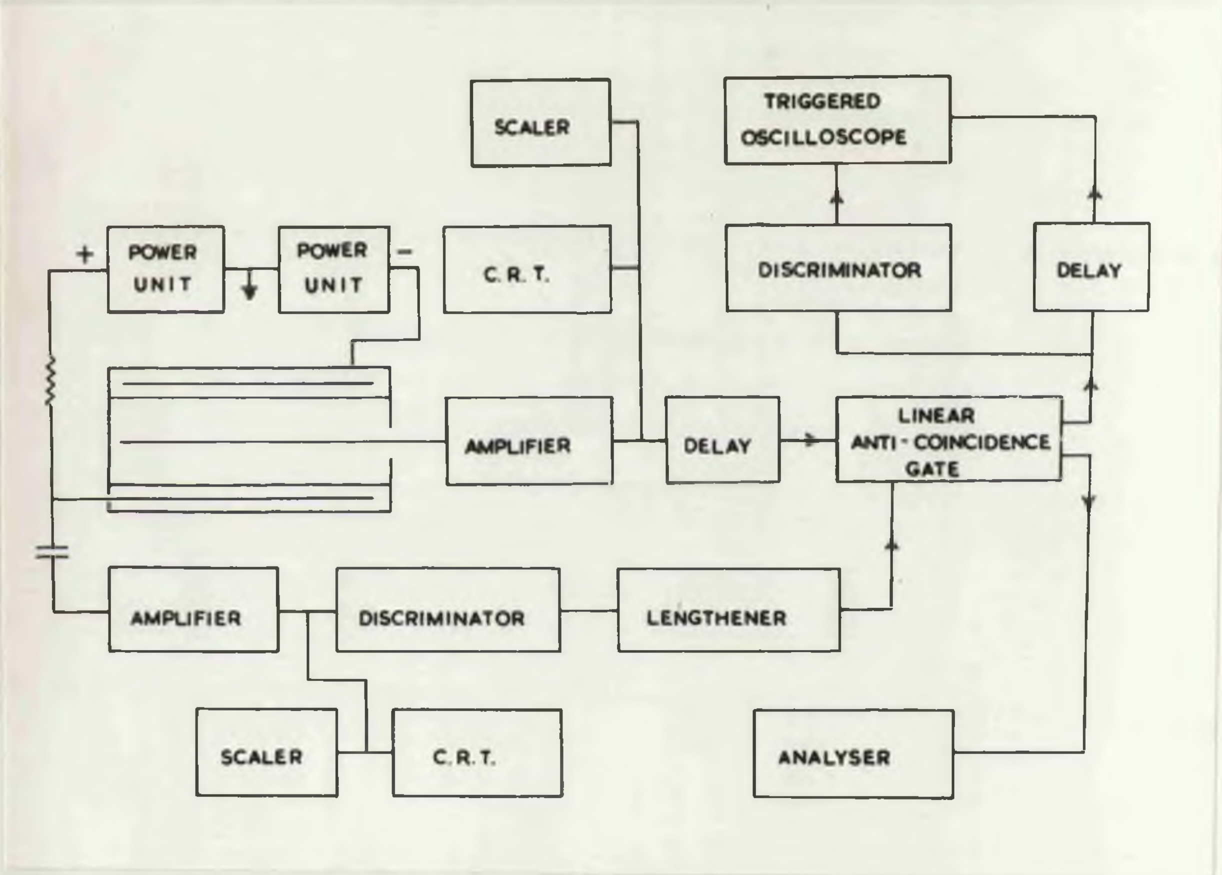


Fig. 3. Block diagram of the electronic equipment used with the multi-wire and plastic counters.

large wall-less counter constructed by Moljk et al. (107) for gas counting of C^{14} has a background of about 2 counts/min. for a sensitive volume of 5.6 litres, which is a considerable improvement on conventional techniques.

(b) Electronics.

A block diagram of the electronic equipment used in the experiment on C^{11} is given in fig. 3. With the exception of the triggered oscilloscope this is the equipment normally used with the wall-less counter.

In order to enable the gains of the central and ring counters to be adjusted independently, a separate E.H.T. power supply is connected to the anodes of the ring counters.

The pulses from the central counter are fed into a high gain low noise linear amplifier and through a 20μsec. delay line to a linear anti-coincidence gate (fig. 4). Pulses from the ring counters are similarly amplified and fed to an amplitude discriminator with an output pulse of width 1μ sec. and height 15 v. This is fed to a double Schmitt circuit and lengthener, emerging as a pulse of amplitude 80 v. and length 200μ sec. This final pulse is fed to one grid of a cathode-coupled double triode, and the pulse from the central counter is applied to the other grid. The arrival of a gate pulse at the left-hand grid has the effect of driving the right-hand valve to cut-off, and suspending its normal action as a cathode follower. This anti-coincidence circuit is

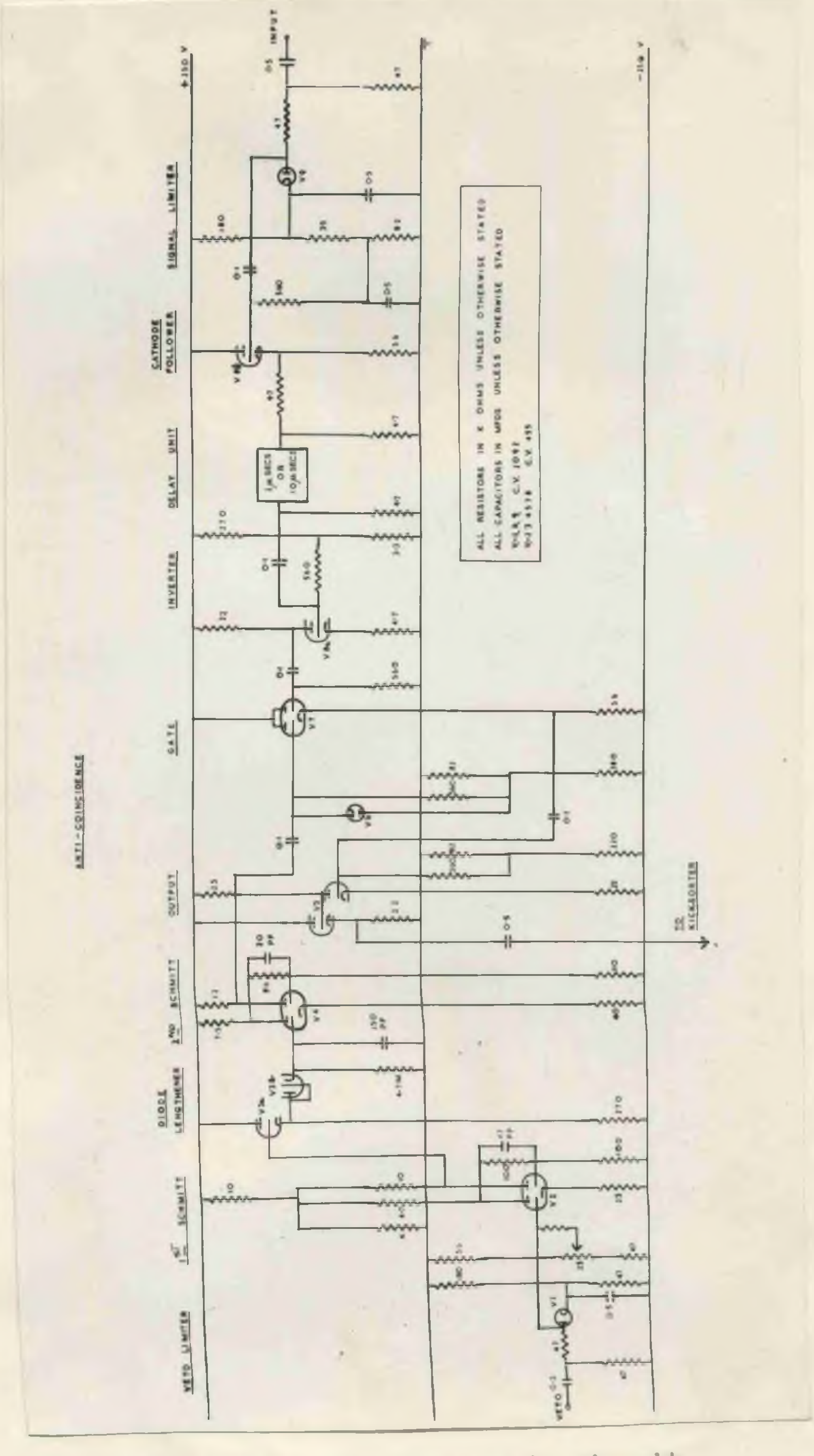


Fig.4. Anti-coincidence gate circuit.

a modification, by the author, of one designed by Drever et al. and is so devised that the gate closes at the beginning of a pulse from the ring counters, and remains closed for the length of the pulse and for a further 200 μsec.

Pulses which pass through the gate are fed to scalers, oscilloscopes, and, when necessary, to a Hutchinson-Scarrott 100-channel kicksorter.

(c) Operation.

Before using the multi-wire counter in the experiments on As^{76} and C^{11} the effectiveness of the anti-coincidence system and the energy resolution of the counter were tested.

The counter was filled with an argon-methane mixture (about 5-10 % methane) to a pressure of two atmospheres, and the natural background in the laboratory without shielding was found to be 610 counts/min. With the anti-coincidence system switched in the background fell to 21 counts/min. With the counter shielded by about 2 in. of lead these figures were 290 c/min. and 10c/min. respectively. In these runs all counts corresponding to energies greater than 1 keV were accepted from the central counter, and the anti-coincidence system was set to trigger on all pulses above 1 keV.

Although the energy resolution, as defined by the width of the 9.2 keV x-ray peak from an external Ge^{71} source, was satisfactory, this did not necessarily imply that the resolution obtained with a gaseous source would also be good, since the x-rays were restricted to a very

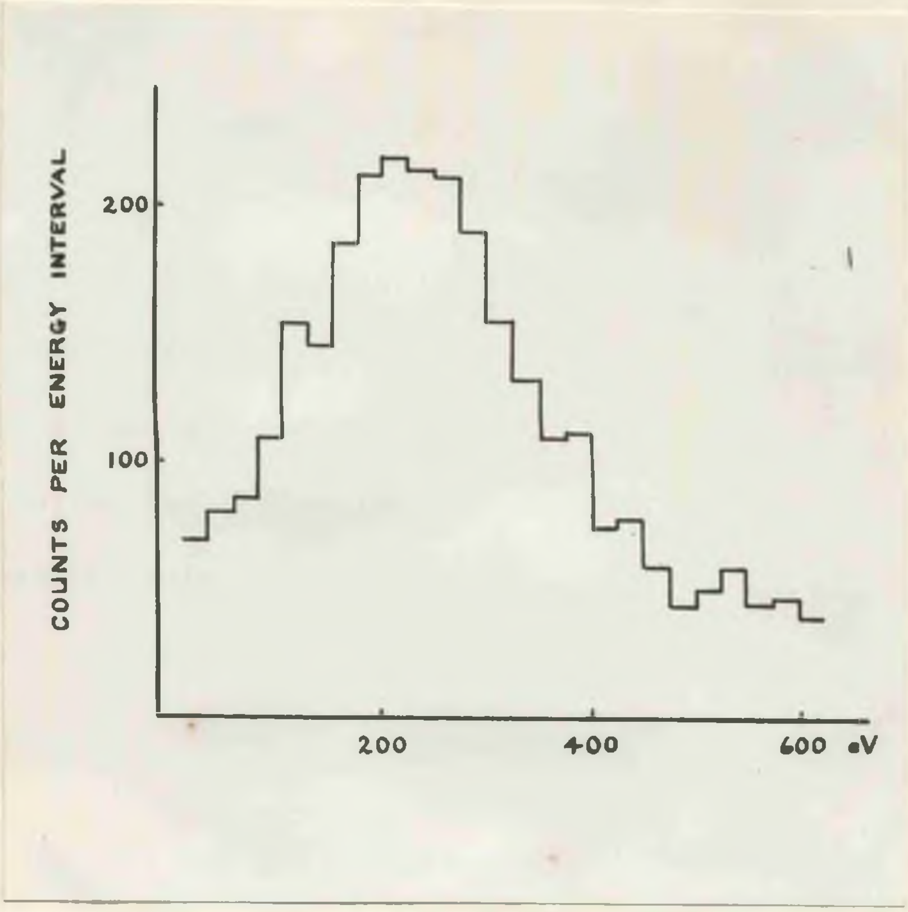


Fig. 5. A^{37} L-peak obtained with the multi-wire counter.

small part of the sensitive volume under the counter window. Accordingly a trace of A^{37} was added to the counter filling and the energy resolution of the 2.8 keV K-peak and 240 eV L-peak was noted and found to be satisfactory. The latter peak is shown in fig. 5. A typical peak due to the 9.2 keV x-rays from an external Ge^{71} source is shown in fig. 2.

It was foreseen that in the experiments on As^{76} and C^{11} it would be necessary to know the ratio of the sensitive volume of the central counter to that of the combined central and ring counters, since, due to the presence of the gaseous source in the anti-coincidence ring, some of the pulses from the central counter are due to particles which originate in the ring and cross over into the central counter. By adding electrically the pulses from the central and ring counters the total number of disintegrations in the counter is obtained, irrespective of the range of the particles. The number of disintegrations in the central counter is then obtained by multiplying the total number by the ratio of the sensitive volume of the central counter to the total sensitive volume. From the dimensions of the counter this ratio is 0.32, and this has been confirmed by a separate experiment with an A^{37} source, which resulted in a value of 0.30 . Since the electron capture radiations from A^{37} have ranges very small compared with the counter dimensions, the counting rates from the two counter systems are quite accurately in the ratio of their sensitive volumes. Since the K-fluorescence yield of chlorine is about 0.07, this fraction of the A^{37} disintegrations

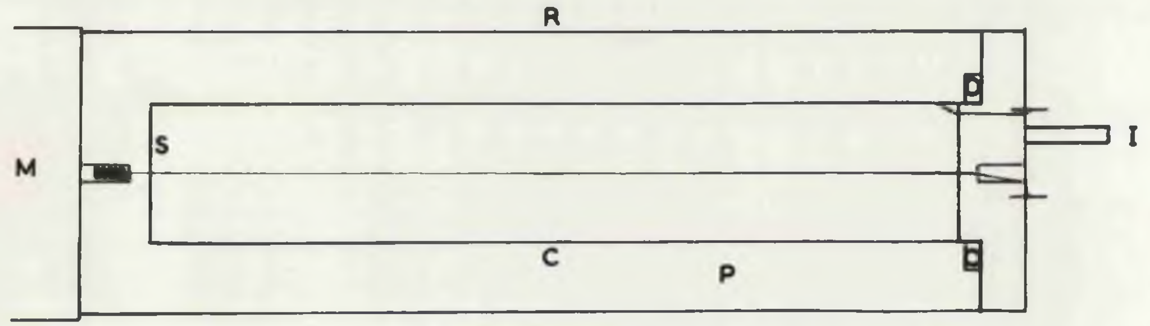


Fig. 6. Construction of plastic counter. C, cathode of aluminium foil; P, plastic scintillator; M, Photomultiplier; S, thin black polythene screen; R, reflector; I, gas input.

gives rise to the emission of K x-rays of chlorine and a small correction has to be made to take account of those x-rays which pass from the ring counter into the central counter and vice versa. This correction was calculated to be 3 % .

3. THE PLASTIC COUNTER.

(a) Construction.

The counter was constructed from a cylinder of plastic scintillator $6\frac{1}{2}$ in. long and 2 in. in diameter. A 1 in. hole was bored in the cylinder to within $\frac{1}{2}$ in. of one end to make a tube of wall thickness $\frac{1}{2}$ in., closed at one end, and a plastic scintillator disc of diameter 2 in. and thickness $\frac{1}{2}$ in. was used to construct a cap for the open end (cf. fig. 6). Six tapped holes were made in the top of the counter, and brass screws passing through holes in the end cap pressed it down on to an 'O'-ring, thus sealing the counter.

Although a coating of evaporated metal would probably give better results, in this case the cathode of the counter was of aluminium foil 0.0002 in. thick, and electrical contact with the high voltage supply was made by means of a wire, soldered to the foil, and passing through a hole in the end cap. The anode wire, which passed through 0.02 in. holes in the plastic at each end, was fixed to a spring at one end of the counter, and at the other end to a screw on the outside of the end cap. All the holes were sealed with Araldite.

Because of the small size of this prototype counter no guard rings were provided. In order to permit the entry of the 9.2 keV x-rays from the Ge^{71} calibration source a $\frac{1}{4}$ in. hole was bored, to within an $\frac{1}{8}$ in. of the inside wall, at about the mid-point of the counter length. However, the wall material being of very low average atomic number, with a reasonably strong source the counter could be calibrated at any point along its length.

Aluminium foil (0.002 in.) was wrapped round the counter to act as a reflector of the light generated in the plastic, and a tube of black polythene was fitted over the outside of this. Due to the presence of the high voltage connection it was not possible to have an aluminium reflector at the capped end of the counter, and, in an attempt to improve light collection, a disc of white Teflon was used here with an outer covering of black polythene. Wrappings of black tape elsewhere made the whole light-tight.

(b) Operation.

The counter was mounted in optical contact with a photomultiplier (Dumont 6292), and some preliminary tests were made on the operation. The same electronics as was used with the multi-wire counter was used with this counter, with the exception that a faster amplifier was used for the photomultiplier pulses than was used for the pulses from the ring proportional counters. Both the proportional counter and the scintillation counter functioned satisfactorily, and, by observing the

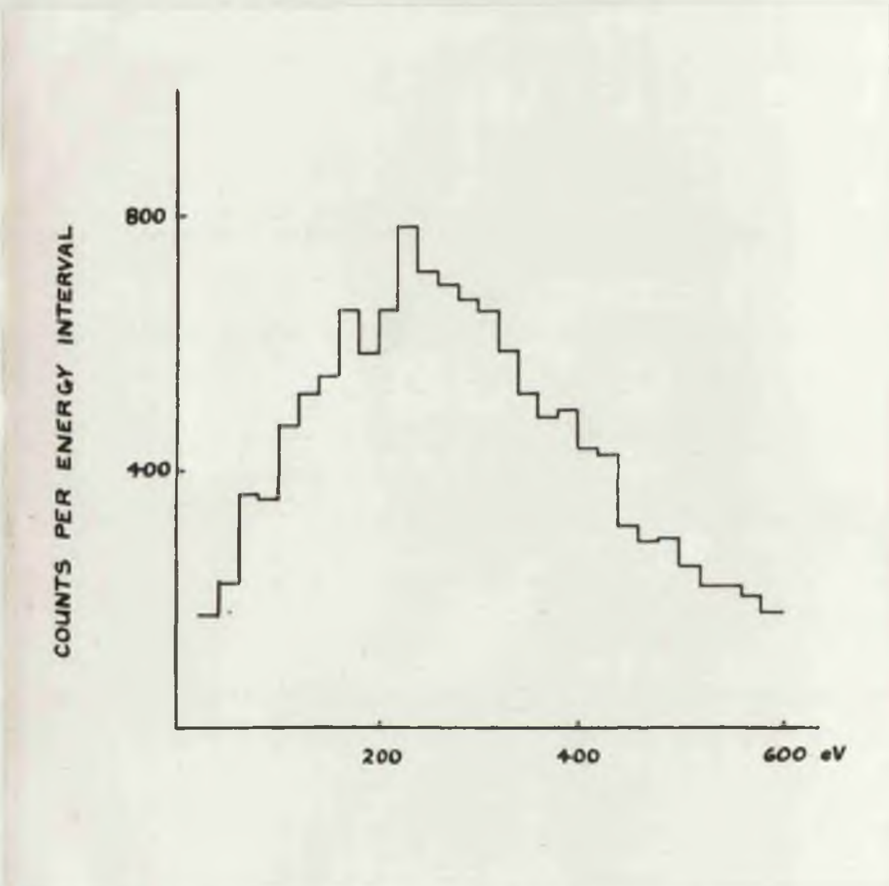


Fig. 7. A^{37} L-peak obtained with the plastic counter.

Compton spectrum caused by gamma-rays of known energy, it was found that electrons dissipating an energy greater than 25 keV in the plastic would give pulses which could be resolved from the photomultiplier thermal noise pulses. Fig. 7 shows the 240 eV L-peak from an A^{37} source introduced into the counter.

It was immediately observed, when switching in the anti-coincidence system, that at very high values of proportional counter gain no pulses at all passed through the gate. This was due to light emitted by excited atoms in the proportional counter discharge reaching the photomultiplier and triggering the anti-coincidence system. Some work on these light pulses has been carried out by Charpak and Renard (108, cf. also 109), who found that the light is emitted by excited states of half-life about 2×10^{-7} sec., and that the intensity is proportional to the energy dissipated in the counter when working in the proportional region. These workers claim that energy releases as low as 1 keV could be detected by this method. In most measurements with the plastic counter it would be necessary to prevent these light pulses from triggering the anti-coincidence system, and this can be done either by making the inside ends of the proportional counter opaque, or by decreasing the counter gain and at the same time using higher amplifier gain.

As a test of the effectiveness of the counter as an anti-coincidence device some runs were made to study the natural background. In this work thin sheets of blsck polythene were inserted at the inside ends to prevent the effect mentioned above. With the counter filled with the usual argon-methane mixture to a pressure of 1 atmosphere the counting rate in the proportional counter was about 120 counts/min. without anti-coincidence, and 10 c/min. with it. The counter was then surrounded by about 3 in. of lead and the measurements repeated; the corresponding counting rates were 32 c/min. and 1 c/min. It is seen that the anti-coincidence system is quite efficient.

After this small prototype counter had been shown to operate satisfactorily, Moljk and Drever constructed a similar counter with a considerably larger volume, and using one 5 in. photomultiplier tube at each end. This counter was used, inside different types of shielding, to study the character of the residual background in low activity equipment, with particular emphasis on applications to C^{14} dating, and results of preliminary work were presented at the U.N.E.S.C.O. Conference on Radio-Isotopes in Scientific Research (Paris 1957). An even larger model is now under construction by these workers.

A much larger version of the plastic counter was also constructed by the author and Dr. G.M. Lewis for further work on low intensity electron capture transitions. This counter was formed from a cylinder of plastic scintillator 12 in. long by 4 in. diameter, and light was collected at each end by a 5 in. photomultiplier. Unfortunately, large cracks appeared in the plastic after the machining, and these increased gradually with time, causing a drastic decrease in optical efficiency. Accordingly no useful work has been done with this counter.

CHAPTER THREE.

THE K/β^+ RATIO IN F^{18} AND THE FIERZ TERM IN THE GAMOW-TELLER INTERACTION.

1. Introduction.

The positron spectrum of F^{18} has been studied in magnetic beta-spectrometers by Blaser et al. (110) and by Ruby and Richardson (111). These workers are in good agreement on all features of the decay, which they find to be simple, with an allowed spectrum shape and an end-point energy of 635 $\pm$ 15 keV and 649 $\pm$ 9 keV respectively. This is in qualitative agreement with earlier work of Snell (112) and of Yasaki and Watanabe (113), who found a simple spectrum ($\sim$600-700 keV), using absorption and cloud-chamber techniques, and contradicts the results of Zah-Wei Ho (114), who obtained an additional high energy positron component and detected several gamma-rays.

Knox (115) found no evidence for gamma-rays with energy greater than 100 keV with intensity greater than 10 % of the beta disintegrations, and Ruby and Richardson agree with this result. The half-life is 112 mins. (110) and the transition is super-allowed with log ft = 3.6 . F^{18} is an odd-odd N=Z nucleus with a spin of 1 (116), assigned on the basis of isotopic spin selection rules which indicate that the transition to O^{18}, with zero spin and isotopic spin one, involves a Gamow-Teller interaction only.

Although electron capture is expected in F^{18} it had not been detected before the present work, mainly because the absence of gamma emission prevents the application of the usual indirect methods of measurement (cf. p.17). K-capture can only be detected by observing the K-radiations of oxygen subsequently emitted. These have an energy of 530 eV and, on account of the low fluorescence yield, 99.5 % of the K-capture events lead to the emission of Auger electrons instead of K x-rays. The suitability of the proportional tube spectrometer for the measurement of electrons in this low energy region has been discussed in chapter 2.

2. PREPARATION OF THE RADIOACTIVE SOURCE.

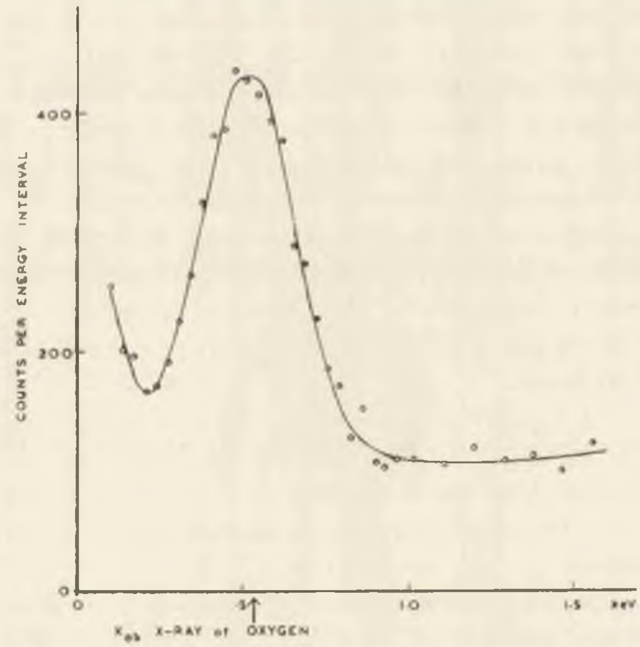
In the choice of a gas to be used as a radioactive source in a proportional counter the primary consideration is that the gas should not have a high attachment coefficient, which would impair the operation of the counter. Fluorine itself is not suitable for this reason and, in any case, would immediately react chemically with the counter wall. It was found in preliminary work that boron trifluoride BF_3 , and methyl fluoride CH_3F, could be used in small quantities.

With methyl fluoride, to a partial pressure of 2 mm. Hg., as the gaseous source in the counter, initial counting rates of about the same magnitude as the natural background were obtained, and, while this was sufficient to indicate the presence of the K-capture peak, the counting statistics were poor and it was evident that an accurate result would necessitate more intense sources. However, it was not desirable to increase the partial pressure of CH_3F in the counter because of the adverse effect on the energy resolution. An attempt to prepare higher specific activity CH_3F sources using a Szilard-Chalmers technique was unsuccessful.

Boron trifluoride gas has been used extensively in proportional counters for the detection of thermal neutrons, and, with three fluorine atoms in the molecule, was expected to give higher counting rates for the same partial pressure in the counter. A considerable improvement was in fact made by using BF_3 , and this gas was employed in most of the measurements. The BF_3 was made by heating radioactive calcium fluoride with boric oxide and sulphuric acid, and was freed from condensable impurities by cooling with solid carbon dioxide. In the preparation of BF_3 it is difficult to avoid making a small amount of silicon tetrafluoride, which has a high attachment coefficient and is therefore detrimental to the operation of the counter. However, extensive purification is required to remove the SiF_4 and this was precluded by the short half-life of F^{18}. The presence of this gas set a limit to the partial pressure ($\sim$ 4 mm. Hg) of the source which could be used in the counter.

A few grams of calcium fluoride were packed into a polythene tube and fixed in a position close up against the vacuum chamber of the Glasgow 350 MeV electron synchrotron. F^{18} was made by the reaction $F^{19}(\gamma,n)F^{18}$, which has a cross-section of 0.11 MeV-barns integrated up to 70 MeV (117). With a maximum output of 2×10^{9} equivalent quanta per min., the intensity of the most effective part of the bremsstrahlung spectrum in the neighbourhood of the (γ,n) "giant resonance" is seen to be just sufficient to give a source of the required specific activity. The CaF_2 was irradiated for periods of about four hours.

In the preliminary experiments with BF_3 a small peak was observed in the pulse spectrum at an energy of about 2.8 keV. It was found that this peak did not decay appreciably over a period of a day, and this fact, together with the measured energy, indicated that the impurity was A^{37}. This isotope decays by orbital electron capture with a half-life of 35 days, emitting the 2.8 keV K x-radiations of chlorine. It was assumed that the A^{37} was produced, in the reaction $Ca^{40}(n,\alpha)A^{37}$, by the fairly strong flux of neutrons at the position, close to the vacuum chamber, in which the target



K-capture peak of ^{18}F measured in a proportional counter with a gaseous source.

Fig. 8.

material was irradiated. This radioactive impurity was removed by freezing the BF_3 with liquid air and pumping.

3. MEASUREMENTS.

The measurements were carried out using a brass proportional counter with a diameter of 10 cm. and a sensitive length of 58 cm., defined by field-correcting tubes. Radioactive BF_3 to a pressure of about 2 mm. Hg was admitted to the counter, and the total pressure increased to three atmospheres by adding an argon-methane mixture ($\sim$10 cm. Hg CH_4). In general it is advantageous to have as high a gas pressure in the counter as possible, since this reduces the rise in the pulse spectrum at low energies caused by escaping positrons (cf. chap. 2), but in this case the advantages did not outweigh the practical difficulties of working at pressures in excess of three atmospheres.

The pulses from the counter were fed to a high gain linear amplifier and the pulse spectrum was analysed with a Hutchinson-Scarrott 100-channel pulse-height analyser. The 9.2 keV x-rays from an external Ge^{71} source were used for energy calibration, and the gain of the counter was adjusted so that the low energy end of the pulse spectrum was analysed. Fig. 8 shows that part of the pulse spectrum in the energy region between 100 eV and 1.6 keV. The peak at about 500 eV is attributed to the K x-radiations of oxygen, emitted in the K-capture decay of F^{18}. It is worth pointing out that this peak can only be caused by K-capture, since the integrating property of a counter with 4π geometry prevents the detection of only the x-rays in a multiple event such as the emission of x-rays following positron decay.

At the high counter gain used in this experiment it is possible for the space charge, created by very large pulses, to reduce the electric field at the wire and thus cause variations of gain and poor energy resolution. The energy of the observed peak was determined more accurately by using the x-radiations from an A^{37} source which was admitted to the counter.

The total counting rate and the counting rate in the peak were found to decay together with a half-life close to the published value for F^{18}. After several half-lives a long-lived activity was observed, but its intensity was low ($< 1\%$) and was allowed for when subtracting natural background.

In electron capture measurements of this type some uncertainty arises in the determination of the number of counts in the peak, because of its superposition on the continuous positron pulse distribution, which rises at low energy. This uncertainty increases with the width of the peak, which is determined largely by the statistical fluctuations in the number of ion pairs formed initially. In the present case this number is about 540/30 = 18, and the width is accordingly rather large. Electron attachment and the effects of the high counting rate of large positron pulses make the peak even wider. To reduce such errors to a minimum the area of the peak was obtained by a comparison with a source which did not emit positrons. To do this a trace of A^{37} was admitted to the counter after the F^{18} source had decayed, and the shapes of the 2.8 keV K-peak and 240 eV L-peak were studied. The shape of the F^{18} peak was compared with these, and the base of the peak, and consequently the area, were thus determined.

Errors due to the escape of K x-rays from the sensitive volume of the counter are completely negligible due to the almost complete absorption in the argon filling, and to the very small fluorescence yield ($\sim$ 0.5%). Accordingly the number of K-capture transitions in the sensitive volume is given by the number of counts in the observed peak.

The number of positron transitions was obtained from the total number of pulses recorded, after correcting for natural background and the low intensity radioactive impurity revealed by the decay curve. It has been the custom in this laboratory to operate proportional counters inside a large electro-magnet which shields the counters quite effectively from natural background. The background of the counter used in these experiments when inside the magnet was 700 counts/min. In most of the later runs the source strength was greater than five times this at the beginning of the run, and counting was stopped when the source intensity became about equal to the background.

The number of positrons recorded requires to be corrected for those positrons which enter the sensitive volume from the insensitive regions at the ends of the counter. This correction was made by drawing a scale diagram of one end of the counter, dividing this up into volume elements and determining graphically the average solid angle subtended at each volume element by the sensitive volume of the counter. Since the average range of the positrons is considerably greater than the distance between the boundary of the sensitive volume and the end of the counter, these effective solid angles can be integrated to give an approximation to the required correction. A more accurate result can then be obtained by correcting this result for the absorption of the positrons, on the basis of the known spectrum and the approximate range-energy relationship. The result of this calculation was that 4.6% of the recorded positrons were due to this effect.

Boron trifluoride is a reactive gas and during some of the later runs measurements were made to determine how much, if any, of the source had been adsorbed on the counter wall. On completion of a run of about one hour duration the counter was pumped out, refilled, and the counting rate measured. By comparing this with the counting rate observed directly before pumping out it was found that 23 % of the F^{18} activity had been deposited on the brass wall of the counter. The measurements on the decay of the positron and K-capture intensities indicated that no loss of source occurred during the experiments, and it was therefore assumed that the adsorption took place within a few minutes of filling. The number of positrons was corrected to take account of this effect. No correction to the observed number of K-capture events is necessary since it has been shown that such an adsorbed source would not contribute to the K-capture peak (118).

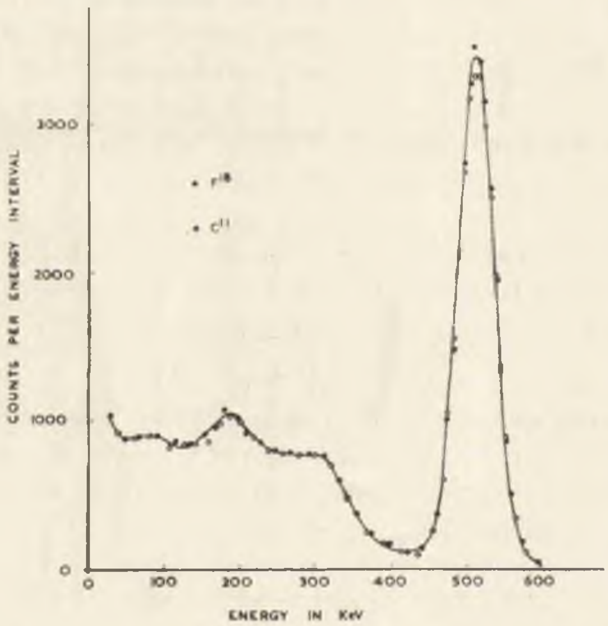
4. THE GAMMA-RAY SPECTRUM.

As noted in section 1 , Knox has placed an upper limit of 10 % on the relative intensity of gamma-rays with energy greater than 100 keV, but this does not exclude the possibility that some of the observed K-capture transitions could go to an excited state of O^{18}. Accordingly a more sensitive experiment to search for gamma-rays was carried out using a NaI scintillation spectrometer.

It was found, after the first run, that considerable difficulty would be experienced in detecting a low intensity photo-electric peak above the continuous Compton distribution due to the annihilation radiation. Accordingly a comparison method was adopted, the F^{18} gamma spectrum being compared with that of C^{11}, another simple positron source.

The NaI crystal, which had a diameter of 4.2 cm. and a height of 2.5 cm., was temporarily encased in a thin Perspex container, with an extension for mounting the radioactive sources on the end of the crystal. The Perspex between the source and the crystal was thick enough to absorb the highest energy positrons emitted. A thin layer of irradiated calcium fluoride was placed on top of the crystal and the F^{18} gamma-ray spectrum between 30 keV and 700 keV was examined with the multi-channel analyser. Spectroscopically pure graphite was used to prepare a C^{11} source by the reaction $C^{12}(\gamma,n)C^{11}$, and this experiment was repeated. Care was taken to ensure that the graphite layer was of the same thickness and in the same position relative to the crystal as the calcium fluoride in order to keep the counting geometry the same.

Carbon 11 is a simple positron emitter with an end-point energy of 970 keV and a half-life of 20 mins. No gamma emission has been detected. K-capture is present in 0.2% of the disintegrations



Scintillation counter spectra of ^{18}F and ^{11}C.

Fig. 9.

but this has no appreciable effect on the gamma-ray spectrum.

Fig. 9 shows the spectra obtained from both the F^{18} and C^{11} sources, normalized to the same total number of counts. No deviation is observed from the shape expected for the annihilation spectrum, and the close correspondence of the two spectra indicates that no gamma-rays with energy greater than 30 keV and relative intensity greater than 0.5% are emitted by either isotope.

During the measurements on the K/β^+ ratio with the proportional counter, an examination was made of the pulse spectrum in the energy region below 50 keV. No peaks were found other than that due to K-capture, and it may therefore be assumed that both the positron and K-capture transitions in F^{18} proceed directly to the ground state of O^{18}.

5. RESULTS AND CONCLUSIONS.

The observed ratio of K-capture to positron emission is $R = \lambda_K/\lambda_{\beta^+} = 0.030 \pm 0.002$, based on the results of a number of separate measurements. The estimated experimental limits are mainly due to the uncertainty in estimating the number of pulses in the K-peak.

Values of the K/β^+ ratio as a function of the energy for several values of Z have been calculated by Feenberg and Trigg (21), and by Zweifel (44). However, in general a rather uncertain interpolation would be required, and in the particular case of a low Z transition this procedure would be very inaccurate. Accordingly the ratio R_o was calculated from the formulae presented in chapter 1. In these calculations a positron end-point energy of 649 keV was adopted, and in the calculation of λ_K the effective value of the atomic number Z' was obtained from the rules of Slater (42) as 8.7 . To obtain λ_{β^+} the positron spectrum was plotted from eqn. 3, using the tables of the Fermi function given by Rose (1), and the area under the curve was measured. The result obtained was $\lambda_{\beta^+} = 0.604$, omitting the nuclear matrix element and the Fermi constant. Although screening effects are very small at low Z values allowance was made for this effect by recalculating λ_{β^+} taking into account the screening correction factors given by Reitz (119). These were obtained from Reitz's values by extrapolation. The final result was $\lambda_{\beta^+} = 0.607$.

When the Fierz interference term is neglected, the value R_o of the K/β^+ ratio is found to be $R_o = 0.0295$, which is very close to the measured value.

As noted in chapter 1 , by comparing the theoretical and experimental values of the K/β^+ ratio it is possible to set a limit to the magnitude of the Fierz interference term. If the K/β^+

ratio calculated for a small finite value of b_{G-T} has the value R, then from eqn. 11

$$b_{G-T} \doteq \frac{R - R_o}{R_o(1 + \langle W^{-1} \rangle)}$$

The values previously calculated for the probability of positron emission at various energies were multiplied by the reciprocals of the energies and the results plotted as a function of the energy. By dividing the area under this curve by the area under the positron spectrum the value of $\langle W^{-1} \rangle$ for F^{18} was obtained as 0.69 . Substituting the present theoretical and experimental values into the above equation gives $b_{G-T} = 0.008 \pm 0.04$, in good agreement with previous results (cf. chap. 1).

It has been shown in chap. 1 that, previous to the discovery of the non-conservation of parity, a small value for b_{G-T} was taken as implying a small value for either C_A or, less likely, C_T . However, the introduction of parity non-conserving terms to the beta-decay interaction leads to the amended formula for b_{G-T}:

$$b_{G-T} = 2\gamma \, \mathcal{R}e \left\{ \frac{(C_T C_A^* + C_T' C_A'^*)}{(|C_T|^2 + |C_A|^2 + |C_T'|^2 + |C_A'|^2)} \right\}$$

and the Fierz condition now implies only that the real part of $(C_T C_A^* + C_T' C_A'^*)$ is small. More explicit relationships between the T and A coupling constants can in principle be obtained by combining this result with those of the numerous other experiments at present being carried out for the purpose of elucidating the character of the beta-decay interaction. In chap. 8 a discussion is presented on the conclusions which can be drawn from the Fierz condition in the light of recent work.

CHAPTER FOUR.

THE K/β^+ RATIO IN C^{11} AND THE FIERZ TERM IN THE FERMI INTERACTION.

1. INTRODUCTION.

The positron decay of C^{11} has been investigated by Townsend (120) and by Siegbahn and Bohr (121). These workers found a simple allowed spectrum with an end-point energy of 981 $\pm$ 5 keV and 993 $\pm$ 10 keV respectively. More recently Richards and Smith (122) have carried out threshold measurements for the reaction $B^{11}(p,n)C^{11}$ which have indicated a positron end-point energy of 958 $\pm$ 3 keV. Sine the difference between this value and the ones obtained by direct measurements lies outside the combined probable errors, the positron spectrum was remeasured by Wong (123), using a magnetic lens spectrometer. He found the Kurie plot of the spectrum to be linear from the end-point of 968 $\pm$ 8 keV down to 255 keV. Together with the half-life of 20.7 mins.(124), this gives a log ft value of 3.62 for the transition, and confirms the super-allowed nature of the decay between the mirror nuclei C^{11} and B^{11}. Wong's result agrees with those of the nuclear reaction measurements within the combined probable errors.

Electron capture is expected in C^{11} but had not been observed before the present work for the reasons mentioned in chap. 3.1 .

The K-fluorescence yield is expected to be about 0.1 % (92), and the energy of the K-radiations of boron is 180 eV.

The lowest energy electron capture radiations observed before the present work were the L x-radiations of chlorine, with an energy of 240 eV, emitted in the electron capture decay of A^{37}. Since there is no positron emission accompanying this decay, the L-peak can be readily observed in a conventional proportional counter. In the decay of C^{11} however, K-capture is expected in only 0.2 % of the disintegrations, and the capture peak will be superimposed on a substantial continuous pulse distribution due to the positrons. The work on F^{18} had shown that this distribution would obscure any low energy peak with an intensity less than about 0.5 % of the positrons, and it was clear that for the C^{11} experiment large reductions would have to be made in the wall effect. The multi-wire and plastic counters described in chap. 2 were accordingly employed.

In the decay of C^{11} both Fermi and Gamow-Teller terms are expected, but the Fermi matrix element predominates (cf. below), and an accurate measurement of the K/β^+ ratio in this isotope can lead to an estimate of the magnitude of the Fierz interference term in the Fermi interaction.

Another matter of interest in this decay is associated with the comparatively large recoil energy of the nucleus in the

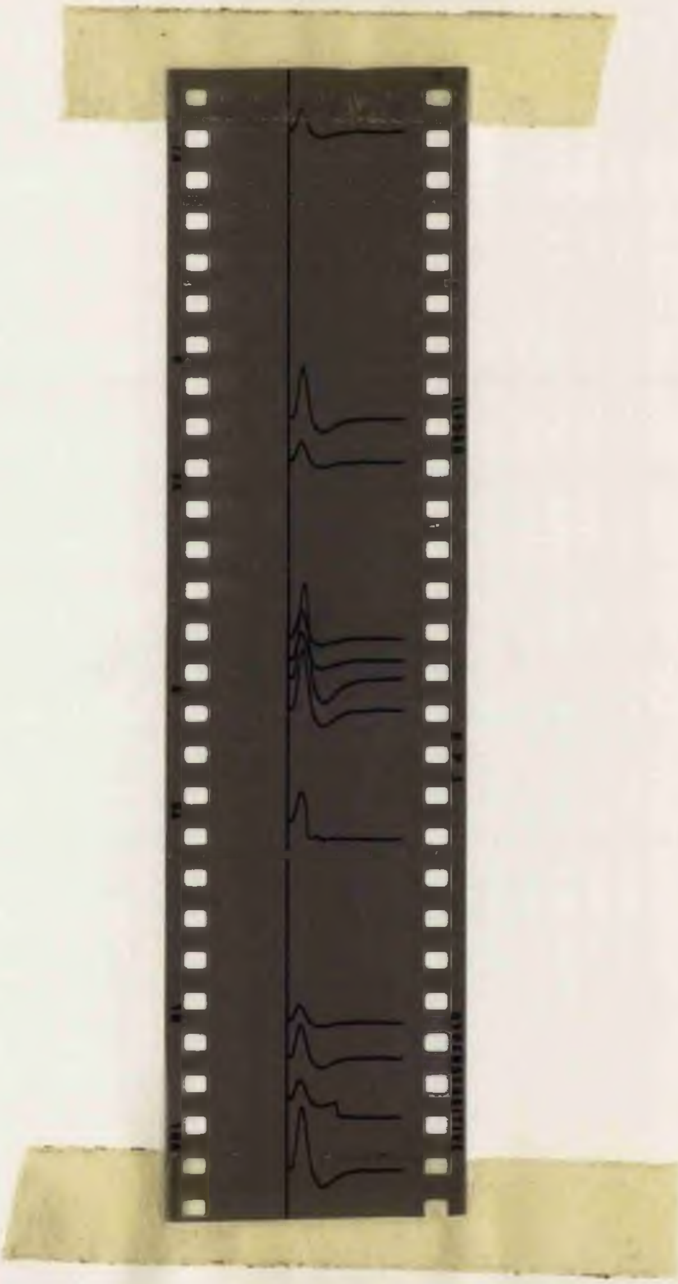
K-capture process. This amounts, for a free B^{11} atom, to 190 eV, whereas the energy of the K-radiations of boron is 180 eV. A shift in energy of the electron capture peak would be observed if the recoil ion were to dissipate a considerable fraction of its energy in ionization of the counter gas. The experiments which have been carried out at low energies indicate that such ionization is small, and that the energy is more rapidly expended in collision processes (125). However, these experiments are extremely difficult to perform, and it seemed of interest to obtain an independent check on this point.

2. APPARATUS.

The multi-wire counter described in chap. 2 was used in most of the measurements, but a few runs were made with the plastic counter to test its effectiveness. In the present application the black polythene discs at the ends of the counter were removed in order to allow positrons escaping from the sensitive volume to enter the plastic scintillator. In this way end effects were kept to a minimum. It was necessary in doing this to ascertain that the light pulses picked up by the photomultiplier from those proportional counter discharges corresponding to the energy region below 1 keV were not large enough to trigger the anti-coincidence unit. The photomultiplier voltage and amplifier gain were adjusted accordingly.

In this experiment some additions were made to the electronics described in chap. 2 . The anti-coincidence gate circuit was so arranged that pulses from the central counter were fed to a potential divider which reduced them to one tenth of their amplitudes and passed them to the input of the anti-coincidence pulse shaper. Since the bias of the Schmitt circuit on this input was set at 5 volts, the result was to eliminate all pulses from the central counter with amplitudes greater than 50 volts. This measure led to a considerable reduction in the number of pulses with which the analysing system had to deal, and also avoided the presence of very large pulses in the later stages of the electronics. Such large pulses are accompanied by negative overshoots of the amplifier baseline which are of considerably greater duration than the positive part of the pulses, and a smaller pulse following closely behind such a pulse is liable to be considerably distorted. However, the anti-coincidence gate pulse is longer than these overshoots, and such spurious events are not transmitted to the analysing system.

The analysis of the spectrum of the pulses which passed through the gate was carried out simultaneously in two completely different ways; by a Hutchinson-Scarrott multi-channel kicksorter, and by photography. The pulses were displayed on a commercial oscilloscope



Section of C^{11} film showing pulses from the multi-wire counter.

whose timebase was triggered by the pulses themselves, and were continuously photographed on 30 mm. film using a camera driven at a constant speed by an electric motor. The input pulses to the oscilloscope were delayed by 10μ sec. relative to the trigger pulses in order to enable the front edge of each pulse to be clearly seen. With this method the whole of each pulse is seen on a separate sweep, and, since the shape and length of the proportional counter pulse are well defined, spurious pulses due to pick-up from other apparatus may be rejected. Furthermore, a number of pulses arrive in coincidence with the edges of the anti-coincidence gate pulse, and are in consequence considerably distorted. These events are also eliminated with this system. The number of such spurious pulses was small ($<$ 1 %). A section of film is attached opposite p.64.

Since the triggering level in the commercial oscilloscope was found to be rather unstable, the trigger pulses were obtained from a standard discriminator unit for which the bias was stable and accurately known. The triggering level of the discriminator was set at the same voltage as the lower level of the kicksorter in order that the two pulse-height spectra obtained would be readily comparable. A block diagram of the electronic system is shown in fig. 3 .

A microfilm reader was employed to measure the heights of the pulses recorded on the film, and the spectrum sorted into thirty channels. Throughout this work the proportional counters were calibrated with the 9.2 keV x-rays from an external Ge^{71} source. The filmed spectrum was calibrated by feeding pulses from a pulse generator simultaneously to the triggered oscilloscope and to the kicksorter. These pulses were adjusted so that they occupied that kicksorter channel which was closest to the mid-point of the 9.2 keV peak. Filming the pulses on the oscilloscope then gave an accurate energy calibration.

3. EXPERIMENTAL PROCEDURE.

A glass vessel, of volume 300 cc., filled to various pressures up to two atmospheres with propane, was placed in a position close to the target of the Glasgow 350 MeV electron synchrotron and irradiated for periods of about half an hour. The C^{11} was formed by the reaction $C^{12}(\gamma,n)C^{11}$.

With propane at a partial pressure of 10 cm. in the multi-wire proportional counter the initial counting rate was as high as 30,000 counts/min. at maximum synchrotron output. The propane source was admitted to the counter and argon added to a total pressure of one atmosphere. Propane was employed in this work because of its high carbon content, and because the main impurities are similar hydrocarbons. Since propane itself is a good stabilising gas, and has been used extensively in proportional counter work for this purpose, it was not necessary to add methane, which is normally used in this laboratory as a stabiliser.

With the object of reducing the effects of positrons escaping from the ends of the counter a total gas pressure of two atmospheres was used in the counter during the early experiments. However, the average pulse height is halved by reducing the pressure to one atmosphere, and consequently the fluctuations in the level of the amplifier baseline due to very large pulses are reduced. Reducing the pressure also has the effect of decreasing the ionization density along the particle tracks, and any consequent distortions due to space charge effects.

Using the Ge^{71} source, the counter gain was adjusted until the peak due to the 9.2 keV x-rays was situated at the upper end of the kicksorter display. The amplifier gain was then increased by a factor of twenty and the low energy part of the pulse spectrum analysed. The accuracy of the amplifier gain settings was checked beforehand with a standard pulse generator. A rapid deterioration of the energy resolution, as defined by the width of the 9.2 keV peak, was observed as the counter gain was increased, showing the effects of local variations of gain due to space charge at the high gain used in this experiment. This effect leads to non-linearity of gain in the counter and set an upper limit to the gain of the whole system, since an increase of amplifier gain would have meant an approach to amplifier noise level.

When using the plastic counter it was found to be necessary to disconnect the anti-coincidence system in carrying out the energy calibration with the 9.2 keV x-rays, since proportional counter discharges corresponding to energies greater than a few keV produced sufficient light to trigger the system.

As far as the author is aware no measurements have been made on the relationship of pulse height to energy dissipation in a propane-argon mixture over the energy range covered in these experiments, and therefore separate measurements were made in order to verify that this relationship was at least approximately linear. A trace of A^{37} was admitted to each counter, which was then filled with the same argon-propane mixture as used during the main runs. For each counter the gain was adjusted until the peak due to the 9.2 keV x-rays was in the same position on the kicksorter display as it was when the counters were being calibrated, and the position of the peak due to the 2.8 keV K-radiations of chlorine was noted. The amplifier gain was increased by a factor of twenty and the position of the peak due to the 240 eV L-radiations of chlorine was noted. The position of this latter peak relative to the 9.2 keV peak provided a very accurate and convenient calibration in the search for the C^{11} K-peak. For convenience, in all of the later runs the 9.2 keV peak was kept in this position. Within the limited accuracy of these measurements it was found that the relationship between pulse height and energy was linear over the energy range of interest here.

In a typical run the initial counting rate in the central counter was about 3000 counts/min., and of these about 15 counts/min. were allowed through the anti-coincidence gate to the analysing system. The total counting rate and the number of counts in the K-peak were observed for several half-lives and found to decay with a half-life of 20 minutes. No appreciable radioactive impurities were present. The pulse spectrum due to natural background was examined, and was found to be very small in magnitude and flat over the energy region under study.

It is essential for measurements on low energy Auger electrons that the source be in gaseous form, and a separate experiment was carried out to confirm that this was the case. The proportional counter was filled with the usual propane source and argon to the usual pressure, and the counting rate was noted. The counter was pumped out, refilled, and the counting rate again noted. No significant increase over the known natural background counting rate was observed, indicating that none of the source had been deposited on the counter wall.

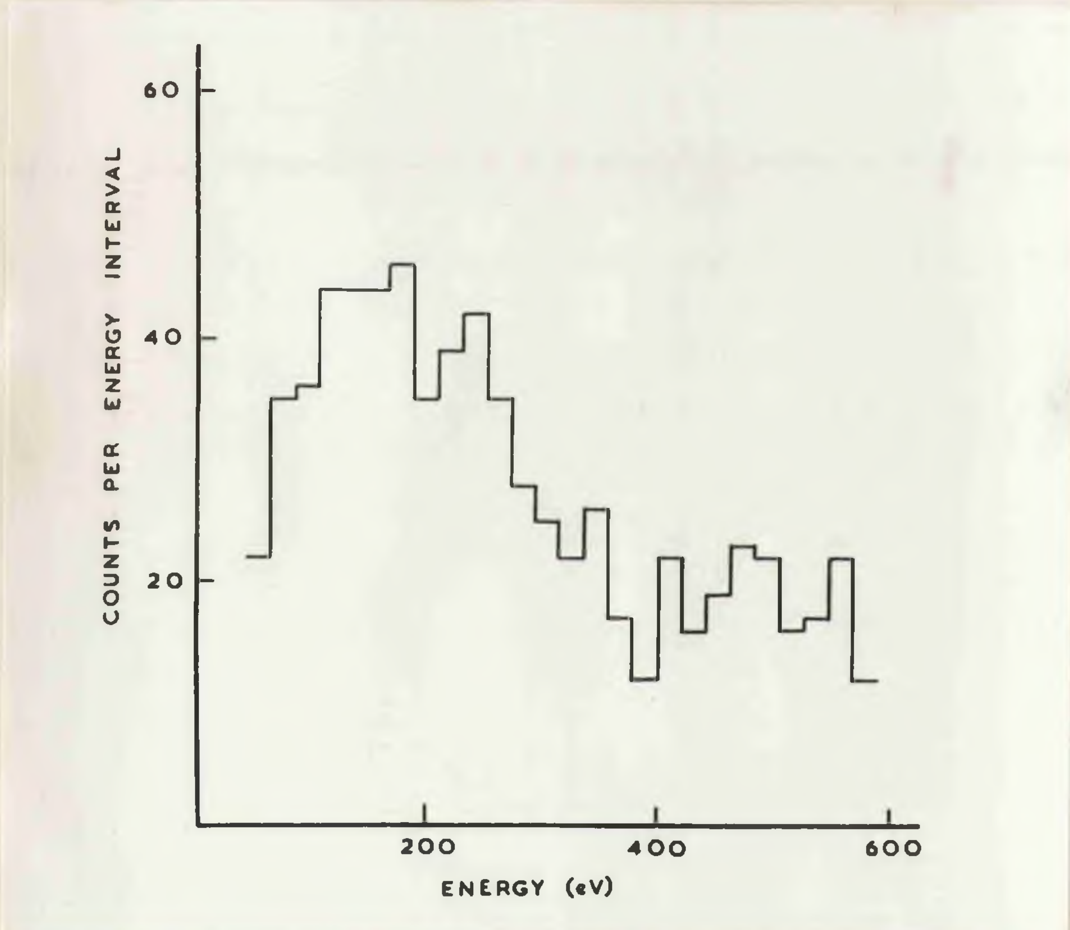


Fig. 10. C^{11} K-peak obtained with the multi-wire counter.

In another experiment it was verified, by irradiating the empty glass vessel, that none of the source activity was due to dust or adsorbed gases coming off the wall of the vessel.

4. RESULTS AND CONCLUSIONS.

On analysing the low energy end of the pulse spectrum obtained with the multi-wire counter a broad peak was observed at an energy of about 180 eV, which was attributed to the K x-radiations of boron emitted in the K-capture decay of C^{11}. Fig. 10, which is the result of three successive runs, shows that part of the pulse spectrum in the energy region between 40 eV and 600 eV. The natural background, which accounts for about three counts per energy interval, has not been subtracted.

The K/β^{+} ratio was obtained from the number of counts in the peak and the total number of counts in the pulse spectrum, after correcting for natural background. The number of positrons was obtained as described in chap. 2.2 , and a correction of about 2% , which was calculated as described in chap. 3, was applied to take account of those particles which enter the sensitive volume from the insensitive regions at the ends of the counter. A further correction of about 2 % was included for the time the gating unit was shut.

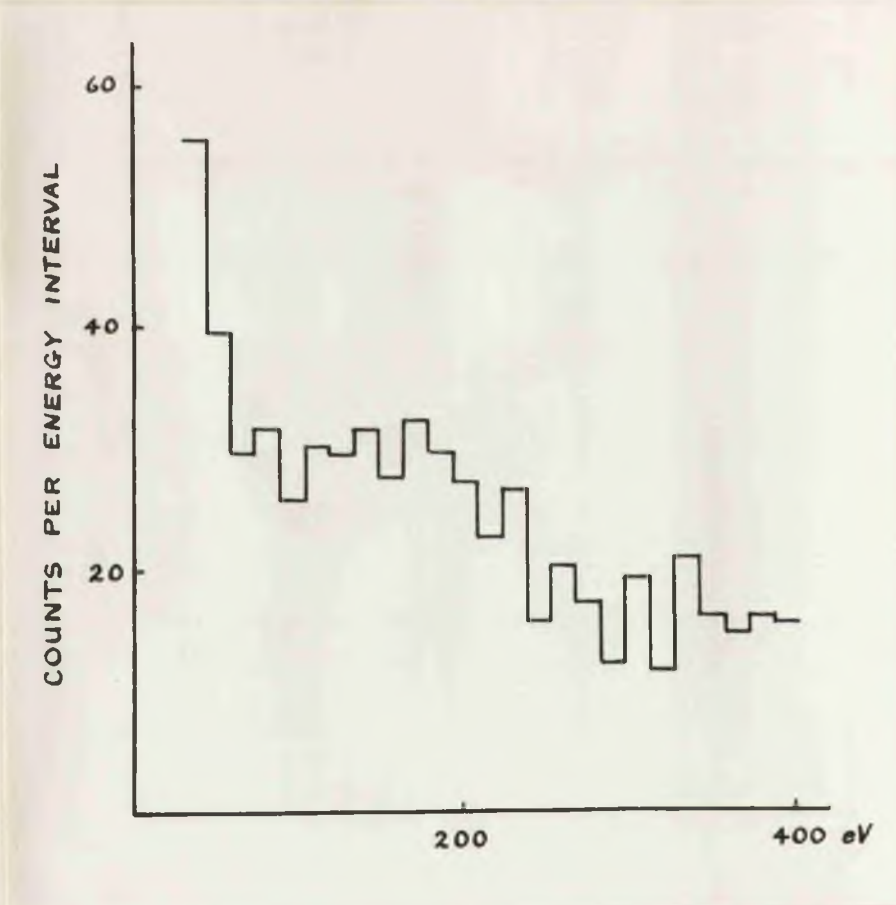


Fig. 11. C^{11} pulse spectrum obtained with the plastic counter.

The observed K/β^+ ratio is 0.0019 ± 0.0003 , the errors being largely due to the uncertainty in estimating the area of the peak.

That part of the pulse spectrum from the plastic counter in the energy region between 40 eV and 600 eV is shown in fig. 11, and the step in the spectrum at about 200 eV is consistent with the presence of K-capture in the above intensity.

The position of the C^{11} K-peak relative to that of the A^{37} 240 eV peak indicates that little or no contribution to the energy of the peak is provided by ionization due to the nuclear recoil, and such ionization must be quite small.

The theoretical value of the K/β^+ ratio for C^{11} has been calculated in the same manner as for F^{18}, with the result that $R_0 = 0.0020$. The positron spectrum end-point energy has been taken as 970 keV (123), and a screening correction to the K-capture decay constant has been applied by taking the effective Z value for carbon as 5.69 (43,126). As in the experiments on F^{18}, the measured value of the K/β^+ ratio is in excellent agreement with the theoretical value.

Substituting R and R_0 into eqn. 11 gives $b = -0.03 \pm 0.10$. To obtain the value of b_F from this value of b requires the knowledge of the relative magnitudes of the Fermi and Gamow-Teller nuclear matrix elements for C^{11}. Since C^{11} is a mirror transition the change in the nuclear radial wave functions can be neglected, and $|M_F|^2$ and $|M_{G-T}|^2$ can be calculated from the angular wave functions alone (26). The calculation of the Fermi matrix elements for mirror transitions depends solely on the assumption of charge independence of nuclear forces, requiring no other assumptions about nuclear structure, and the result is that $|M_F|^2 = 1$ for all such transitions. Semi-empirical values of the corresponding Gamow-Teller matrix elements have been obtained by Trigg (28) and by Kofoed-Hansen and Winther (26) from the measured nuclear magnetic moments, and have been used by these workers to determine $|C_F|^2/|C_{G-T}|^2$ (cf. chap. 1).

However, the value of $|M_{G-T}|^2$ used here for C^{11} was obtained from eqn. 6 by substituting the known value of the ft product, $|M_F|^2$, B and x. The values of B and x were taken as 2787 and 0.560 respectively from (27), and in this way it was found that $|M_{G-T}|^2 = 0.41$, which is close to the value of 0.45 calculated by (26).

By substituting these values of the matrix elements into the equation for b, and by using the value of b_{G-T} given in chap. 3.5, an estimate can be made of the magnitude of b_F. In this way it is found that $b_F = -0.04 \pm 0.18$. The estimated maximum positive value is 0.14, which is considerably smaller than the value of 0.29 required by Kofoed-Hansen and Winther (27) to obtain the best fit for their B-x lines. However, the lower limit of about 0.08 given by these workers lies within the estimated limits of the present result, and therefore there is no clear-cut disagreement between the two estimates.

Substituting the present result into the equation for b_F gives $\mathrm{Re}\,(C_S C_V^* + C_S' C_V'^*) = -0.02 \pm 0.09$. The conclusions to be drawn from this condition are discussed in chap. 8.

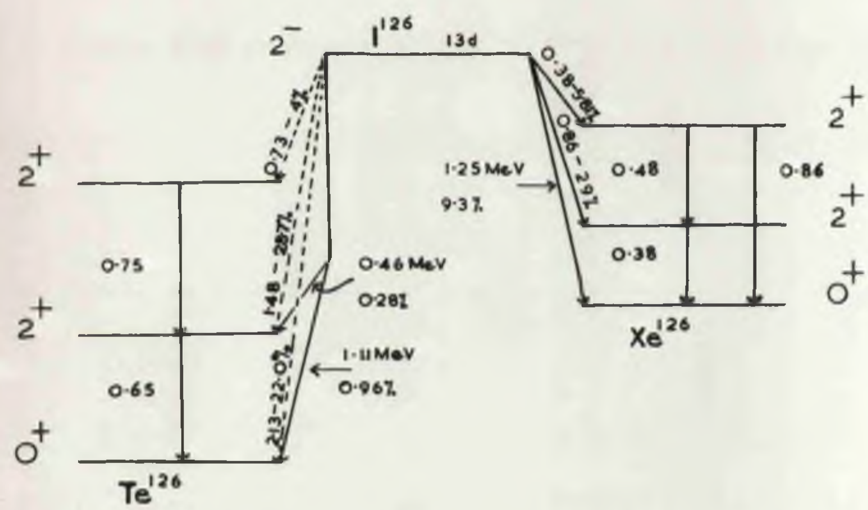


Fig. 12. Decay scheme of I^{126}.

CHAPTER FIVE.

THE L/K-CAPTURE RATIO IN I^{126}.

1. INTRODUCTION.

The most recent investigation of the decay scheme of I^{126} has been carried out by Koerts et al. (62), using a beta-gamma coincidence solenoidal magnetic spectrometer and gamma-gamma NaI scintillation spectrometers. Unique first forbidden shapes were observed for the spectra of both the negatron and the positron transitions from the ground state of I^{126} to the ground states of Xe^{126} and Te^{126}, indicating that the I^{126} ground state has spin 2 and odd parity. This conclusion is strengthened by the fact that allowed shape spectra were observed for the negatron and positron transitions to the excited states. Measurements on the electron capture decay led to values for the K/β^{+} ratios for the transitions to the ground and first excited states of Te^{126}, and these results are discussed in chap. 1.2 . The decay scheme proposed by these workers is shown in fig. 12, and is in good agreement with that given by Perlman and Welker (127, cf. also 128, 129).

As pointed out in chap. 1.4 , a measurement of the L/K ratio for a fairly heavy nucleus would be of some importance because of the reliance which has been placed on the theoretical results to deduce transition energies and L-fluorescence yields for nuclei in this mass region.

The I^{126} decay has several features which make such a measurement both simple and accurate. Firstly, the source can be formed inside the NaI crystal, in which the measurements on the x-radiations take place, simply by bombarding the crystal with the photon beam from the Glasgow synchrotron. Corrections for geometry, efficiency, absorption and scattering, and for L and K-fluorescence yields are then almost negligible. Both the L and K x-radiations are detected with effectively 100 % efficiency.

Secondly, the half-life of 13 days allows a reasonably strong activity to be formed, and is long enough to allow accurate measurements to be made over a period of several weeks.

Thirdly, by taking coincidences with the gamma-rays from the excited states in Te^{126}, the orbital capture radiations are isolated from the particle radiations, and, in addition, the photomultiplier thermal noise contribution is very considerably reduced. The latter advantage is of critical importance in the measurement of the 4.9 keV L x-radiations.

Finally, although very many photonuclear reactions are possible in I^{127}, Na^{23}, Tl^{203} and Tl^{205}, most of these are of very low cross-section relative to the (γ,n) process (130,131,153), and in any case

. . . .

do not lead to radioactive nuclei with characteristics similar to those of I^{126}. For a radioactive impurity to be detected by the present technique it would have to be an electron capture transition, with x-ray energies close to those of tellurium, followed by a gamma-ray of energy greater than 550 keV, or, alternatively, an isomeric transition in which a gamma-ray of about the same energy as the tellurium x-radiations is followed by a gamma-ray of energy greater than 550 keV. In either case the half-life would have to be close to that of I^{126}. On examination of the isotopes which could be produced in significant quantities it was found that none satisfied the above requirements, and it was estimated that less than 1 % of the observed x-gamma coincidences were due to impurities. The good energy resolution of the observed x-radiation peaks and the absence of any unexpected peaks in the coincidence gamma-ray spectrum confirm this (cf. below).

The scintillation spectrometer has been employed by several workers for the measurement of x-rays with energies as low as 2 keV (132-134), and, in view of the considerable reduction in photo-multiplier thermal noise obtained in the present work by using a coincidence technique, no difficulty was envisaged in measuring the 4.9 keV L x-radiations of tellurium.

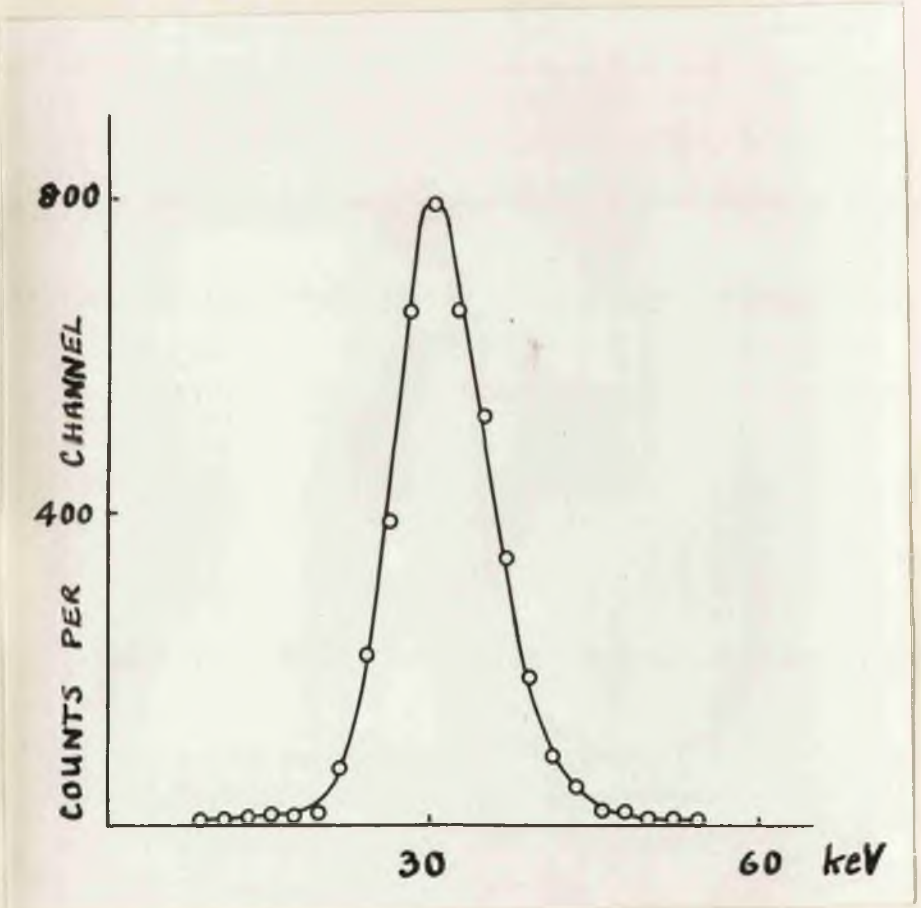


Fig.13. Part of the I^{126} coincidence pulse spectrum from the sou

crystal showing the K-capture peak.

2. EXPERIMENTAL PROCEDURE.

A NaI crystal 1 in. long and $1\frac{1}{2}$ in. in diameter was irradiated in the photon beam from the 350 MeV synchrotron for about two days. At the position, close to the collimator, in which the crystal was situated the beam is about $\frac{1}{4}$ in. in diameter, and it can be assumed that the activity resulting from the reaction $I^{127}(\gamma,n)I^{126}$ is concentrated in a cylinder of about $\frac{1}{4}$ in. diameter along the axis of the crystal. This assumption is not critical. Preliminary experiments were carried out with a smaller crystal of diameter $\frac{1}{2}$ in. and length $\frac{3}{4}$ in.

The source crystal was mounted on a photomultiplier (Dumont type 6292) which had been selected for its low thermal noise. Another NaI crystal 2 in. long and $1\frac{3}{4}$ in. in diameter was mounted on a photomultiplier and was placed close to the source crystal. A coincidence unit with a resolving time of 1μsec. controlling a linear coincidence gate unit with a gate length of 10μsec. was used to select those pulses from the source crystal which were in coincidence with pulses from the other crystal corresponding to energies greater than 550 keV. This section of the apparatus was calibrated with annihilation radiation from Na^{22} and with the 661 keV gamma-rays from Cs^{137}.

The low energy end of the pulse spectrum passing through the

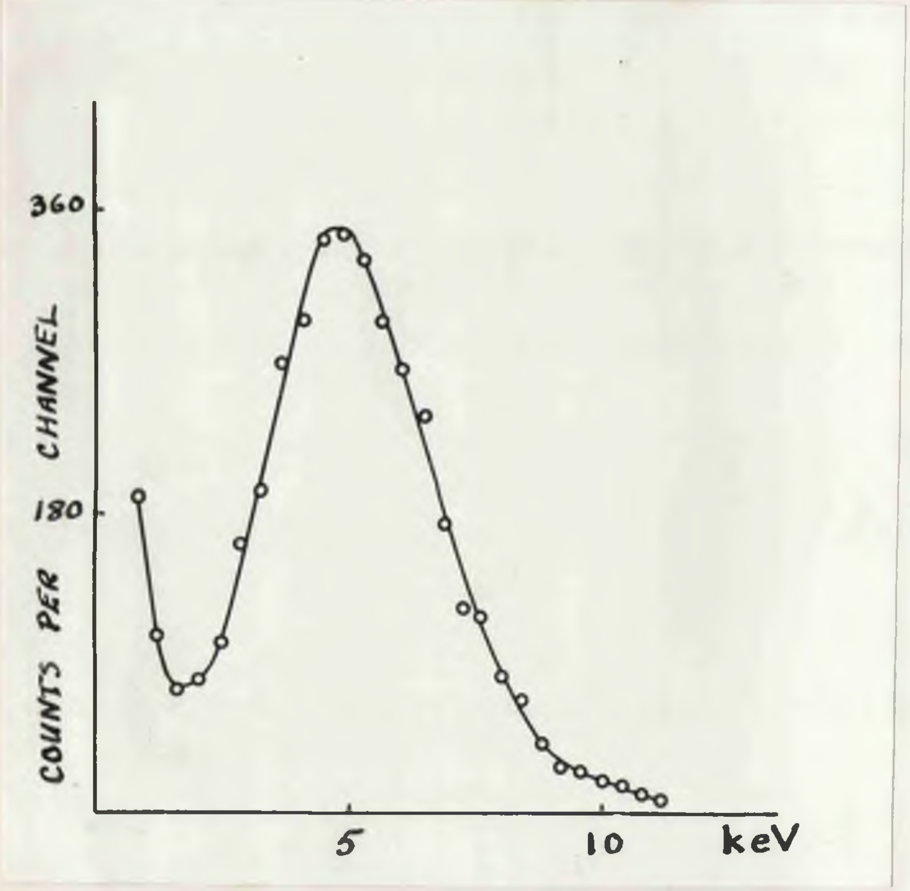


Fig. 14. Part of the I^{126} coincidence pulse spectrum from the source crystal showing the L-capture peak.

gate was analysed with the multi-channel kicksorter, and a prominent peak was observed (fig.13) whose energy, by comparison with the 46.5 keV calibration peak from a RaD source, was found to be 32 keV, corresponding to the K-absorption energy of tellurium. The amplifier gain was increased by a factor of six, and the pulse spectrum analysed as before. Another peak was observed (fig.14) at 4.9 keV, corresponding to the L-absorption energy of tellurium. It was not possible to analyse the two peaks simultaneously, but a check was made on the stability of the electronics by ascertaining that the total number of pulses above 12 keV in each run remained constant.

The intensity of I^{126} gamma-rays detected in the large crystal was about one half that of the natural background, and an approximation to the accidental coincidence spectrum in the source crystal was obtained by separating the two counters. This spectrum was of negligible intensity, with the exception that a large part of the thermal noise distribution was present. In the spectra shown this has not been subtracted.

The gamma-ray spectrum from the large crystal in coincidence with x-radiations in the source crystal was examined with the multi-channel kicksorter. A prominent peak was observed whose energy, by comparison with the Cs^{137} 661 keV calibration peak, was found to

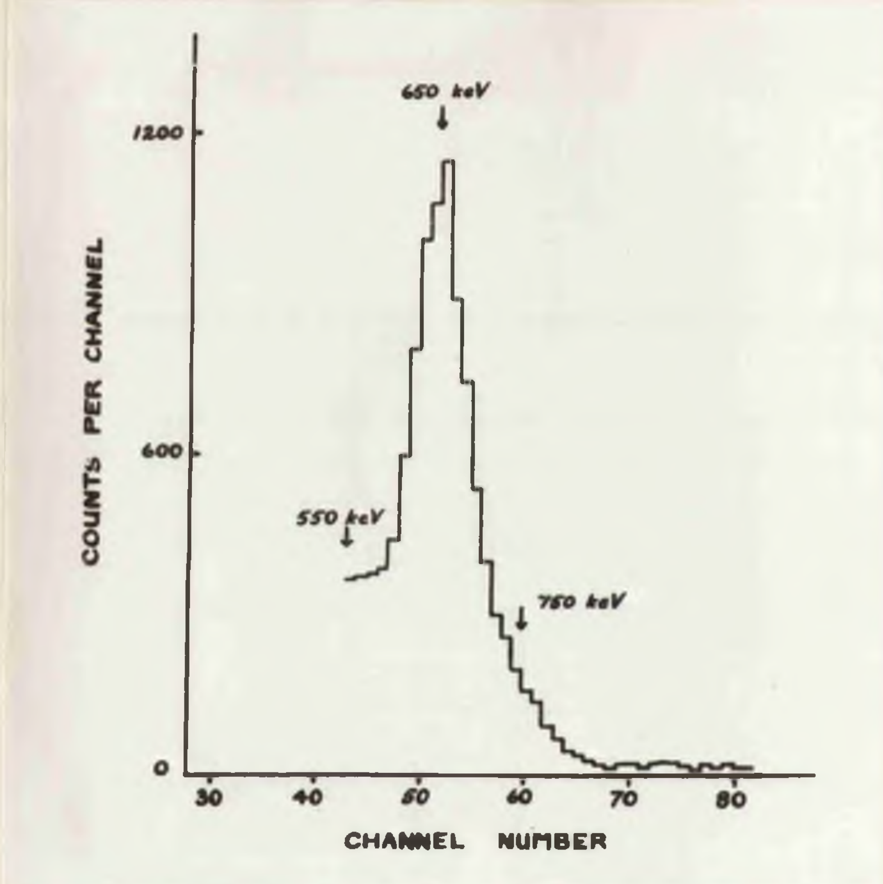


Fig. 15. Part of the gamma-ray spectrum in the large crystal in coincidence with the I^{126} electron capture x-radiations in the source crystal.

be 650 keV. This is shown in fig. 15. The effect of the discriminator which selects those pulses corresponding to energies greater than 550 keV is shown by the cut-off in the spectrum at this energy. The energy resolution is not good enough to distinguish the low intensity 750 keV gamma-ray from the main 650 keV peak. No other peaks were observed in the energy region from 550 keV to 2 MeV with an intensity greater than 2 % of that of the 650 keV peak. However, the possibility cannot be ruled out that part of the observed pulse distribution in the energy region from 550 keV to 800 keV is due to radioactive impurities. As pointed out in section 1 , this is unlikely to be the case.

Some 60 day I^{125} is formed in the crystal due to the reaction $I^{127}(\gamma,2n)I^{125}$, but the electron capture decay of this isotope is not followed by a high energy gamma-ray and did not therefore contribute to the observed peaks (87).

3. RESULTS.

The ratio of counts in the L-peak to counts in the K-peak was observed to be $0.144 \begin{smallmatrix} +.005 \\ -.018 \end{smallmatrix}$, the mean of several measurements. The area of the L-peak was measured assuming a Poisson distribution for the pulse heights, and the possible errors are mainly due to the uncertainty in estimating this area.

A K-capture event in which a K_α x-ray escapes from the source crystal gives rise to the detection of an L x-ray which does not correspond to L-capture, and accordingly a small correction has to be applied to take account of this effect. It is assumed that escape occurs mainly from the ends of the crystal, and an approximate calculation (135) leads to the result that 0.2 % of the K x-radiations are not absorbed. This assumption is not critical, since in the extreme case of the source being distributed uniformly throughout the crystal the correction to the K x-radiation intensity would still be only 0.9 % (136). The above fraction of K-capture events then gives rise to an increase of 1.4 % in the number of counts in the L-peak. The corrected value of the L/K ratio is $0.142 {}^{+.005}_{-.018}$.

4. DISCUSSION.

The decay energies of the electron capture transitions studied here are given by Koerts et al. as 1.48 MeV and 0.73 MeV, in the intensity ratio of seven to one. Using the curves of Dirac wave function ratios given by Brysk and Rose (48), the L/K -capture ratios for these transitions were calculated to be 0.125 and 0.129 respectively. The theoretical ratio for the sum of these transitions is then 0.126 , which is just within the experimental limits on the observed value. However, on the basis of the Poisson fit which was made to the L-peak it appears to be more likely that the experimental ratio is slightly larger than the theoretical.

CHAPTER SIX.

ELECTRON CAPTURE IN As^{74}.

1. INTRODUCTION.

The decay of As^{74} has been studied by several groups since the energy spectrum of the emitted particles was first measured, in a cloud chamber, by Sagane et al. (137), and found to contain both negatron and positron components. Using a magnetic lens spectrometer, Deutsch and Roberts (138) found that a gamma-ray with an energy of 0.582 MeV was emitted in the decay of As^{74}. Mei et al. (139) made some observations on the beta and gamma-ray spectra of As^{74}, in the process of studying the decay of As^{72}. They established the presence of two negatron groups of end-point energies 1.45 MeV and 0.82 MeV, and one positron group of energy 0.96 MeV, the ratio of negatrons to positrons being about two. They also fixed the energy of the gamma-ray at 0.593 MeV. The most recent work on the decay scheme of As^{74} is by Johansson et al. (140), who report the presence of an additional high energy component in the positron spectrum, and show that the previously observed gamma-ray spectrum is composed of two lines, with energies 0.5963 MeV and 0.6352 MeV, and with the intensity ratio about four to one. It was found, by means of beta-gamma coincidence techniques, that the gamma-rays follow the two

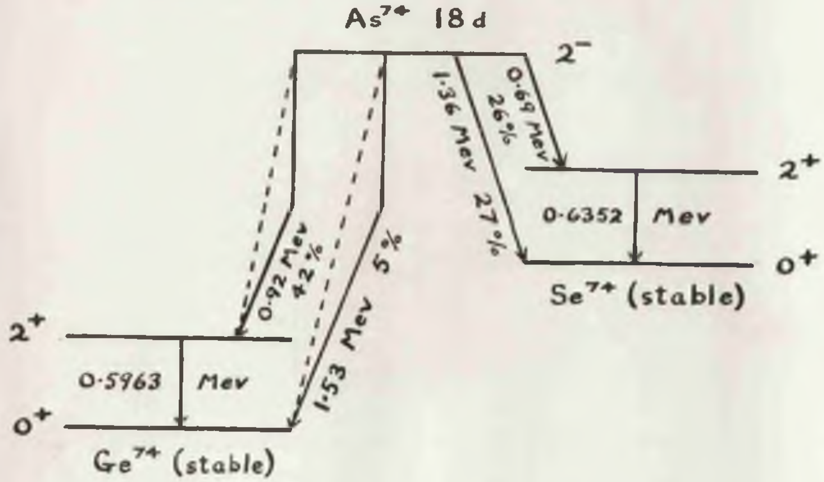


Fig. 16. Decay scheme of As^{74}.

softer components. The decay scheme proposed by these workers is shown in fig.16. The spin and parity assignments were made on the basis of the experimental ft values and spectrum shapes of the various beta-transitions, taking into account the fact that the end nuclei are of even-even type, with even parity and zero spin.

From the measured intensities of the beta-components and the gamma-rays, the ratio of electron capture to positron emission for the transition to the excited state in Ge^{74} was found by Johansson et al. to be 1.5, as compared with the theoretical allowed value of 1.2. However, this result is less accurate than those of the beta intensity measurements and these workers claim that their value is in good agreement with theory.

The present measurements on the electron capture decay of As^{74} were carried out for several reasons. No measurement had been made of the branching ratio $\epsilon/\beta^{+}+\beta^{-}$, and a determination of this ratio would lead to a value for the ϵ/β^{+} ratio for the first forbidden ($\Delta I = 0$, yes) transition to the first excited state of Ge^{74}. It would be of interest to confirm the findings of Johansson et al. that this ratio is larger than the theoretical allowed value. It was pointed out in chap. 1.4 that very few direct measurements have been made of the L/K-capture ratio where the transition energy is known, and that a large proportion of the measured values are in disagreement with the theory. The discrepancies found by the French workers for Ge^{71} and Kr^{79} are particularly serious. The author is of the opinion that if these workers had filled their proportional counters to pressures of two or three atmospheres with argon they would have obtained pulse spectra in which the ratio of the area of the L-peak to that of the K-peak would have been smaller than they in fact obtained for the final L/K ratio, after applying their corrections for K x-ray escape. The measurement of the L/K ratio in As^{74} was expected to demonstrate this. Finally, arsenic is a useful addition to the list of elements which can be studied by the gaseous source technique.

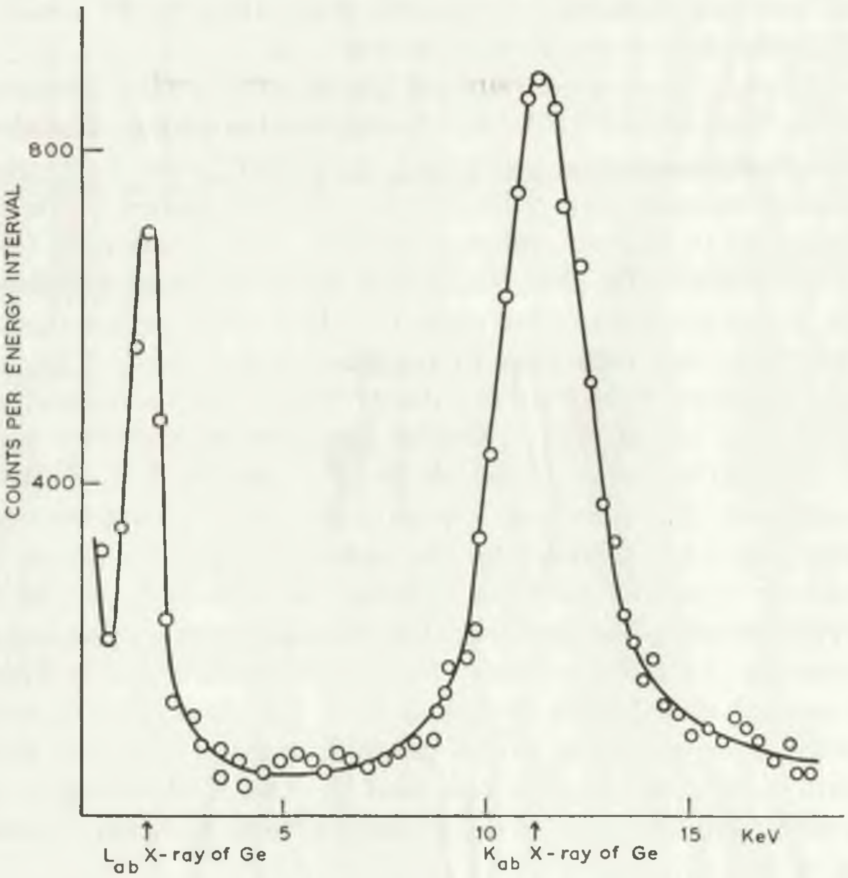
2. EXPERIMENTAL PROCEDURE.

The As^{74} was made by the (γ,n) reaction on arsenic using the photon beam from the 350 MeV synchrotron. Arsenious oxide was compressed in a ring of glass tubing, and fixed in a position in front of, and concentric with, the beam collimator. In this way the source was left in position for five weeks without disturbing other workers using the machine. Arsine gas was made by dropping magnesium turnings on a solution of the radioactive arsenious oxide in dilute HCl. Because of the toxic properties of this gas, even in very small quantities, the preparation and ultimate disposal of the arsine were carried out with great caution.

Part of the arsenious oxide was mounted, as an internal solid source, on the walls of a small proportional counter, and the decay of the total counting rate and of the K-capture counting rate observed for six half-lives. The counting rates decayed together with a half-life of 18 days, which is close to previously published values (143). A long-lived component, with an intensity of about 1 % of the As^{74} activity, was observed in the decay of the total counting rate, and the experimental results were adjusted accordingly. The results of experiments on the yields from various photonuclear processes in arsenic (131,153) indicate that most of this long-lived component was due to 80 day As^{73}, produced by the $(\gamma,2n)$ reaction.

In these experiments the proportional counter used had a diameter of 14 cm. and a sensitive length of 35 cm., defined by field-correcting tubes. Arsine to a pressure of about 1 mm. Hg was introduced into the counter, and methane to a pressure of 7 cm. Hg and argon to two atmospheres, were added. The presence of the arsine in the counter did not seriously affect the energy resolution, as defined by the width of the peak due to the 9.2 keV x-rays from the external Ge^{71} source used for calibration.

That part of the pulse spectrum in the energy region between 2 keV and 20 keV was examined with a multi-channel kicksorter, and a prominent peak was observed whose energy, by comparison with the Ge^{71} calibration peak, was found to be 11.1 keV, corresponding to the K-absorption energy of germanium. The amplifier gain was increased by a



~~Fig. 1~~. Part of the proportional counter pulse spectrum of As^{74} including both electron capture peaks. This has been constructed from separate curves of the K and L peaks.

Fig. 17.

factor of eight, and the pulse spectrum between 0.2 keV and 3 keV was analysed as before. Another peak was observed at about 1.3 keV, corresponding to the L-absorption energy of germanium. Fig. 17 shows that part of the pulse spectrum in the energy region between 0.2 keV and 18 keV, and includes both electron capture peaks. This curve has been constructed from separate curves of the K and L-peaks, fitted at about 4 keV. An L-peak is shown in fig. 18.

The relative intensity of electron capture was determined from the number of counts in the two peaks and the total number of counts in the pulse spectrum. Due to the fact that a fraction of the K x-rays emitted in the K-capture decay escape from the counter, the areas of the peaks observed in the spectrum do not correspond directly to electron capture from the L and K shells. However, as explained below, most of the escaping K x-rays give rise to counts in the L-peak, and the sum of the areas of the two peaks corresponds closely to the total electron capture intensity. The effect of capture from the M and higher shells is neglected here since the probability of M-capture is expected to be at most 10 % of that of L-capture for transitions where the decay energy is much greater than the L x-ray energy. It was calculated that only 3.2 % of K-captures yielded x-rays which escaped completely and did not lead to a count in either peak.

The number of electrons, obtained from the total number of pulses recorded, was corrected for those particles entering the sensitive

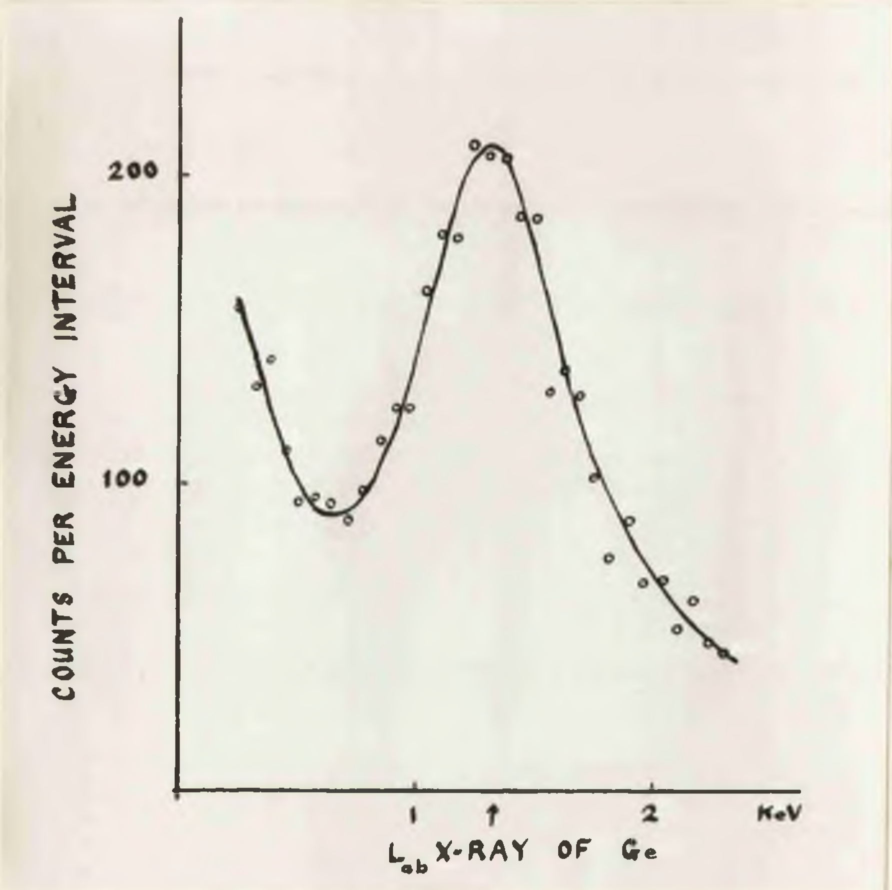


Fig. 18. Typical L-peak of As^{74}.

volume from the ends of the counter. This correction was calculated graphically to be 7.5 % of the number of electrons. A similar correction, calculated to be 1 %, was applied to the observed number of x-rays.

On completion of the measurements the proportional counter was pumped out and refilled with an argon-methane mixture as before.. The pulse spectrum was analysed as before, and it was found that 18 % of the As^{74} activity had been deposited on the walls of the counter. Total counting rates, and the areas of K and L-peaks observed at different times, indicated that no loss of source occurred during the measurements, so it may be assumed that adsorption takes place immediately after filling. The difference between the observed spectra, after allowing for decay, is the pulse spectrum due to the gaseous source.

3. RESULTS AND CONCLUSIONS.

After applying the above corrections, and taking into account the uncertainty in estimating the areas of the capture peaks, the branching ratio of electron capture to electron emission was found to be 0.67 $\pm$0.04. Using the latest values of the relative intensities of the electron groups (140), namely a β^+/β^- ratio of 47/53 , the ratio of electron capture to total positron emission becomes 1.42 . Combining this with the theoretical ratio (0.8) for the low intensity first forbidden unique transition to the ground state, a value of 1.49 is obtained for the first forbidden ($\Delta I = 0$, yes) transition to the first excited state of Ge^{74}. Since the theoretical value for an allowed transition of the same energy is 1.2, this result would seem to confirm the theoretical prediction (cf. chap. 1.2) that in forbidden transitions of this type the ϵ/β^+ ratio should be close to the allowed value. Unfortunately our present knowledge of the relative magnitudes of the nuclear matrix elements is so slight that detailed theoretical predictions are not possible. However, it is interesting to note that the increase over the allowed value observed here is of the same magnitude as that observed by Welker and Perlman (63) for a similar transition in Rb^{84}.

It has been assumed in the above argument that it is valid to accept as correct the theoretical ratio for the unique forbidden transition to the ground state. In fact no consistent experimental evidence on this point is available at present (cf. chap. 1.2), but, in view of the success of the theory in the allowed calculations, it seems likely that the theoretical results are valid. In any case the unique transition is of low intensity, and is unlikely to affect the above discussion critically.

From the relative areas of the observed peaks the L/K ratio can be obtained. The observed K-peak is due to Auger electrons and to the K x-rays which are absorbed in the counter gas. The observed L-peak arises from Auger electrons and x-rays emitted following L-capture, and from L-radiations emitted after those K-capture events in which a K_α

x-ray escapes from the counter gas. To obtain the L/K ratio from the relative areas of the two peaks it is necessary to know:

(a) The fraction of K x-rays which escape from the counter. Because of the low energy of the L x-rays and the low value of the L-fluorescence yield the fraction of L x-radiations escaping is negligible.

(b) The K-fluorescence yield of germanium.

(c) The fraction of K_α x-rays in the K-series.

Because of the effect of the field-correcting tubes, which are maintained at a potential two-thirds that of the counter case, the sensitive volume is quite accurately a right cylinder. The fraction of photons, emitted by a source distributed uniformly throughout a cylinder, which escape from the cylinder can be calculated to a high degree of accuracy, using formulae given by Hammersley (136). This calculation was carried out for the K x-rays of germanium, using values of 65 cm.^{2}/gm. and 49 cm.^{2}/gm. for the absorption coefficients of the K_α and K_β radiations respectively (141), and the escape fractions were found to be 29 % and 42 % respectively. The accuracy of these values is dependent only on the accuracy with which the x-ray absorption coefficients are known.

The value of the K-fluorescence yield of germanium was obtained from the semi-empirical formula of Broyles et al. (93) as 0.52 , and the ratio of K_α intensity to the total K x-ray intensity was taken as 0.85 (141).

Using these values it was calculated that 12.8 % of the K-capture events gave rise to pulses in the L-peak. The ratio of counts in the L-peak to counts in the K-peak was observed to be 0.250 ± 0.025, the errors largely being due to the uncertainty in estimating the areas of the peaks. From this value the L/K ratio is deduced to be 0.085 ± 0.020, in agreement with the theoretical value of 0.095, obtained from the graphs of this ratio given by Brysk and Rose (48). This result is comparatively insensitive to variations in the value assumed for the K-fluorescence yield, a change of 10 % in the latter giving rise to a change in the L/K ratio of only 0.017 . If the value for the K-fluorescence yield is taken from the new data of Laberrigue-Frolow et al. (94) the value of the L/K ratio becomes 0.090 .

It is seen that the ratio of counts in the L-peak to counts in the K-peak, namely 0.25 , is lower than the values obtained by the French workers for the L/K ratios in Ge^{71} and Kr^{79}, namely 0.30 and 0.26 (cf. chap. 1.4), as predicted in section 1. The present result confirms that the discrepancy, if any, between the experimental and theoretical results is comparatively small.

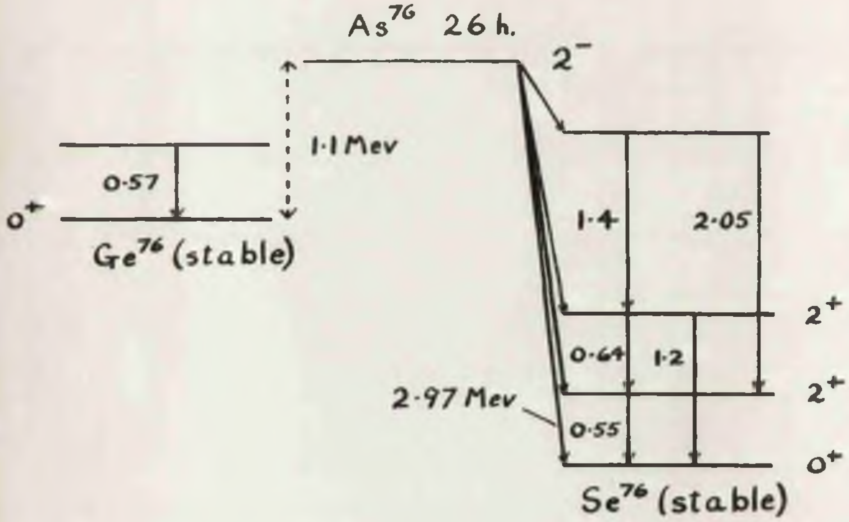


Fig. 19. Decay scheme of As^{76}.

CHAPTER SEVEN.

A SEARCH FOR ELECTRON CAPTURE IN As^{76}.

1. INTRODUCTION.

The decay scheme of As^{76} has been investigated by numerous workers since the discovery of the isotope by Amaldi et al. (142). A comprehensive summary of the more recent information obtained up to 1955 is given in Nuclear Level Schemes (143). Several investigators have established the presence of four negatron groups and several gamma-rays, but the actual number of gamma-rays and their relation to the excited states of Se^{76} have been in doubt.

Kurbatov et al. (144) have carried out the most recent detailed investigation of the decay scheme, and have obtained reasonable agreement with earlier work of Kraushaar and Goldhaber (145). The decay scheme proposed by the former group is shown in fig. 19, where the spin and parity assignments have been made from considerations of the beta-spectrum shapes, ft values and gamma-gamma angular correlations.

Fig. 19 also shows the expected position of the ground state of Ge^{76} relative to that of As^{76}. The energy difference between these states was computed to be 1.10 ± 0.12 MeV , from the weighted average of three measurements. Two of these (146,147) were mass-spectroscopic determinations of the atomic mass difference between Ge^{76} and Se^{76}, from which the required As^{76} - Ge^{76} decay energy was obtained by subtraction from the known As^{76} - Se^{76} decay energy. The third estimate of the decay energy was obtained from the difference in the neutron binding energies of the two nuclei. (148).

Since the requirement for the emission of positrons is that the decay energy be greater than 1.02 MeV, there might well be a low energy positron transition in the decay of As^{76}. However, from considerations of the expected ft value, such a transition would be expected to be of low intensity relative to that of the negatron transitions, and in fact several workers have placed quite low upper limits on the β^+/β^- branching ratio. Probably the most reliable work is that of Mims and Halban (149), who searched for positron activity by attempting to observe the gamma-gamma coincidences due to annihilation photons. A positive result of 7×10^{-4} positrons per disintegration is explained by these workers as being due to pair production by hard gamma-rays from the source, followed by the annihilation of the pair positron. This at any rate places an upper limit on the β^+/β^- ratio. Subsequently, Murray and Kurbatov (150) claimed to have observed positrons, with a maximum energy of 670 keV, from an As^{76} source in a cloud chamber, the β^+/β^- ratio being about 10^{-3}. However, it is reported in ref. (143) that these workers now consider that the positrons which they observed were of non-nuclear origin, and it is probable that the result of Mims and Halban is valid.

In a case such as As^{76}, where there is very little energy available for the positron transition to Ge^{76}, the K/β^{+} ratio is very large, due to the fact that in the capture process there is approximately 1 MeV more energy available than in the positron process. For As^{76}, taking the decay energy as the average value above (1.10 MeV), and the maximum value (1.22 MeV), the theoretical allowed values of the K/β^{+} ratios are about 4000 and 160 respectively. Hence, even if positron emission is possible, electron capture should be much more intense. However, the detection of a low relative intensity K-capture transition in competition with a beta-transition is much more difficult than the detection of positrons, since with any external source method there is the likelihood of detecting K x-rays of As and Se, emitted in the negatron decay and in the subsequent gamma emission. It would be difficult to distinguish between these x-rays and the K x-rays of Ge emitted in the K-capture decay. However, as explained in chap. 2.1 , the integrating property of the proportional counter used with a gaseous source results in the elimination of x-ray peaks due to particle and gamma emission.

An excited state of Ge^{76} at 0.57 MeV above the ground state has been observed recently in observations on the reaction $(\alpha,\alpha'\gamma)$ (145), and if, as seems likely, this is a 2^{+} state, some of the electron capture transitions would be expected to take place to this level. The presence of electron capture would then be shown by the appearance, in the As^{76} gamma-ray spectrum, of a line at 0.57 MeV. However, the presence of a 0.55 MeV gamma-ray following the negatron decay makes a search for electron capture by this means rather insensitive, and Backstrom (151), who has carried out such a search using a double-focussing magnetic spectrometer, concludes that electron capture to this state exists at most in 5 % of the disintegrations. No other search for electron capture in As^{76} has been reported, and, in view of the large energy difference, it seemed desirable to carry out such a search.

2. EXPERIMENTAL PROCEDURE.

The As^{76} sources were obtained from A.E.R.E. Harwell in the form of arsenious oxide, and arsine gas was prepared as in chap.6. A portion of the arsenious oxide was mounted in a 2 in. $\times$ 2 in. NaI crystal scintillation spectrometer, and the characteristic gamma-rays of As^{76} were observed in a multi-channel kicksorter. Further confirmation of the identity of the source was obtained by observing, over a period of several days, that the counting rate in the proportional counter decayed with a half-life of 26 hours.

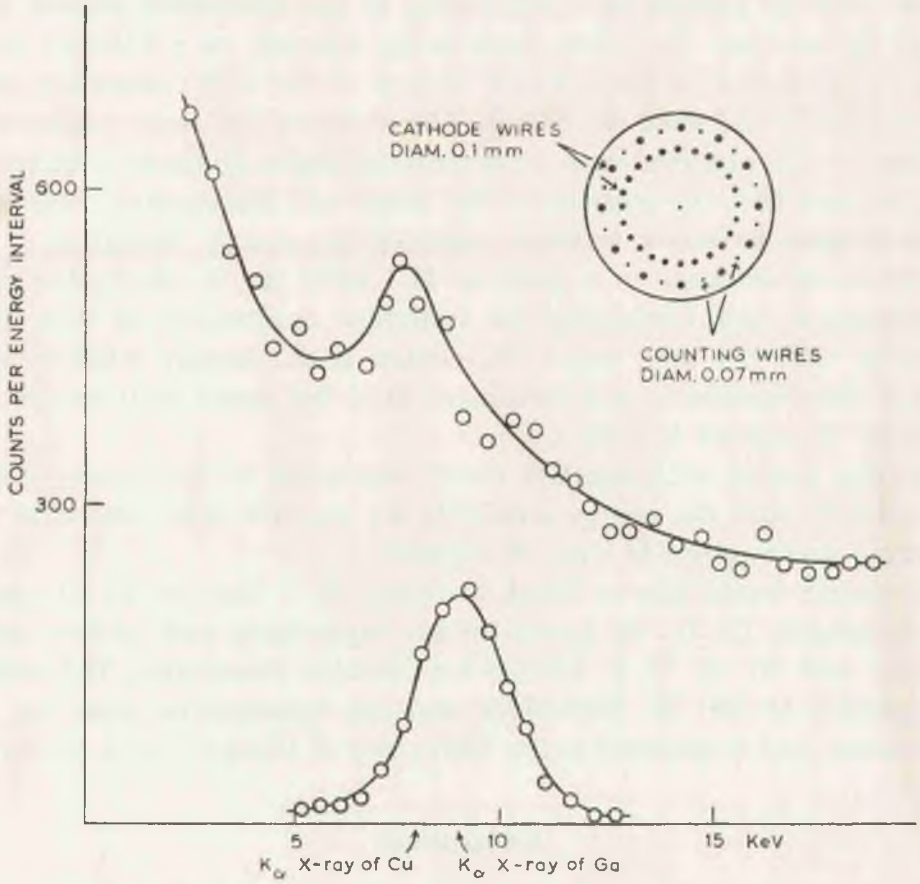
A proportional counter of the soft glass external cathode type described by Cockcroft and Valentine (152) was used for the first experiments. This was one of several such counters constructed by the author, and was 5 cm. in diameter with a sensitive length of 70 cm. The external cathode and guard rings were of colloidal graphite.

Arsine to a pressure of a fraction of a mm. Hg was introduced into the counter, which was then filled to various pressures up to two atmospheres with an argon-methane mixture. The spectrum of pulses from the counter was analysed with the multi-channel kicksorter, and the operation was checked by observing the energy resolution of the 9.2 keV x-ray peak from an external Ge^{71} source.

That part of the pulse spectrum in the energy region between about 2 keV and 18 keV was examined with the kicksorter. No evidence was found for a peak at 11.1 keV, the K-absorption energy of germanium, and it was concluded that the upper limit for K-capture was about 2 % of the disintegrations.

These experiments were continued with a large brass counter, resulting in a lowering of the upper limit to 0.4 % of the disintegrations.

The final experiments were carried out using the multi-wire counter. Pressures of up to two atmospheres of argon-methane were used. After the experiments the counter was pumped out, refilled, and the observed counting rate compared with the known natural background. It was found that about 6 % of the source had been deposited on the counter wall.



~~Fig. 2.~~ Part of the proportional counter pulse spectrum of As^{76}. The lower curve is the peak due to the 9.2 keV X-rays from an external Ge^{71} source. Inset is a cross-section of the proportional counter used.

Fig 20.

That part of the pulse spectrum in the energy region between 2.5 keV and 17.5 keV is shown in fig.20. A small peak at about 8 keV is superimposed on the continuous electron spectrum, which has been greatly reduced by the operation of the anti-coincidence system. This peak is interpreted as due to the K x-rays of copper produced by the excitation of atoms in the counter wall by cosmic rays and by gamma-rays originating in the insensitive regions at the ends of the counter. Part of this peak appears also in the natural background spectrum. The lower curve in fig.20 shows, on a different vertical scale, the peak due to the 9.2 keV x-rays of the Ge^{71} calibration source, observed under the same conditions. The width of this latter peak is about the same as that obtained with a conventional counter, and it is clear that the presence of the arsine and the unusual construction of the counter have not seriously affected the energy resolution.

There is no evidence of a peak at 11.1 keV and, comparing the statistical uncertainty at that part of the curve with the area under the copper peak, namely 0.035 % of the electron disintegrations, it is concluded that the upper limit for the probability of K-capture is 0.02 % .

A similar search, with negative result, was made for L-capture, although it is unlikely that the energy available for capture is so low as to make L-capture more probable than K-capture.

CHAPTER EIGHT.

CONCLUSIONS.

1. K/β^+ RATIOS.

The present work on the simple allowed transitions C^{11} and F^{18} satisfactorily confirms the validity of the theoretical results for such transitions, at least for light nuclei. For heavy nuclei the effects of screening and of the finite size of the nucleus become important, and Zweifel's calculations (45) show that corrections for these effects reduce the theoretical K/β^+ ratio by about 25 % for $Z = 84$ and $W = 3\ mc^2$. Accurate measurements are needed in the high Z region to compare with these results.

As pointed out in chap. 1.2 , little accurate work has been done so far on K/β^+ ratios for forbidden transitions, mainly because of the complex decay schemes in which such transitions occur. However, the author is of the opinion that present experimental techniques are adequate for accurate investigations of this type, should theoretical progress justify this. At present unambiguous theoretical values of the K/β^+ ratio can be calculated only for the unique forbidden transitions, and it is particularly important that these be submitted to experimental test. For other forbidden transitions the theoretical transition probabilities are complicated functions of the energy, of the coupling constants, and of several different nuclear matrix elements, and unambiguous results will be available only when the relative magnitudes of the latter can be assessed. However, the few experimental results which have been obtained (cf. chap.1.2 and chap.6) are significantly different from the relevant theoretical allowed values, and, when better estimates of the nuclear matrix elements are available, it is probable that the K/β^+ ratio will be a more sensitive test for the theory than spectrum shape measurements.

2. THE FIERZ INTERFERENCE TERMS.

The results of the present work strengthen the conclusion that the Fierz interference term is very small for both the Gamow-Teller and the Fermi interactions, although the evidence is less conclusive in the latter case. Previously the Fierz condition led unambiguously to the conclusion that in the general beta-decay interaction only one of each of the pairs S,V and T,A was present (cf. eqn. 15). However, the introduction of the parity non-conserving terms into the interaction has led to the more complicated expression for b (eqn.4), and the Fierz condition can now be interpreted in several different ways. Several schemes have been proposed for the relationships between the various coupling constants, but so far none of these has been completely successful in explaining the experimental results.

The two-component neutrino theory of Lee and Yang (154, cf. also

155,156) requires a combination of the coupling constants $C'_k = \pm C_k$, and it is seen from eqn. 4 that in this case the Fierz condition reduces to $\mathrm{Re}\; C_T C_A^* = 0$ and $\mathrm{Re}\; C_S C_V^* = 0$. If it is then assumed that time reversal invariance is valid (i.e. the C_k, C'_k are real) the condition reduces to the earlier form (eqn. 13). This theory receives support from the results of experiments on electron polarization (157-166), on the angular distribution of electrons from aligned sources (7), and on beta-gamma circular polarization correlation. (171). The results of Ridley's experiment on Ne^{23}, and that of Ambler et al. (170) on the angular distribution of positrons from aligned Co^{58} nuclei, might constitute a difficulty if the validity of time reversal invariance were assumed. However, the interpretation of both of the latter experiments is dependent on estimates of the relative magnitudes of Fermi and Gamow-Teller matrix elements, and these estimates may be incorrect.

In an attempt to resolve these latter difficulties more complicated theories have been proposed. Preston (167) and also Mayer and Telegdi (168) have suggested that in a β^--decay due to S and/or A a right-handed anti-neutrino is emitted, and that in a β^--decay due to T and/or V a left-handed anti-neutrino is emitted. The combination of coupling constants becomes $C'_S = C_S$, $C'_A = C_A$, $C'_V = -C_V$ and $C'_T = -C_T$. It is seen that the Fierz term is then identically zero. This theory predicts the absence of any terms due to interference between T and S, and between V and A, which is in agreement with the findings of Ambler et al. for Co^{58} but not with those of experiments on beta-gamma circular polarization correlation, in which large interference terms have been observed (171)

Recently a four-state neutrino theory has been proposed by Cavanagh et al. (172), which requires that neutrinos emitted in a T interaction have the opposite sign of spin to those emitted in an A interaction. The combination of coupling constants becomes $C'_T = -C_T$, $C'_A = C_A$, $C'_S = -C_S$ and $C'_V = C_V$. This scheme then implies that the electron polarization will be $\pm$ v/c irrespective of the ratio of T to A or S to V, and agrees with the two-component in predicting the presence of S-T and V-A interference terms. The Fierz term again becomes identically zero without any restrictions on the relative magnitudes of the coupling constants.

Recently Goldhaber et al. (173) have found that the neutrino in a pure Gamow-Teller electron capture decay is left-handed, which, taken together with electron polarization results, indicates that the Gamow-Teller interaction is predominantly A. This is in marked disagreement with the He^6 and Ne^{23} results, and makes it imperative that the He^6 experiment be repeated. An experiment similar to that of Goldhaber et al. should also be performed for a negatron emitter, since it is possible that the form of the interaction may be different in this case.

If the He^6 result is found to be wrong, a large amount of other experimental data would lead to the conclusion that the dominant interaction combination is A,V , and the two-component theory would then appear to be able to explain a large fraction of the experimental results so far obtained. The results of the work on the Fierz term described here would then lead to the conclusion that the S and T interactions are relatively small. If the He^6 result is found to be correct then it would appear that a more complicated theory of the beta-decay interaction is required.

Should it be found that time reversal invariance is not valid, the interpretation of the Fierz condition would become much more difficult. The coupling constants C_k, C'_k would no longer be real quantities and the Fierz term could vanish from phase considerations irrespective of the relative magnitudes of $|C_k|$ and $|C'_k|$.

3. L/K RATIOS.

The present measurements on As^{74} and I^{126} confirm that the discrepancy, if any, between the theoretical and experimental values of the L/K ratio is comparatively small. This is in agreement with the result of Drever and Moljk (95) for Ge^{71}, but contradicts the earlier work on Ge^{71} (89,72) and Kr^{79} (50,90,91), which might indicate that incorrect values of the K-fluorescence yield were assumed in the latter work.

It is clear that in general little reliance can be placed on methods of measuring the L/K ratio which require an accurate knowledge of K-fluorescence yield values, since the experimental limits on the latter are still relatively high. In this respect the internal source technique appears to be both the simplest and the best method, provided that close to 100 % of the K x-radiations can be absorbed in the detector. The proportional counter method of Drever and Moljk could be used for the region $15 \lesssim Z \lesssim 35$; above this region the high pressures which would be necessary would constitute a considerable practical difficulty. For the region $45 \lesssim Z \lesssim 70$ the method employed here for I^{126} would be suitable if small quantities of the source could be introduced into the melt from which NaI crystals are grown (cf. 87,169).

It would probably be advisable in the immediate future to concentrate on L/K ratio measurements for those transitions for which the theoretical ratio is effectively independent of the relative magnitudes of the nuclear matrix elements. Only when completely satisfactory agreement is obtained between experiment and theory for these simple cases would it be profitable to turn attention to those forbidden transitions in which the decay energy is low enough for the relative magnitudes of the matrix elements to become important. However, accurate measurements of the L/K ratio for such transitions could yield valuable information about the latter, and this also would probably be a better method for obtaining such information than the study of spectrum shapes.

APPENDIX.

THE BETA-SPECTRUM OF He^6.

1. INTRODUCTION.

The decay of He^6 by beta emission to Li^6, with a spin change of one and no parity change, is not allowed by the Fermi selection rules, and, since its ft value classifies it as a super-allowed transition, it is among the most important pieces of evidence for the presence of the Gamow-Teller interaction in beta-decay. An accurate knowledge of the ft value of this transition is of importance in the determination of the relative magnitudes of the Fermi and Gamow-Teller interactions (3), and many measurements of the half-life have been made, so that this quantity is known to a high degree of accuracy. However, the measurement of the beta-spectrum, and the determination of the end-point energy, are very difficult to carry out, due to the short half-life and the difficulty of obtaining strong activities, and only two such experiments have been performed.

The first accurate measurement of the He^6 spectrum was made, on a semi-circular focussing magnetic spectrometer, by Perez-Mendez and Brown (174), who obtained an end-point energy of 3.215 ± 0.015 MeV, which, together with a half-life of 0.799 sec. (175), gives an ft value of 568 sec. The theoretical interest in the spectrum led to its being remeasured by Wu et al. (176) with a magnetic solenoidal spectrometer, resulting in an end-point energy of 3.50 ± 0.05 MeV, with a Kurie plot linear from the end-point down to about 600 keV. This result gives an ft value of 792 sec.

The difference between the end-point energies obtained by these two groups is considerably larger than the combined probable errors, and, in view of the importance of this spectrum, measurements were started in this laboratory using a scintillation counter technique. Although the energy resolution of the scintillation counter is worse than that of the magnetic spectrometer, it was hoped that measurements could be made of the low energy part of the spectrum, and that better counting statistics would be obtained. It was hoped to improve in both of these respects on the previous measurements.

The method adopted here consisted of bombarding a LiI(Eu) crystal with the photon beam from the 30 MeV electron synchrotron, thus forming He^6 by the reaction $Li^7(\gamma,p)He^6$. The energy spectrum of the He^6 activity, distributed fairly uniformly throughout the crystal, was then studied using the multi-channel kicksorter. During the preliminary experiments described here a $\frac{1}{2}$ in. cube crystal was used, but it was intended that a larger crystal should be used in the later work.

Single crystals of lithium iodide, with thallium activator, have been grown by Hofstadter et al. (177), who have studied its properties as a detector of thermal neutrons. The neutrons are captured by Li^6

(7.5 % abundance), with a large cross-section (~900 barns), in the reaction $Li^6(n,\alpha)H^3$. The combined energy of the alpha-particle and triton is approximately 4.8 MeV, which gives a light pulse which can easily be distinguished from those due to other radiations, which are mainly of lower energy. These workers found that the pulse heights are of the order of one tenth the size of NaI pulses for equal energies.

Similar work has been done by Schenck and Heath (178) using LiI crystals activated with tin.

More recently Nicholson and Snelling (179) have grown LiI crystals with Tl, Sn, Eu and Sm activators, and have studied their properties as detectors of slow neutrons. It was found that LiI(Eu) was the most efficient of these, with a light output about five times that from LiI(Sn) for a given energy dissipation in the crystal. In similar work with gamma-ray sources it was found that the pulse height from a LiI(Eu) crystal was about one third of that from a NaI crystal for a photo-electron of given energy.

2. EXPERIMENTAL PROCEDURE.

The $\frac{1}{2}$ in. cube LiI crystal was mounted in paraffin oil in a polystyrene box which was strapped in optical contact with a photo-multiplier (Dumont 6292). Pieces of 0.0002 in. aluminium foil were placed between the walls of the box and the crystal to act as reflectors. Although a 6 sec. activity is produced in aluminium by the (γ,n)

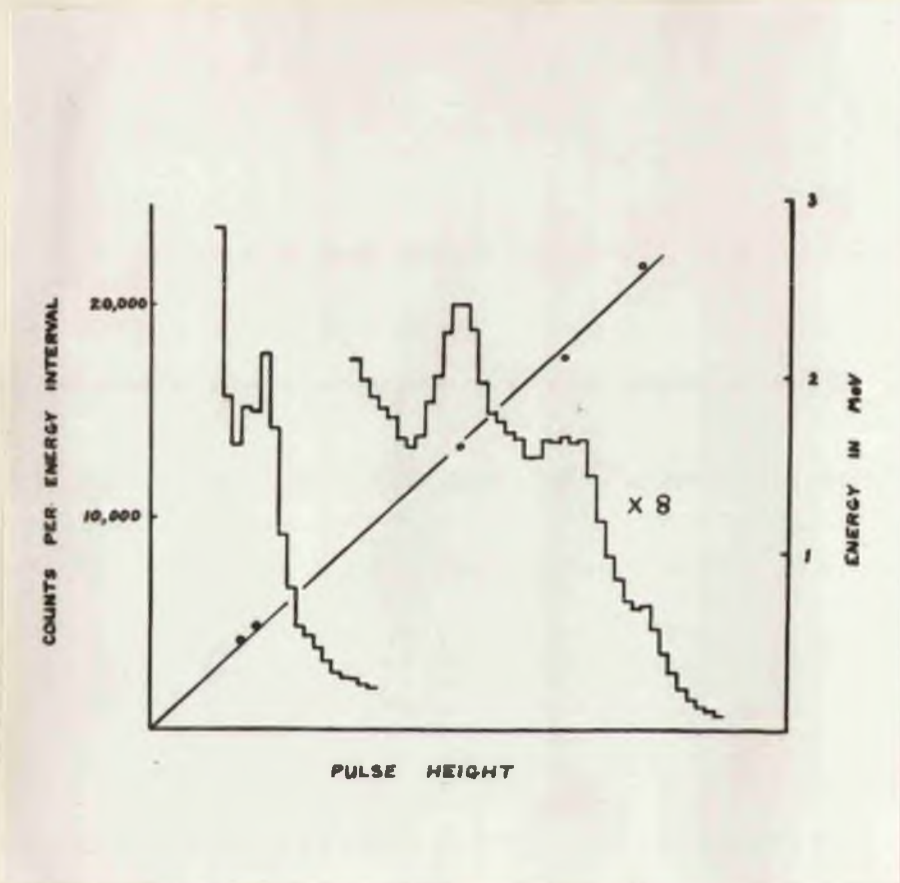


Fig. 21. Radiothorium gamma-ray spectrum obtained with LiI crystal showing peaks at 1.6 MeV (electron pairs), 2.1 MeV (electron pairs + one annihilation quantum) and 2.6 MeV (electron pairs + both quanta); also photoelectric peak at 0.58 MeV.

process, the mass of the aluminium is so much smaller than that of the crystal (~ 0.1 %) that a negligible amount of the activity detected in the crystal is due to this.

Since little work had been reported on the properties of LiI as a detector of electrons, some preliminary measurements were carried out to determine the variation of light output with energy dissipation in the crystal. The pulses from the photomultiplier were amplified and the pulse spectrum analysed with the multi-channel kicksorter. Gamma-rays from Na^{22} and radiothorium sources mounted on top of the crystal were used to produce electrons of accurately known energy. Fig. 21 shows the pulse spectrum obtained with a radiothorium source, which emits gamma-rays with energies of 0.582 MeV and 2.615 MeV. It is seen that the plot of pulse height against energy, as defined by the positions of the peaks, is a straight line within the accuracy of the experiment.

The apparatus was then set up in the beam room of the 30 MeV synchrotron, with the LiI crystal close up to the lead collimator so that most of the beam passed through the crystal, with very little reaching the photomultiplier. The photomultiplier was shielded from the magnetic field of the synchrotron by Mu-metal tubes, and it was ascertained, by taking gamma-ray spectra with the magnet energised, that the apparatus was operating satisfactorily in this position. In order to prevent damage to the photomultiplier the high voltage supply was controlled from the synchrotron control room, and was always switched

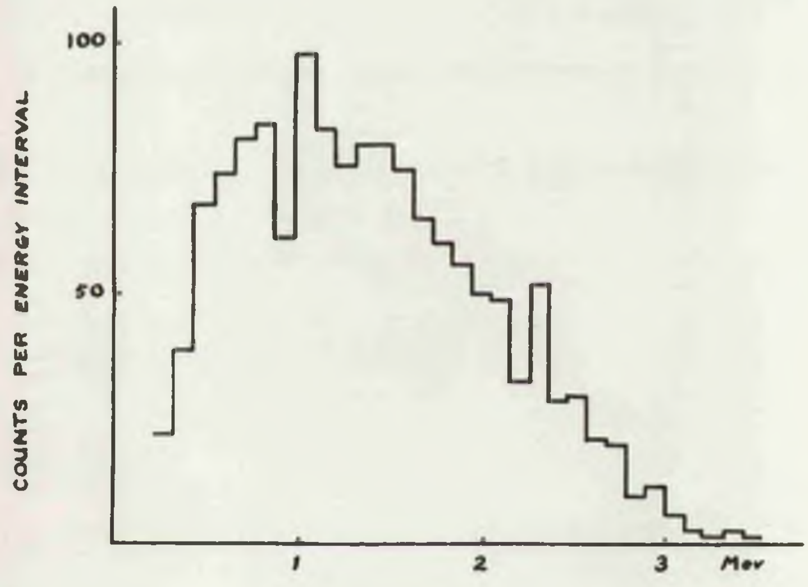


Fig. 22. He^6 beta-spectrum obtained with LiI crystal.

off when the photon beam was passing through the crystal.

In the three preliminary runs which were carried out the following cycle was adopted:

Synchrotron beam on for 3 seconds.

" " off.

Photomultiplier tube E.H.T. on.

Kicksorter on for 3 seconds.

" off.

Photomultiplier tube E.H.T. off.

Synchrotron beam on for 3 seconds.

etc.

In this preliminary work the switching was done manually, but it was intended that this should be done automatically in later measurements. Fig. 22 shows the pulse spectrum obtained in these runs, and, although the statistical errors are large, it has the shape to be expected for the He^6 spectrum. There is a slight deficiency of pulses at energies above 1.5 MeV which is due to beta-particles escaping from the crystal without dissipating their full energy therein, but this effect would be negligible with a larger crystal.

A separate experiment was carried out to obtain an approximate value of the half-life of the activity formed in the crystal. The pulses were fed to a scaler, the display of which was photographed by a constant speed (32 frames/sec.) 16 mm. camera. From the plot of total counts against time the half-life was found to be 0.80 $\pm$ 0.1 seconds, in good agreement with previous more accurate measurements.

Work on this experiment was suspended partly because of breakdowns in the synchrotron and also because of the pressure of other work, but a larger LiI crystal has been obtained and it is hoped to complete this experiment in the near future.

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