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The author is indebted to the following for their assistance in the preparation of this thesis: Mr. J. G. ...
The author is also indebted to the following for their assistance in the preparation of this thesis: Mr. J. G. ...

**THE LIFETIMES OF POSITRONS
IN MATTER**

by

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**Presented as a thesis for the degree of Ph.D
in the University of Glasgow
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P R E F A C E

The writer has attempted to show in this thesis how the scintillation counter may be used for the measurement of short time intervals in nuclear physics and how this technique may be applied to the study of positron annihilation.

In the first section, the significance and the occurrence of short lifetimes in the general field of nuclear physics is briefly surveyed, and the general methods of measuring short time intervals are discussed. A more detailed study is made of positron annihilation. The material of this section is drawn mainly from the literature.

Section two consists of the analysis leading to the design of a delayed coincidence apparatus using scintillation counters. The design and construction of the author's apparatus are described and evidence of its performance is presented and discussed. The analysis and designs which are presented here are generally original though their basic principles are not.

In section three are described the experimental investigations of the lifetimes of positrons in solids. The matter here is original, all the work having been carried out solely by the author who was also responsible for the interpretation of the results.

Section four describes the experimental investigation of the spectrum of annihilation γ -rays from positrons annihilating in neon and oxygen. This work was carried out in collaboration with Mr. G. M. Lewis who had the major responsibility for the experimental work. The interpretation

and discussion were shared equally by us.

Work done by the writer in the field of γ -ray spectroscopy using scintillation counters and a pulse amplitude analyser is presented in Appendix I. The study of Ia^{176} was initiated by Mr. D. Dixon who suggested the application of scintillation counter techniques. The scintillation counter measurements were made by the writer. In Appendix II the final circuits of the coincidence units are shown by diagrams.

Thanks are due to Professor P. I. Dee, Dr. S. C. Curran and Mr. G. M. Lewis for their interest in this work, Professor J. C. Gunn and Dr. G. A. P. Wyllie for helpful discussions on points of theory and the technical staff of the laboratory for their co-operation.

Publications

The majority of the results of sections III and IV have been published in two papers:-

On the Annihilation of Positrons in Solids,
by A. T. G. Ferguson and G. M. Lewis, Phil.
Mag., 44, 1339, 1953:

On the Annihilation Spectrum of Positrons in
Freon and Oxygen, by G. M. Lewis and A. T. G.
Ferguson, Phil. Mag., 44, 1011, 1953.

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SECTION I

GENERAL INTRODUCTION

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CHAPTER I

The historic importance of the study of the time dependence of nuclear processes and a survey of those which might be of short duration.

It was not long after the discovery of Radioactivity by Becquerel in 1896 that the transient and time dependent nature of many radio-active phenomena was appreciated. The study of this aspect of the subject has, over and over again, proved fruitful for our understanding of nuclear processes. In recent years, the introduction of new techniques has made possible the extension of these studies to events of very short duration, and a new field of investigation has thereby been opened up.

The fundamental laws of Radio-active change were deduced by Rutherford and Soddy in 1902 from their studies of the thorium emanation and its active deposit. In their words "..... the activity in each case diminishes regularly with the lapse of time, the intensity of radiation at each instant being proportional to the amount of energy remaining to be radiated." These authors then formulate this law in its well-known mathematical form and go on to explain the more complex cases of growth and decay of activity encountered in their experiments.

From the laws for large numbers of atoms, Rutherford postulated the law for an individual atom in terms of probability. He proposed that for each species of radio-active atom, there was a constant probability per unit time of disintegration. The reciprocal of this quantity can readily be shown to be the "mean life" of an atom.

After the discoveries of Rutherford, the mean life became one of the most important characterising properties of a radio-active species and the

measurement of the variation of activity with time was one of the most powerful techniques in the unravelling of the natural radio-active series.

The relationship of mean life to other quantities.

Even within the relatively limited field of the natural radio-elements which emit α -particles, the magnitudes of the mean lives encountered vary from 1.3×10^{10} years in the case of Thorium, to 3×10^{-7} s. for Th.C¹. This would seem to indicate a large variation in the degree of instability of active nuclei.

It was found by Geiger, (1910), that the α -particles from a particular material had a definite range in matter. Here was another measure of nuclear instability. Geiger and Nuttall (1911) related the range of the α -particle and the mean life of the emitting nucleus by an empirical expression

$$\log \lambda = A + B \log R$$

$$\lambda = \frac{1}{\text{Mean life}}$$

$$R = \text{Range}$$

$$A, B = \text{Constants}$$

A theoretical explanation of this law by Gamow (1928) was the first successful application of quantum mechanics to nuclear physics. Gamow's expression (see page 6) depends strongly on the nuclear radius. Consequently, a measurement of the mean life of an α -emitting nucleus gives a value for this important quantity. There are many other examples of similar relationships of the mean life to fundamental properties of nuclei. In β -decay the product of the lifetime and a function of the maximum energy should have a constant value which indicates the degree of forbiddenness. This

constant also is a measure of the magnitude of the β -nucleon coupling constant. Similarly, the mean lives of mesons indicate the strength of meson interactions. The long mean life of some nuclear isomers drew attention to the emission of γ -rays by higher multipoles (Von Weissäcker, 1936).

In all these fields the measurement of mean life has been a most valuable tool. The measurements referred to have, in the main, been made by observing the change in activity with lapse of time, the intervals being long enough to be measured with a clock or stop-watch. New methods of timing have made it possible to study processes of very much shorter duration where the time intervals may be as short as 10^{-10} s. It would be of interest to consider what nuclear and fundamental particle processes might have short mean lives.

Before discussing particular examples of processes which might have short mean lives, there are some general relationships that are important here. Any quantum mechanical system is described by an eigenfunction, whose eigenvalues give the energy states of the system. For stable systems, these eigenvalues are definite or "sharp". When there is a finite probability of decay this is represented by a time dependent exponential term in the eigenfunction, which gives rise to an "uncertainty" in the eigenvalues. The mean width of the energy uncertainty of the state, denoted by Γ , is related to the mean life τ by the equation

$$\Gamma \tau = \frac{1}{2\pi} \times \text{Planck's constant}$$

Thus, a state having a mean life of 10^{-12} s, has a mean width of about 4×10^{-3} e.v.

The probability of decay per unit time, λ , is equal to $\frac{1}{\tau}$

and so

$$\Gamma = \frac{h \lambda}{2 \pi}$$

If a state can decay in several alternative ways which have respectively probabilities $\lambda_1, \lambda_2, \lambda_3, \dots$ then the total probability per unit time of decay is $\lambda = \lambda_1 + \lambda_2 + \lambda_3 + \lambda_4 + \dots$

Thus, the total width of the state is the sum of the partial widths for the various modes of decay. In any experiment, it is the total width of the state which we will measure. The partial widths may be found from the relative intensities of the various modes of decay.

Nuclear and Fundamental Particle processes of short duration.

There are a great many time dependent nuclear processes. Natural and artificial radio-elements emit α and β particles and de-excite with the emission of γ -rays. The compound nucleus, the intermediate state in nuclear reactions, is known to emit neutrons, protons and other charged particles. All the known mesons decay spontaneously into other species. Electromagnetic interactions of γ -rays with charged particles have intermediate states of finite durations. Positrons in matter annihilate with electrons producing two or more photons. Short lifetimes could be associated with many of these processes and their expected magnitudes will now be discussed briefly.

Nuclei may be found in either their ground or in one of a series of excited states. The first excited states of light nuclei lie about 1 M.e.v. above the ground state, while for heavy nuclei, the first level (A. Bohr

and Mottelson, 1951) may be only a few tens of kilovolts above the lowest state. In heavy nuclei, and with excitation energy of the order of 8 M.e.v., levels are about 1 e.v. apart.

When the excitation energy of a nucleus is less than the separation energy of any nucleon or group of nucleons, the state can decay only by the emission of electromagnetic radiation. When sufficient energy is available, the emission of heavy particles and γ -rays respectively are competitive processes.

If a bombarding particle enters a nucleus, the strong interaction between nucleons rapidly causes its binding energy and kinetic energy to be shared among the nucleons, and the resulting compound nucleus (Bohr 1936) "forgets" how it was formed. Except for its high degree of excitation, (~ 8 Mev for proton or neutron capture), it does not differ from an ordinary nucleus. This nucleus is energetically unstable. A more stable configuration may always be reached by the emission of particles or γ -rays.

Such a nucleus has a finite lifetime due to two factors. Firstly, for a particle to be emitted it must, in the random collision processes within the nucleus, acquire an appreciable fraction of the energy of excitation. Secondly, such a nucleon is prevented from leaving the nucleus by a potential barrier for which there is only a definite probability of penetration. For charged particles the high coulomb barrier makes this probability small. The emission of particles with spin is inhibited by the centrifugal barrier and all particles may be reflected at the change of potential.

Blatt and Weisskopf (Theoretical Nuclear Physics, page 389) give the formula

$$\lambda = \left(\frac{4R}{K} \nu_e \right) \left(\frac{D}{2\pi\hbar} \right)$$

k = wave number of the particle outside the nucleus

K = wave number of the particle inside the nucleus

V_1 = penetrability of centrifugal or coulomb barrier.

D = average spacing between levels of the same spin and parity.

The first bracket represents the probability of emission of a particle with suitable energy, the second represents the frequency with which this energy is concentrated on one particle.

For a neutron of 20 ev. and $D = 10$ ev., the mean life of neutron emission is about 10^{-10} s. For higher energies the lifetime is correspondingly shorter. For protons of 3 Mev. channel energy, the mean life will exceed that for a corresponding neutron by a factor of 40. ($Z = 20$, see Blatt and Weisskopf, p. 363).

In competition with these processes, γ -rays may be emitted. The lifetime for emission of γ -rays such as those following neutron capture is estimated to be about 10^{-12} s. Thus the mean life of the compound nucleus cannot be ^{greater} less than this.

In a number of heavy nuclei, the separation energy of an α -particle is negative and these particles may be emitted from the ground state. No competing processes are present in this case. Gamow (1928) calculated the emission probability here and found

$$\lambda = 10^{21} e^{-G}$$

$$G = \left(\frac{8\pi\gamma Z e^2 b}{\hbar^2} \right)^{\frac{1}{2}} \left[\cos^{-1} \left(\frac{R}{b} \right)^{\frac{1}{2}} - \left(\frac{R}{b} - \frac{R^2}{b} \right) \right]$$

Z = nuclear charge

where

- z = charge of emitted particle
 b = width of the potential barrier for a particle with energy E
 R = nuclear radius

With a suitable value of R this formula gives the correct order of magnitude of λ for a very wide range of elements. Amongst the heavy nuclei the shortest lived α -emitter is ThC^1 which has a mean life of 3×10^{-7} s. (Dunworth 1939). The energy of α -particles from the ground state of this nucleus is greater than those from the ground state of any other nucleus. This is therefore likely to be the shortest mean-life that may be found amongst the heavy α -emitters. In the light elements however the low potential barrier of the Be^8 leads one to expect a mean life in the region of 10^{-16} s.

The emission of γ -rays.

γ -rays are emitted when a nucleus goes from a higher to a lower energy level. For the purpose of calculation the nucleus is considered as an oscillating system of charges and currents and its radiation expressed as the sum of a series of electric and magnetic multipoles. Angular momentum and parity selection rules determine the lowest multipole order that may contribute.

For electric transitions with a change of energy E and of angular momentum l , Blatt and Weisskopf give (p. 627)

$$\lambda = \frac{4 \cdot 4 (l+1)}{e^2 [(2l+1)!!]^2} \left(\frac{3}{l+3} \right)^2 \left(\frac{E}{197 \text{ Mev.}} \right)^{2l+1} R^{2l} \times 10^{21}$$

R = nuclear radius in units of 10^{-13} cm.

This formula is deduced on the basis of the independent particle model of the

nucleus. λ varies rapidly with l . For E about 200-kev and $R = 6 \times 10^{-13}$ cm changes by 10^5 for a unit change in l .

The corresponding magnetic transitions are less probable by a factor of about 1,000. A transition of energy 200-kev and $l = 1$ i.e. electric dipole would have a mean life of 5×10^{-14} s., while corresponding magnetic dipole or electric quadrupole transitions would have mean lives of about 5×10^{-10} s., and 5×10^{-9} s. respectively. The wide spacing of the lifetimes for the various transition types leads one to hope that a measurement of γ -ray mean lives in this energy region would lead to the assignment of spins and parities.

Internal conversion

For transitions with energies below a few hundred kilovolts orbital electron capture competes strongly with the emission of γ -rays and the mean lives of these states are therefore reduced. The ratio of the number of conversion electrons to the number of corresponding γ -rays, i.e., the conversion coefficient, increases with z and l and decreases with E . The order of magnitude of the mean life for this process is seldom more than an order of magnitude different from that for γ -ray emission in the region where it is important.

β -Ray Emission

The lifetime of a β -emitting state decreases slowly with increasing transition energy. The β -particles of Li^8 have an energy limit of 12-Mev. (Lewis, Burcham and Chang 1937) and a corresponding mean life of 0.11s. Hence it is unlikely that β -emitting states with lifetimes of less than 10^{-6} s will exist. Due, however, to the slow variation of transition probability with transition energy, daughter nuclei are frequently formed in their lower excited states. This fact has greatly facilitated the study of the mean lives of such states against de-excitation.

Electromagnetic and Other Processes

Electro-magnetic processes, e.g., Compton effect, proceed through intermediate states. The intermediate state in the Klein-Nishina treatment of the Compton effect has a lifetime of about 10^{-20} s., while on the alternative theory of Bohr and Kramer the times of appearance of the electron and scattered quantum are unrelated.

It is not proposed to discuss meson lifetimes further than to note how closely they are grouped within a few orders of magnitude of 10^{-10} s.

The lifetimes predicted by electro-magnetic theory for positrons range from about 10^{-9} s. for solids to 10^{-7} s. for gases on the simplest theory. The magnitudes to be expected depend greatly on the physical assumptions made and their correctness verified by experiment. This subject will however be discussed in more detail in Chapter II.

In Chapter VI we shall see that, using scintillation counters, lifetimes of the order of 10^{-10} s. may be measured. From the above discussions we see that two processes other than the decay of mesons have lifetimes of this order uncomplicated by much faster competing processes. The first is the emission of γ -rays from the lower excited states of nuclei, the second the annihilation of positrons in solids.

States having a measurable lifetime for the emission of γ -rays are to be expected amongst the isotopes of medium and heavy nuclei and are most simply reached (from an experimental point of view) by the decay of β -active isotopes. Many of the latter have half-lives of one day or less and it was felt that such studies could best be pursued where a pile was available.

Although some low lying levels produced in H.T. set reactions were thought to warrant study, e.g., the 30 keV level in A^{28} (Smith and Anderson, 1951), it was felt that at the time the technique of time measurement was not sufficiently far developed for experiments with machines.

Further consideration of the state of knowledge of positron annihilation showed the need for further study in this field. For this work a long-lived source of positrons Na^{22} was available, and the relatively high energy of the nuclear and annihilation γ -rays involved suited the state of development of the apparatus. This field of study was therefore chosen as the main subject of the work of this thesis.

CHAPTER II

Prior to the year 1932, the known fundamental particles of nuclear physics were the massive, positively charged proton and the light electron of opposite charge. The asymmetry of this situation made inevitable, speculation on the possible existence of negative protons and of electrons with positive charge.

The existence of positive electrons was predicted theoretically by Dirac (1930) in his "hole theory". In this paper, Dirac, though he refers to the particles as protons, takes their mass equal to that of the electron and deduces many properties of the positron which have since been verified experimentally. The first experimental proof of the existence of light positively charged particles was that of Anderson (1932) and this was verified by Blackett and Occhialini (1933) in the following year. These workers identified the positron by the curvature of its track in a cloud chamber situated in a strong magnetic field. Curie and Joliot (1933) discovered that many artificial radioactive isotopes were positron emitters. This greatly facilitated the experimental study of positrons in the laboratory.

The Hole Theory

Dirac's relativistic wave equation for a free electron has solutions corresponding to states in which the energy of the particle is negative. This arises from the double valued nature of the square root, i.e.

$$E = \pm \sqrt{(mc^2)^2 + p^2 c^2}$$

m = mass of electron
p = momentum of
electron
E = energy of Elec-
tron

Clearly an electron cannot have energy between $\pm mc^2$. If an electron of energy $+ E$ should jump to the level $- E$, the energy difference, viz. $2 E$ would appear as radiation. In seeking the lowest available level, we should

expect all electrons to jump into the negative energy states. To overcome this difficulty, Dirac proposed that, normally, all the states of negative energy should be filled with electrons, though these electrons should not contribute to the total energy and momentum of the system. The absence of one of these electrons manifests itself as a positively charged particle of positive energy, i.e., a hole is equivalent to a positron.

The conception of the positron as a vacancy or "hole" in the negative energy states of electrons suggests two of its most important properties. First, by the action of an electro-magnetic field, e.g., a γ -ray, a positron-electron pair may be created. By raising one of the infinite sea of electrons in -ve energy states to a state of positive energy, there appears an electron and a hole - in other words, a positron-electron pair. Secondly, if we have initially an electron and a positron, the latter represents a vacancy in the negative energy states into which the electron may fall, with the emission of energy in the form of γ -rays.

Using the hole theory, it is possible to predict the rate of annihilation of positrons. The simplest case is that in which a free positron annihilates in collision with a free negative electron. It may be shown (see e.g. Fermi, page 54) that energy and momentum can be conserved in such a process only when the energy of annihilation is emitted in the form of two or more quanta. When the electron is tightly bound to a nucleus these considerations do not apply, as the nucleus may act as a momentum sink. Hence under these conditions, annihilation with the emission of a single quantum may occur. This process is relatively unimportant except for positrons of high energy in heavy materials (see Heitler, page 209). This effect will not be considered further here.

The two quantum process in which an initial state of a positron and electron goes to a final state of two quanta is a second order process, i.e., the matrix element for the direct transition is zero and hence it can only take place through an intermediate state. Similarly, annihilation processes in which three or more quanta are emitted are of third or higher order. Since the probability of a process decreases rapidly with increasing order, the most important and probable event is two quantum annihilation.

Two Quantum Annihilation

The calculation of the annihilation probability per unit time for a free positron and electron uses a perturbation method. The probability per unit time P is given by

$$P = \frac{2\pi}{\hbar} |H_{IF}|^2 P_F \quad \left\{ \begin{array}{l} \text{See e.g.} \\ \text{Schiff p. 193} \end{array} \right\} \dots\dots\dots (1)$$

\hbar = Planck's constant

P_F = density of final states per unit energy interval

H_{IF} = matrix element for the transition

For a second order process involving an intermediate state, H_{IF} is of the form

$$H_{IF} = \frac{H_{IB} H_{IF}}{E_I - E_B} \dots\dots\dots (2)$$

E_I, E_B are the energies of the initial and intermediate states respectively.

H_{IB} , H_{IF} are the matrix elements for the transition from the initial to the intermediate and intermediate to final states respectively.

The summation is over all spin directions and energy signs. It is found that only those cases in which the initial spin is zero give non-vanishing contributions to H_{IF} , i.e., two quantum annihilation from the triplet state is forbidden.

If the calculation is performed in detail, representing the incident positron as a plane wave, the formula first deduced by Dirac is obtained for the annihilation cross-section.

The positron has energy E_+ and the electron is at rest.

$$\phi = \pi r_0^2 \frac{1}{\gamma + 1} \left[\frac{\gamma^2 + 4\gamma + 1}{\gamma^2 - 1} \log(\gamma + \sqrt{\gamma^2 - 1}) - \frac{\gamma + 3}{\sqrt{\gamma^2 - 1}} \right] \dots \dots \dots (3)$$

$$\gamma = \frac{E_+}{m_0 c^2}$$

$$r_0 = \text{classical radius of the electron} \\ = \frac{e^2}{m_0 c^2}$$

The above formula describes the variation of the annihilation cross-section with energy. During a fraction of its path dx the positron has energy E . The probability of annihilation with energy E in dx is

$$P = \phi(E) dx \quad (\text{for unit electron density}) \\ = \phi(E) \left(- \frac{dx}{dE} \right) dE \\ = \frac{\phi(E)}{-\frac{dE}{dx}} dE$$

Assuming that the electron density is NZ (where N = no. of atoms per c.c.) and using his calculated values of $\frac{dE}{dx}$, Heitler (p. 231, fig. 6) shows that in lead, less than 3% of positrons of initial energy 500 keV annihilate in flight. The remainder annihilate at rest and this is the dominant process.

When the positron kinetic energy becomes small, i.e. $\gamma \rightarrow 1$, the cross-section ϕ diverges as $\frac{1}{\text{velocity of positron}}$. Therefore the probability per unit time for annihilation $v \times \phi$ becomes constant.

$$v\phi \rightarrow \pi r_0^2 D \quad \text{for } v \rightarrow 0 \text{ i.e. } \gamma \rightarrow 1$$

Hence in an electron atmosphere of density n per c.c., a slow positron has a probability of annihilation per unit time $n\pi r_0^2 C$

$$n\pi r_0^2 C = \text{constant} = \lambda \text{ say}$$

The emission of γ -rays in the annihilation of a slowed down positron is thus analogous to the emission of radiation by a nucleus where the probability of emission of radiation is also constant. A slow positron may be said to have a mean life in the same sense as an excited nucleus.

Numerical substitution shows that $v\phi$ is sensibly constant below a few keV. The time to reach such energies is calculable roughly from the range, energy relationship for electrons. In aluminium for example it is not greater than about 10^{-11} s. for 500 keV positrons, while the expected lifetime of the slow positron is at least one order of magnitude greater. We may thus speak of the mean life of the positron in the material.

Three quantum annihilation of a positron and a free electron

As was pointed out above, three quantum annihilation is a third order

process. No general formula corresponding to equation (3) has been deduced but the limiting case for small velocities has been worked out by Ore and Powell (1949). The general method of calculation is the same as the two quantum case.

The probability of annihilation per unit time P is given by a formula similar to equation (1) and Ore and Powell show that this quantity differs from zero only when the initial spin state of the positron and electron is a triplet.

At small velocities the cross-section is proportional to
$$\frac{1}{\text{velocity of positron}}$$

Ore and Powell calculate the ratio of the two quantum annihilation cross-section to be 1100 : 1. As, however, triplet states have three times the statistical weight of singlet states, three quantum annihilation is favoured by this factor in free collision. Under these circumstances

$$\frac{\text{Number of three quantum annihilation}}{\text{Number of two quantum annihilation}} = \frac{1}{370}$$

The methods and assumptions of the above theory are closely related to the calculations of the Klien-Nishina theory of the Compton effect. The predictions of the latter have been thoroughly verified experimentally. We should therefore expect the positron calculations to be sound as far as they go. Nevertheless, their experimental verification, especially of the third order processes, would be of itself of some interest. Some indirect experiments however suggest that the application of these results to real materials may involve complications which add further interest to these studies.

Simple theory applied to real materials

If λ is the probability of annihilation per unit time

$$\lambda = \lambda_2 + \lambda_3 \quad \text{where}$$

λ_2 = probability of two quantum annihilation

λ_3 = probability of three quantum annihilation

$$\lambda_3 = \frac{\lambda_2}{370}$$

$$\therefore \lambda = \lambda_2 + \frac{\lambda_2}{370} = \lambda_2$$

Thus we should observe a single lifetime approximately equal to the two quantum lifetime.

In a rough attempt to estimate positron lifetimes in materials, Heitler (p. 208) assumed that to the slow positron the material appeared as an electron atmosphere of density NZ electrons per c.c. where N = no. of atoms per c.c. Since $Z = \frac{A}{2}$

$$\lambda_2 \propto NZ \propto \frac{NA}{2} \propto \text{density of the material}$$

Therefore, the lifetime observed should be inversely proportional to the density of the material and in the case of gases to their pressure.

Typical values to be expected on this view are given in tables I and II below.

TABLE I (Solids)

Material	Li	H ₂ O	Al	Fe	Pb
τ	10×10^{-10} s.	5×10^{-10} s.	1.8×10^{-10} s.	0.6×10^{-10} s.	0.5×10^{-10} s.

Gases at N.T.P.

Gas	H ₂	O ₂	Xe	CO ₂	CCl ₂ F ₂
τ	$2.5 \times 10^{-6} \text{ s.}$	$3 \times 10^{-7} \text{ s.}$	10^{-7} s.	$2 \times 10^{-7} \text{ s.}$	10^{-7} s.

The effect of Coulomb forces

The main defect of the theory presented above and the difficulty of applying it to real material arises from the neglect of the coulomb forces acting on the positron. An attractive force increases the concentration of positron wave function while a repulsive force has the opposite effect.

The repulsive force between the positron and a nucleus decreases the positron amplitude in the region of the inner electron shells and hence makes annihilation with these electrons improbable. This has the effect of reducing the number of electrons available for annihilation and consequently increasing τ . There is some experimental evidence (which will be discussed later) that only the valence electrons are available.

On the other hand, the attractive force between a positron and electron would tend to increase the annihilation probability, i.e., decrease the lifetime. In the absence of general solution of the wave equation of a positron and electron, no accurate theory of this effect has been worked out. A slightly better approximation suggested by Heitler (p. 84)

$$\psi = \psi_{\text{plane}} \times f\left(\frac{r}{a_0}\right)$$

$$f = \frac{Ze^2}{h\nu}$$

ν = vel. of positron.

The correction is clearly strongly velocity dependent (see e.g. Heitler, p. 84) and hence the lifetime would depend on the energy distribution of the available electrons in the material and so in the solid state would be influenced by the material structure.

For an adequate solution of this problem the interaction of the positron with the whole atom would need to be considered and in condensed matter the atomic interactions would be an added complication.

An experimental study of positron lifetimes might be expected to show which of the opposing effects discussed above is the more important. The crude picture first presented (P₁) does, however, lead one to expect lifetimes that might be measurable by the techniques discussed in Chapter V

A further effect of the attractive force would be to make possible the formation of bound states of the positron and electron. This bound system is known as positronium. If positronium formation was strongly competitive with other modes of annihilation, the positron lifetime would become largely independent of the material in which it was taking place. The properties of this peculiar and intriguing atom will now be discussed.

Positronium

The possibility of a bound positron-electron state was first pointed out by Ruark (1945) who named such a system "Positronium". In 1946 Wheeler was attempting to explain mesons as aggregates of positrons and electrons, calling them polyelectrons. He started by investigating the simplest of such systems, the bi-electron. He showed that positronium was stable by 6.77 ev. against dissociation and should have a half-life against annihilation considerably longer than its period of revolution.

A very detailed study of positron-electron interactions has been made by Pirene (1946) in a series of papers in the last of which he discusses positronium.

Positronium may exist in either a singlet or triplet state depending on whether the spins of the particles are anti-parallel or parallel respectively. As has been pointed out above, annihilation by two quantum emission is forbidden when the spins are parallel. In this case annihilation is by three quantum emission. It has been shown by Wheeler that the lifetime against annihilation (two quantum) of all S states is

$$\bar{\tau}_2 = 1.25 \times 10^{-10} \times n^3 \text{ sec.} \quad \text{where } n \text{ is the principal}$$

quantum number. The fact that this increases rapidly with n , while the lifetime against optical de-excitation decreases, suggests that most of the annihilation will be from the ground state.

The annihilation quanta will share the rest energy of the positron and electron equally and will be emitted in opposite directions.

The lifetime against annihilation from the triplet state is calculated by Ore and Powell (1949) to be $1.4 \times 10^{-7} \times n^3$ sec. Simple dynamics show that the three quanta must be coplanar and that the greatest energy that any one photon may have is $M_0 c^2$.

We should therefore expect the annihilation γ -rays from triplet positronium to have a continuous spectrum. Ore and Powell have deduced the shape of this spectrum. This is discussed more fully in Chapter XI.

Positronium is formed in the triplet and singlet states in proportion to the statistical weights $2S + 1$ of the states.

$$\text{i.e. } \frac{\text{no. of atoms in triplet state}}{\text{no. of atoms in singlet state}} = \frac{2 \times 1 + 1}{2 \times 0 + 1} = \frac{3}{1}$$

Thus we should expect to be associated with this phenomenon two independent half lives and a large proportion of three-quantum annihilation. The half lives which would be observed might differ from those predicted by the theory above owing to phenomena, e.g. electron exchange, whereby a positronium atom in a triplet state might change to a singlet state or vice versa. Let λ_1 + λ_3 be the probability of annihilation per unit time from singlet and triplet states respectively, and a , be the probability per unit time of a transition from one state to the other. Suppose positronium is formed in the triplet state. Probability of annihilation

$$\begin{aligned} \text{per unit time} &= \lambda_3 + a\lambda_1 + a^2\lambda_3 + a^3\lambda_1 + \dots \\ &= \lambda_3 + a\lambda_1 \text{ if } a \text{ is small.} \end{aligned}$$

Similarly for positronium formed in the singlet state.

$$\begin{aligned} \text{Probability of annihilation per unit time} &= \lambda_1 + a\lambda_3 + a^2\lambda_1 + \dots \\ &= \lambda_1 \text{ if } a \text{ is small since } \lambda_1 \gg \lambda_3 \end{aligned}$$

These considerations lead us to expect one lifetime of about 1.25×10^{-10} s. for the singlet state and another lifetime lying between this value and 1.4×10^{-7} s. for the triplet state. If positronium were formed in a condensed material, one might expect compression of its wave function and consequent departures from the times discussed above, but the characteristic of two independent lifetimes would not be altered. They might, however, not be resolved experimentally.

If the probability of formation were high, one would expect the lifetime of the positron annihilation from the bound singlet state to be independent of the material.

While much theoretical work has been published regarding the annihilation of positronium and about its stable states, no calculations have been made of the cross-section for its formation in practical cases. Such work would seem to be very valuable. The formation of positronium would take place in competition with direct annihilation in free collisions and would thus reduce the free annihilation lifetime.

A number of other suggestions have been made regarding possible positron annihilation reactions. The formation of compounds in which a positron replaces a hydrogen ion, e.g., β^+Cl^- (Ore, *Hylleraas*, 1948). The theoretical analyses concern themselves mainly with stability of such compounds, and not with the likelihood of formation and decay. It would seem probable that if formed they would possess a single lifetime of the same order as that of positronium.

From these discussions it is certainly clear that the simple picture presented on page 17 is unlikely to give more than an order of magnitude for the positron lifetime and that the additional phenomenon of positronium formation may introduce two further lifetimes.

The effects of solid state structure both in the case of free annihilation and positronium formation would be difficult to analyse accurately by theoretical means, and would make the annihilation processes there more complicated and consequently more difficult to interpret experimentally than in the case of gases.

The decision to study the annihilation of positrons in solids despite its complication was made because at the time these experiments were begun there was practically no direct measurements of positron lifetimes in solids, while Deutsch had begun to publish his now well-

known experiments which established the formation of positronium in gases.

The experimental study of Positron lifetimes

At the time the writer's work began, the generally accepted picture of positron annihilation was the rather crude one described on page above. A few experiments had been done to investigate its validity and their results may be summed up as having cast considerable doubt on this interpretation without being able to replace it with a more certain one.

In several gases, Shearer and Deutsch (1949) measured a lifetime which was shorter than that predicted by the formula

$$\frac{1}{\tau} = \pi r_0^2 C \times NZ \quad (N = \text{no. of atoms per c.c.})$$

by a factor of about 2. Further, they found that the lifetime did not vary with pressure in the expected manner, i.e., inversely. They suggested that positronium might be formed and later were able to establish this very elegantly (Deutsch 1951).

The most definite evidence about annihilation in solids at this time came from indirect experiments. These sought to examine the velocity distribution of the positron-electron systems at the time of annihilation. In two quantum annihilation the motion of the positron-electron centroid would show itself in two ways. First, the γ -rays would not be emitted at exactly 180° . The deviation from 180° would depend on the velocity of the centroid and the angle between its direction and that of one of the γ -rays. Thus, corresponding to a given

velocity, there would be a characteristic distribution about 180° whose width would be a measure of the velocity. Allowance would require to be made for the finite angular resolution of any measuring system. Such an experiment was carried out by DeBenedetti et al. (1950) who used two NaI scintillation counters in coincidence. The crystals were 120 cm. apart, subtending only a few minutes of arc at the source on the mid point of their axis. The source was Cu^{64} wrapped in the minimum quantity of gold needed to stop the positrons in order to reduce scattering. The dimensions of the source were such that it also subtended only a few minutes of arc. By moving one of the counters transversely and plotting the number of coincidences against angle, the angular distribution was measured. From this experiment it was concluded that the motion of the centroid did not correspond to an energy greater than a few e.v. It was therefore suggested that only the outermost shell electrons took part in the annihilation process. This would result in lifetimes an order of magnitude greater than those expected if all Z electrons were available.

The second effect of the motion of the centroid would be a slight spread in the energy of the annihilation γ -rays for $\text{Cu}^{64}\beta^+$ annihilating in Cu. Using a very high resolution curved crystal spectrometer, Du Mond, Lind and Watson (1949) studied this effect. Allowing for all instrumental broadening of the line, the residual effect could be explained by assuming an energy at annihilation of less than 4 e.v., in good agreement with DeBenedetti's experiment. These authors point out that in copper the only electrons having energies of this order are the conduction electrons which have a distribution of energies up to 7 e.v.

Assuming only the conduction electrons to be available, the lifetime expected would depend on their density. A large difference of the order of several times 10^{-9} s. was expected between the lifetime in lead and in potassium say which has a small number of such electrons. A measurement of the difference in lifetime using a delayed coincidence technique was reported by DeBenedetti and Richings (1951) in abstract form. Contrary to the result expected, they found the difference to be not greater than 10^{-9} s.

Moore (1951) reported a lifetime of $1 \pm .5 \times 10^{-9}$ s. for positrons in stilbene and Millet a lifetime of about 10^{-9} s. in plexiglass, both results claimed to be in agreement with an "all electron" theory.

From a study of these rather inconclusive and indeed conflicting results, it appeared that much more work was required and that in particular more accurate measurements were needed before ~~some~~ a quantitative picture could be obtained.

CHAPTER III

Methods of measuring short time intervals in Nuclear and Fundamental Particle Physics

There are two general approaches to the problem of measuring lifetimes of unstable nuclei and fundamental particles. As it was pointed out in Chapter I, page 3 the lifetime and the level width of an unstable nucleus are related by the uncertainty principle. Experiments may therefore measure either quantity. The measurement of very short times and very narrow widths presents considerable experimental difficulty. It is perhaps fortunate therefore that these quantities are reciprocal. In one case, viz. the 411 kev. state of Au^{198} , both the level width (Davey and Moon, 1953) and the lifetime (Bell et coll. 1952) have been measured but, in general, only one quantity has been experimentally accessible.

Measurement of Widths of Nuclear Levels.

When the width of a level excited by charged particle bombardment is large compared with the homogeneity of the incident beam, a measurement of the excitation function for the reaction at the resonance may give a value for the level width. Many widths have also been deduced in reactions initiated by slow neutrons. Here, the high energy resolution obtainable with mechanical velocity selectors enables widths of hundredths of an ev. to be measured accurately, e.g., the 1.44 ev. resonance in In^{115} which has a neutron width of 2.4×10^{-3} ev. and a γ -ray width of .09 ev. (Havens et al. 1947).

An indirect application of width measurement is that of Elliot and

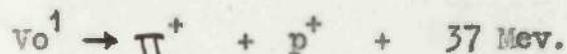
Bell (1949). The width of a γ -ray line may be increased by Doppler effect if the nucleus from which it is emitted is still in motion after particle emission. The γ -ray chosen was that from the 479 kev. state of Li^7 and the width was measured using a high resolution β -ray spectrometer. From the width, the velocity of the nucleus and hence the time of emission, the mean life of 0.75×10^{-10} s. was deduced.

Direct Methods.

A great variety of direct methods of measuring lifetimes have been described in the literature and a few typical examples will be discussed, illustrative of the more important techniques.

When the unstable particle has high energy and either is, or decays into an ionizing particle, the cloud chamber is amongst the best instruments for lifetime measurements. This application of cloud chambers is discussed by Wilson and Butler (1952). They show that for each event we require a knowledge of three quantities - x , x_0 and p , where x is the distance travelled in the chamber before decay, x_0 is the maximum possible path length in the chamber, and p is the momentum of the particle. From a knowledge of these quantities and applying the analysis of Peirls (1935), an estimate may be made of the lifetime.

e.g. For the Vo^1 particle (Page and Newth, 1954)



From the mechanics of the disintegration the momentum and direction of the Vo^1 particle may be deduced. From this knowledge, and the geometry of the events in the chamber, the mean life of 3.7×10^{-10} s. was obtained.

Similar techniques are possible with photographic emulsions. One of the earliest measurements of the π^+ lifetime was made using photographic plates arranged round the circumference of a cyclotron tank in which the mesons were produced. The radius and field determined the momentum of the particles and the number decaying in each plate gave a measure of the distances travelled before decay.

Such techniques are not suitable when the momentum of the unstable particle is low and it is being rapidly brought to rest, e.g., a compound nucleus. Here the particle is brought to rest in an immeasurably short length and it is not certain that emission occurs before the nucleus comes to rest. It is also unsuited for the study of positron annihilation at low energies due to the scattering of the particle in numerous collisions. For high energies it offers the most direct if rather tedious method of studying annihilation in flight.

Where the photographic techniques are inapplicable, counters may sometimes be used. By accurate collimation they may be made sensitive only to radiations from a limited region of space. This may be used to define the distance travelled (say by an unstable nucleus, before particle or γ -ray emission), from a target in which the nucleus is produced. Its velocity may be deduced from the dynamics of the reaction.

This type of method was used by Devons et al (1949) to measure the lifetime of the pair emitting state of O^{16} . This they found to have a mean life of about 10^{-16} s. Since it is necessary to assume that the decaying body moves in a straight line, the method could not be applied to the study of slow positrons due to their tortuous paths and short ranges.

Yet another method - the use of electronic delayed gates, finds its application when the particles are produced in short bursts, e.g. by a synchrotron or linear accelerator, or by an interrupted neutron beam from a pile.

Here a counter is used to detect the emitted secondary particles. Its output is fed to a block of electronic delayed gates, the first of which is made sensitive by a pulse from the machine. The latter pulse defines the instant of creation of the states. The other gates open successively at fixed intervals and count the number of states decaying at that time. From the number of pulses in each gate, the lifetime of the state may be deduced. This technique has been applied by Reid and McNeil using the 30 Mev. synchrotron at Glasgow.

Lastly we wish to consider the method of delayed coincidences using counters. Here, a nuclear event detected in a counter announces the "birth" of the state, and the detection of the decay radiation announces its "death". In order to pick out the events in which one is interested, separate counters (A and B say) may be used to detect the announcing and decaying radiations. This enables one to use geometry and shielding to increase the selectivity of the counters.

A coincidence circuit gives an output pulse only when pulses from A and B reach it simultaneously. The amount by which event A has to be delayed in order for it to coincide in time with event B is a measure of the delay in emission of B. In the measurements described above, e.g., those using cloud chambers, the velocity of the unstable secondary particle itself provided the scale for time measurements. Here the velocity of an electrical pulse in a cable or delay network is taken as

a secondary standard. This velocity may be measured with very great accuracy by resonance techniques and errors from this source can be made very small.

This method has wide general applications. It is particularly suited to the study of delays between light particles and γ -rays for which the other methods are unsuitable, and it has been widely applied to the study of the lifetimes of isomeric states.

Delayed coincidence techniques would seem well suited to the study of positron annihilation. Either the positron itself or a nuclear γ -ray associated with it could mark the emission of the particle, and the annihilation γ -rays could mark its "death". We would thus require an apparatus suitable for detecting β - γ or γ - γ coincidences, and, in the latter case, of distinguishing between the announcing and decaying γ -ray.

SECTION II

DELAYED COINCIDENCE TECHNIQUES

AND ANALYSIS

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CHAPTER IV

The method of delayed coincidences between counters

In this chapter we are concerned with those analytical considerations which affect coincidence counting. In particular its object is to set out the analysis by which information may be obtained from delayed coincidence experiments. The limitations of these techniques are discussed and guidance is obtained in the design of practical apparatus.

A coincidence unit may be defined as a device whose function is to detect events occurring simultaneously in several counters. We shall be concerned mainly with the simplest case of two counters. However, due to practical limitations in the speed of response of the circuital elements used, and of the counters themselves, the instrument can only judge when an approximation to simultaneity has been achieved. Thus, if events occur within the same interval τ_0 they are judged by the instrument to be simultaneous. This time τ_0 is characteristic of the particular counters and circuits and is called the resolving time of the coincidence unit. Suppose we have two counters looking at a source emitting N pairs per second of coincident radiations A and B. Let us assume for simplicity that each counter is sensitive to only one of the radiations and that $\eta_A + \eta_B$ are the efficiencies of detection of A and B by their respective counters. Then

$$\text{Number of counts due to A in counter 1} = N \eta_A$$

$$\text{Number of counts due to B in counter 2} = N \eta_B$$

$$\begin{aligned}
 \text{No. of coincidences} &= \text{No. of counts due to A} \times \text{probability per unit time} \\
 \text{per unit time} &= \text{that a radiation B will} \\
 &= N \eta_A \times \eta_B \\
 &= N \eta_A \times \eta_B
 \end{aligned}$$

For two identical counters the number of coincidences is double this number.

If the counters are irradiated by separate sources such that there is no time relationship between the radiations emitted, it is still possible that a radiation may be detected in each counter simultaneously. If the counters detect N_1 , N_2 counts per second respectively, then a number of coincidences $2 N_1 N_2 \tau_0$ will be recorded. This may be seen as follows. Each time a radiation is detected in counter 1 the coincidence unit becomes sensitive for a time τ_0 . If a pulse occurs in 2 during this interval, a coincidence is recorded. Due to counts in 1 the coincidence unit is sensitive for a time $N_1 \tau_0$ per second, and this fraction of the counts in 2, i.e., $N_1 \tau_0 \times N_2$ will give rise to coincidences. Similarly, due to the N_2 counts in 2, $N_2 \tau_0 \times N_1$ coincidences are recorded. Therefore in all $N_1 \tau_0 \times N_2 + N_2 \tau_0 \times N_1$ i.e., $2 N_1 N_2 \tau_0$ coincidences are recorded. These are called random or statistical coincidences. Thus in actual experiment the number of coincidences observed will be the sum of the numbers of real and random coincidences. The number of random coincidences can be regarded as a sort of background which we must try to reduce. The ratio of real to random coincidences is of importance. Consider the simple case above where we have a source emitting pairs of radiations A and B with two counters each sensitive to only one.

$$\frac{\text{Number of real coincidences}}{\text{Number of random coincidences}} = \frac{N \eta_1 \eta_2}{2 N \eta_1 N \eta_2 \tau_0}$$

$$= \frac{1}{2 N \tau_0}$$

We wish this ratio to be as large as possible and hence require τ_0 to be small. We wish to have N small and hence to obtain a suitable coincidence counting rate we must have η_1, η_2 as large as possible. Thus two of the most important features of a coincidence apparatus of any type are detectors of rapid response and high efficiency.

Suppose we have a pair of simultaneous radiations detected by two counters connected to a coincidence unit of resolving time τ_0 . Let us suppose that delay x is inserted between one of the counters and the coincidence unit. In an ideal case, so long as x is less than τ_0 a coincidence will be recorded. If x is τ_0 no coincidence will be recorded. Thus if we have N_c coincidences per second and the number of random coincidences is negligible, the graph of N_c as a function of x will be a rectangle of width $2\tau_0$, x having both positive and negative values (corresponding to delaying the other counter). This function we call $P(x)$. In the case where random coincidences are not negligible, for $x < \tau_0$ the number of coincidences recorded will be the sum of the real coincidence and random coincidences, for $x > \tau_0$ it will equal the number of random coincidences only. Clearly a convenient means of measuring the background of random coincidences is to insert a delay greater than the resolving time and count the number of coincidences under these conditions.

We have considered above the simple case where the relative radiations A and B are emitted simultaneously. Suppose now that B is always emitted at a time t after A. The number of coincidences as a function of delay x inserted $F(x)$ will still give a rectangular distribution but this will be displaced by an amount t with respect to distribution in the case of simultaneously emitted radiations. If now the related particles A and B are separated by a variable time t where the probability of delay t is given by a function $f(t)$, $F(x)$ will no longer be rectangular and its shape will be governed by $f(t)$. Clearly for x large compared with τ_0 the distribution will have the form $f(x)$.

In practical cases (which will be discussed in Chapter V), a variety of instrumental delays of a random nature may occur, and any real systematic delays in the emission of one radiation with respect to another are superimposed on these. Clearly random delays will increase the resolving time just as amplifier noise limits the energy resolution of an ion chamber. The case in which these random delays have a Gaussian shape has been discussed by Binder (1950) and that in which we have a triangular distribution of delays by Newton (1950). Macintyre has shown that the analysis of Binder is in good agreement with experiment.

In these cases the resolving time as defined above loses much of its significance and is not readily observable from experiment. It is found convenient to redefine τ_0 in terms of measurable quantities by the equation

$$\text{Number of random coincidences} = 2 N_1 N_2 \tau_0$$

The modification in the shape of the graph of number of coincidences as a function of delay inserted when random delays are present is shown in

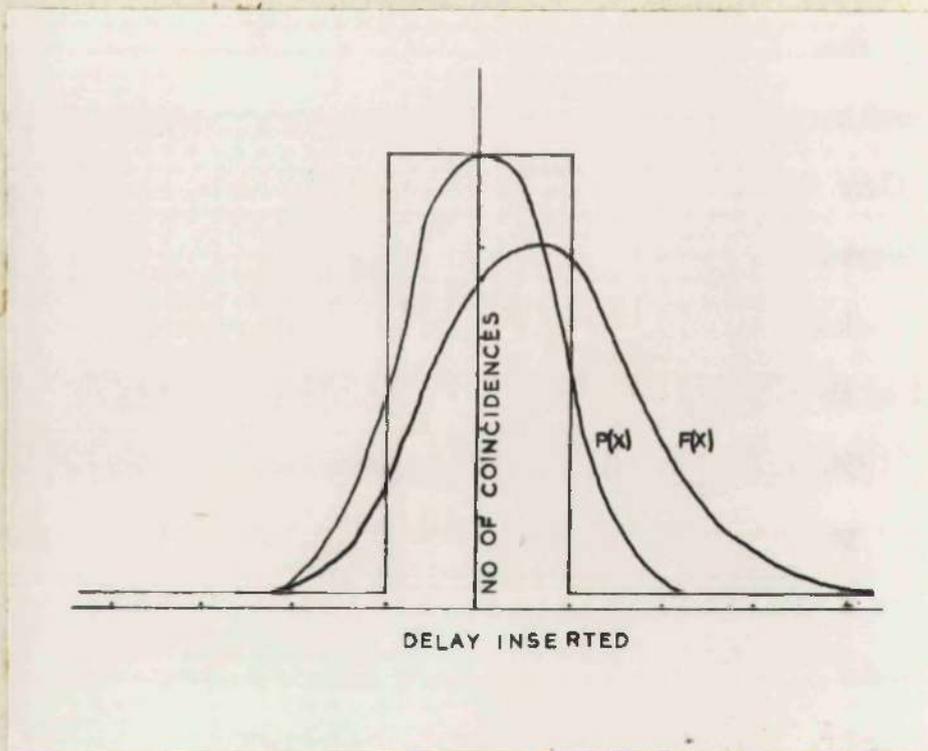


Fig. 1

This figure shows a typical set of resolution curves $P(x)$ and $F(x)$. The rectangle shows an ideal resolution curve for the same resolving time.

fig 1. In the case where there is no delay between the pairs of radiations being counted this curve, which is characteristic of the apparatus and radiations, is called the "prompt coincidence resolution curve $P(x)$ ". Clearly the mean width of this curve will be $2\tau_0$. When one of the pairs of radiations detected is systematically delayed with respect to the other, the corresponding curve is called the "delayed coincidence resolution curve $F(x)$ ". With suitable normalization (to unit area) we may re-interpret these curves. In the case of $P(x)$ the ordinate represents the probability that a coincidence will be recorded when two radiations are emitted simultaneously and detected, one of them being delayed by an amount x . A similar interpretation can be given to $F(x)$.

An important relationship exists between $P(x)$ and $F(x)$ when the only difference between the two cases is that in the latter one of the radiations is delayed with respect to the other. This relationship was pointed out by Newton (1950). It may be seen as follows.

Let the probability per unit time that a radiation will be delayed by an amount t with respect to the other member of the pair be $f(t)$. Suppose a pair of radiations is emitted with delay t between them and an artificial delay x is inserted.

$$\begin{array}{l} \text{Probability that this pair will} \\ \text{be recorded as coincident} \end{array} = P(x - t) dt \quad \dots\dots (1)$$

$$\begin{array}{l} \text{Probability of recording a coin-} \\ \text{cidence with delay } x \text{ inserted} \\ \text{and a delay } t \text{ between the} \\ \text{radiations} \end{array} = f(t) P(x - t) dt \quad \dots\dots (2)$$

$$\therefore \begin{array}{l} \text{Probability of recording a} \\ \text{coincidence with delay } x \\ \text{inserted} \end{array} = \int_{-\infty}^{\infty} f(t) P(x - t) dt \quad \dots\dots (3)$$

But this is $F(x)$

$$\therefore F(x) = \int_{-\infty}^{\infty} f(t) P(x-t) dt \quad \dots\dots (4)$$

If $f(t)$ has the form $\lambda e^{-\lambda t}$ which would arise if the first of the pair of radiations announced the formation of an exponentially decaying state whose decay was marked by the emission of the second radiation, this case becomes

$$F(x) = \lambda e^{-\lambda x} \int_{-\infty}^{\infty} e^{\lambda t} P(t) dt \quad \dots\dots (5)$$

If we now differentiate this equation we obtain

$$\frac{dF}{dx} = \lambda (P - F) \quad \dots\dots (6)$$

$$\frac{d}{dx} (\log. F) = -\lambda \left(1 - \frac{P}{F} \right) \quad \dots\dots (7)$$

We may deduce from these equations certain properties of the curves which are useful in the interpretation of experimental results. From equation (7), if x is such that $P \ll F$ then

$$F \propto e^{-\lambda x} \quad \dots\dots (8)$$

and thus we are able to measure λ from a logarithmic plot of the "tail" of $F(x)$ for large values of x in the region where $P(x)$ is small.

The accuracy with which we may measure this slope depends on the statistical accuracy of the points. This quantity depends largely on the ratio of the number of delayed counts compared to the number of random

coincidences. As the resolving time is increased the number of random coincidences rises. Further, the $P(x)$ curve is broadened and hence the delay x at which the condition $F(x) \gg P(x)$ is satisfied is increased and consequently $F(x) \propto e^{-x}$ decreased. Thus we see that increase in resolving time worsens the delay coincidences to random ratio, and therefore the accuracy with which decay constants can be measured. Looked at from another point of view, if we have a given resolving time, then as the decay period to be measured decreases, so the accuracy measurement decreases, and for a given resolving time this sets a lower limit to the times measurable by this method.

A further analysis, however, shows an alternative method in which these restrictions are avoided. Starting with equation (6) we have

$$\int_{-\infty}^{\infty} x \frac{dF}{dx} dx = \lambda \left[\int_{-\infty}^{\infty} x P(x) dx - \int_{-\infty}^{\infty} x F(x) dx \right]$$

$$\text{L.S.} = \int_{-\infty}^{\infty} x \frac{dF}{dx} dx = \left[x F(x) \right]_{-\infty}^{\infty} - \int_{-\infty}^{\infty} F(x) dx$$

Provided $F(x)$ diverges less rapidly than $\frac{1}{x}$

$$\text{mean life} = \tau = \frac{1}{\lambda} = - \int_{-\infty}^{\infty} F(x) dx = -1$$

$$= \int_{-\infty}^{\infty} x P(x) dx - \int_{-\infty}^{\infty} x P(x) dx \dots\dots (9)$$

With the above normalization $\int_{-\infty}^{\infty} x F(x) dx = \text{centroid of } F(x)$
 Similarly $\int_{-\infty}^{\infty} x P(x) dx = \text{centroid of } P(x)$

Hence $\tau = \text{displacement of the centroid of } F(x) \text{ with the centroid of } P(x).$

This was first shown by Bay (1950) using a more general analysis discussed below.

It is clear that if we can produce a corresponding pair of curves $P(x)$ and $F(x)$, the limits of measurement of τ depend only on the stability of the centroids of $P(x)$ and $F(x)$ and on the statistical accuracy with which the centroids can be determined. Since all the counts, and not just those in the tail, can be used in this method of analysis, the statistical accuracy is much greater than that in the first method described above, and in the author's experience in practice seldom forms the final limitation.

From the equations (5), (6) and (7) above, Bell (1952) makes several deductions which act as checks of the consistency of the results.

- (a) $F(x)$ and $P(x)$ rise from the same point on the left. We have found this a useful check against centroid shift due to instrumental drift.
- (b) If only a single lifetime is present in $F(x)$, then from equation (6) $P(x)$ and $F(x)$ intersect at the maximum of $F(x)$. This criterion is found to be rather difficult to apply with any accuracy when τ is short.
- (c) If only a single lifetime is present in $F(x)$, and if the curvature of $\log P(x)$ is nowhere positive, then the curvature of $F(x)$ is nowhere positive. The author has found the positive "kink" very valuable in the detection of complex lifetimes.

The above analysis is a particular case of a much more general theorem proposed by Bay in a brief letter where he gives a relationship between the n^{th} moment of the curve $F(x)$ and the moments of the curves $P(x)$ and

$f(t)$. Bay, in this letter, promises a demonstration of this in a later paper, but this has not appeared to the writer's knowledge. We have constructed a proof and the necessary assumptions are noteworthy. We start from equation (5) above.

We define in the usual way $M^n F(x) = \int_{-\infty}^{\infty} x^n F(x) dx$

By equation (6) $M^n F(x) = \int_{-\infty}^{\infty} dx \int_{-\infty}^{\infty} x^n P(x-t) f(t) dt$

By Taylor's theorem

$$P(x-t) = P(x) - P'(x)t + \frac{1}{2} t^2 P''(x) + \dots$$

$$+ \frac{(-1)^n t^n P^{(n)}(x) + \dots$$

$$M^n F(x) = \int_{-\infty}^{\infty} P(x)x^n dx \int_{-\infty}^{\infty} f(t)dt - \int_{-\infty}^{\infty} x^n P'(x) dx \int_{-\infty}^{\infty} t f(t) dt$$

$$+ \frac{1}{2} \int_{-\infty}^{\infty} x^n P''(x) dx \int_{-\infty}^{\infty} t^2 f(t) dt \dots$$

$$\dots \frac{(-1)^n}{n} \int_{-\infty}^{\infty} x^n P^{(n)}(x) dx \int_{-\infty}^{\infty} t^n f(t) dt + \dots$$

$$= M^0(f(t))I_0 - M^1(f(t))I_1 + \frac{1}{2} M^2(f(t))I_2$$

$$\dots \frac{(-1)^n}{n} M^n f(t) I_n + (-1)^{n+1} \frac{I_{n+1}}{n+1} M^{n+1} f(t)$$

Now $I_0 = \int_{-\infty}^{\infty} P(x)x^n dx = M^n P(x)$

$$I_r = \int_{-\infty}^{\infty} x^n P^{(r)}(x) dx = \left[\frac{x^n P^{(r-1)}(x)}{=0} \right]_{-\infty}^{\infty} - n \int_{-\infty}^{\infty} x^{n-1} P^{(r-1)} dx$$

The condition that the bracketed term be zero is that all the derivatives of $P(x)$ should converge at infinity more rapidly than $\frac{1}{x^n}$. By repeated

integration by parts we obtain

$$I_r = (-1)^{n-r} n(n-1) \dots (n-r+1) \int_{-\infty}^{\infty} x^{n-r} P(x) dx$$

$$= (-1)^{n-r} \frac{\lfloor n \rfloor}{\lfloor n-r \rfloor} M^{n-r} P(x) \quad r \leq n$$

If $r > n = \lfloor n \rfloor$ $\int_{-\infty}^{\infty} P^{(r-n)}(x) dx = \lfloor n \rfloor \left[\frac{P^{r-n+1}(x)}{=0} \right]_{-\infty}^{\infty}$

Thus we have with Bay

$$M^n F(x) = M^0 f(t) M^n P(x) + n M^1 f(t) M^{n-1} P(x)$$

$$\dots + \frac{\lfloor n \rfloor}{\lfloor r \rfloor \lfloor n-r \rfloor} M^{n-r} P(x) M^r f(t) \dots + M^0 P(x) M^n f(t)$$

..... (10)

Equation (10) shows that given $F(x)$ and $P(x)$ we may in principle deduce the moments of $f(t)$ and hence $f(t)$. In practice we have found it more useful to use a fit and try method.

Some simple cases deserve study and comment, and we will now consider these. In the case above where one detector is sensitive to the announcing radiation and the other to the decay radiation where the decay is exponential, we take first moments.

$$f(t) = \lambda e^{-\lambda t} \quad t \geq 0$$

$$M^1 F(x) = M^1 P(x) M^0 \lambda e^{-\lambda t} + M^0 P(x) M^1 \lambda e^{-\lambda t}$$

Now $M^n \lambda e^{-\lambda t} = \frac{\lfloor n \rfloor}{\lambda^n}$

$$\therefore M^1 F(x) = M^1 P(x) \cdot 1 + M^0 P(x) \cdot \frac{1}{\lambda}$$

$$\therefore \tau = \frac{1}{\lambda} = M^1 F(x) - M^1 P(x) \quad \text{as we have shown above.}$$

..... (11)

There may arise cases in which it is not possible to satisfy the condition that only one of the counters is sensitive to the decaying radiation and the above analysis does not apply. In this case we have

$$f(t) = \lambda e^{-\lambda(t)}$$

Integration by parts shows that

$$\begin{aligned} M^n \lambda e^{-\lambda(t)} &= \frac{M^n}{\lambda^n} && n \text{ even or zero} \\ &= 0 && n \text{ odd} \end{aligned}$$

..... (12)

Applying Bay's expression for $M^1 F(x)$ we find that the centroid $F(x)$ coincides with that of $P(x)$. However for $M^2 F(x)$ we obtain

$$M^2 F(x) = M^2 P(x) \cdot x + M^0 P(x) \cdot \frac{2}{\lambda^2}$$

$$2\tau^2 = \frac{2}{\lambda^2} = M^2 F(x) - M^{(2)} P(x)$$

..... (13)

Hence from a measurement of the second moments of $P(x)$ and $F(x)$ τ may be determined.

It is clear that if all the pairs of radiations are separated by a

fixed delay the curve $F(x)$ will be similar in all respects to $P(x)$ but each point (and hence the centroid) displaced by the amount of the delay.

We have found the Dirac δ function useful in dealing with more complex cases, (if $F(t) = \delta(t-a)$ this represents a fixed delay of a), using

the property that
$$\int_{-\infty}^{\infty} X(x) \delta(x-a) dx = X(a)$$

From first principles we have

$$F(x) = \int_{-\infty}^{\infty} f(t) \cdot P(x-t) dt$$

If $f(t) = \delta(x-a)$

$$F(x) = \int_{-\infty}^{\infty} \delta(t-a) P(x-t) dt$$

$$= P(x-a) \quad \text{— the result deduced above}$$

Since $\int_{-\infty}^{\infty} \delta(x) dx = 1$ and $\int_{-\infty}^{\infty} \delta(x) f(x) dx = f(0)$

Clearly $M^n \delta(x) = 1 \quad n = 0$
 $= 0 \quad n > 0$

We use these in a final example of a more complex case. Suppose the announcing radiation signifies the formation of one of several possible separate states with decay constants $\lambda_1 \dots \lambda_r$ of which decay promptly. The former have individual $f(t)$ of $\lambda_1 e^{-\lambda_1 t} \dots$ and the latter $\delta(t)$. $f(t)$ for the whole system will have the form

$$a_1 \lambda_1 e^{-\lambda_1 t} + a_2 \lambda_2 e^{-\lambda_2 t} \dots a_r \lambda_r e^{-\lambda_r t} \dots + b \delta(t)$$

where $\sum_r a_r + b = 1$

For a solution to this problem we will clearly require $2r$ equations and these can be obtained from the first $2r$ moments of $F(x)$ and $P(x)$.

It must, however, be remembered that each moment has an associated probable error which increases as the order of the moment increases. It may be shown that the solutions of these 2 ζ equations for the moments of $f(t)$ (which determine the latter function) consist of a series of terms of the form

$$M^n F(x) - M^n P(x)$$

As the number of unknowns increases these terms become less and less accurate and the method breaks down.

Conclusions

The power and limitation of delayed coincidence technique depend greatly on our knowledge by other means of the complexity or otherwise of the delays which will be encountered. If it is certain that only one period is present the analytical method above may be applied and the limit to the accuracy of the result is likely to be set more by the stability than the resolving time of the apparatus. However, as the known complexity increases so the accuracy possible decreases, until the method becomes no longer profitable.

When, on the other hand, there is no fore-knowledge of the "delay scheme", the analysis above is of little value. It then becomes necessary to examine the "tail" of the resolution curve. We have discussed the limitations of this method above. While it is difficult to measure a period shorter than the resolving time in this way, care is needed lest a long period associated with only a fraction of the decays be overlooked. This would be observed more readily with a longer resolving time.

Combining these considerations with those set out earlier, the following requirements for a high resolution delayed coincidence apparatus may be deduced.

- (1) Detectors of high efficiency to reduce counting time and hence "improve" stability. The ratio of real to random coincidences would also be increased.
- (2) Minimum random delays to give shortest possible resolving time.
- (3) High stability in order to make fullest use of analytical techniques.

These are therefore the considerations to be kept in mind when designing a delayed coincidence unit, though a certain degree of compromise is necessary between the requirements of stability and resolving time.

CHAPTER V

Delayed Coincidence Techniques

Historical Survey

The first application of electrical coincidence counting to the study of nuclear phenomena was made by Bothe and von Baeyer (1925), though as early as 1910 Geiger and Marsden had viewed with the eye, scintillations appearing to occur simultaneously on two zinc sulphide screens. Dunworth (1939), however, was the first to use such methods in the measurement of very short time intervals when he measured the 3×10^{-7} s. lifetime of ThC' . His apparatus consisted of two Geiger counters connected to a coincidence unit of variable resolving time.

Dunworth pointed out that a lower limit to the useful resolving time of his apparatus was set at about 5×10^{-7} s. by the random delays inherent in the operation of Geiger counters. Subsequent investigations have confirmed that this is so (e.g. Den Hartog et al, 1947, Sherwin, 1948). This is a defect of gas counters and represents the drift time of electrons towards the region of multiplication round the collecting wire. In addition to this limitation, their low efficiency for the detection of γ -rays makes them unsuitable for coincidence experiments of the type proposed by the writer.

The great advance in delayed coincidence technique came with the introduction of scintillation counters (McGowan et al. 1949) as detectors with such apparatus. Scintillation counters may readily have efficiencies of the order of 10% and more for γ -rays of 1 Mev and

detect β -rays with similar efficiency to a Geiger counter. With regard also to speed of response, the scintillation counter is from ten to a hundred times faster than any "gas counter". This aspect of the scintillation counter will be examined in more detail below.

The great improvement in the speed of response of the counter was an incentive to develop electronic circuits of shorter resolving time. The early scintillation counter gave output pulses of the order of 10^{-2} to 10^{-3} volts due to the low gain of the available photo-multipliers and imperfect phosphors. Considerable amplification was required before application to coincidence circuits. This increased the rise time of pulses to about 1.5×10^{-7} s., e.g. McGowan et al. (1947). To overcome this limitation, amplifiers of greater bandwidth were designed, culminating in the distributed amplifier of Gintzon et al. (1948) which had a bandwidth of 200 megacycles. Another approach was that of Bell and Petch (1949) who, with selected photo-multipliers, applied inter-stage voltages considerably in excess of the recommended values, and in this way obtained pulses of several volts from a single electron leaving the photo-cathode of the photo-multiplier. This method produces little or no increase in rise time.

Since the scintillation counter produces pulses proportional to the energy expended in the phosphor, some form of amplitude discrimination is usually required. With pulses of finite rise time, a large pulse triggers a discriminator earlier than a smaller one, thus introducing spurious delays. Special circuits to minimise these effects were introduced by P. R. Bell et al. (1947). Such difficulties were removed by Bell and Petch (1949), who used amplifiers and discriminators

in parallel with the fast coincidence channel to measure the pulse height from each of two counters. A triple coincidence unit fired only when both amplitude and coincidence channels were satisfied. The "side-channels" and triple coincidence unit could be of conventional "slow" design as they had no effect on the resolving time of the circuit proper. This arrangement has since been universally adopted.

Coincidence Circuits

In the work with Geiger counters, and in the early experiments with scintillation counters, the coincidence circuit commonly used was that due to Rossi, of DeBenedetti and McGowan (Phys. Rev. 70, 569, 1946). This consists of two pentodes with a large common anode load so that each acts as a clamp on the anode voltage when the other is cut off by a single pulse. Coincident pulses cut off both giving a large output. The resolving time of this circuit is limited to about 5×10^{-8} s. by the rise time of the anode pulse. Pulses of about 4-v are required to operate such a circuit. Similar to the Rossi circuit was that of Baldinger et al. (1949) which was two triode valves with a common cathode load. The pulse at the cathode was measured by a low capacity diode discriminator circuit. With the low output impedance for non-coincident pulses due to the cathode follower action and the reduced stray capacity, a resolving time of 5×10^{-9} s. was achieved.

Cathode ray tubes were used as delayed coincidence units by some workers. Krashaar et al. (1950) measured the mean life of the π^+

meson. The pulses representing the stopping of the \bar{V}^+ meson and the emission of the μ^+ meson were displayed on a time base. This was photographed and the separation in time of the pulses measured from the photographs. The resolving time was about 3×10^{-8} s. Used in this way, the cathode ray tube is a multi-channel time-sorter (by analogy with a multi-channel pulse amplitude analyser) and greatly reduces the time required for experiments. This is especially valuable when events of infrequent occurrence are being studied. The main disadvantage of the cathode ray tube lay in its poor deflection sensitivity such that pulses of tens of volts were required to produce a measurable deflection. This effect was accentuated when the duration of the applied pulse was less than the transit time of the electron beam through the deflecting plates. The recent introduction of traveling wave amplifiers and deflection systems (Pierce 1949) have largely removed these objections, but there seems to the writer to be little advantage in the use of such complex devices when comparable performance can be attained with the simpler circuits to be described below.

Circuits specially designed to take advantage of the time-resolving properties of scintillation counters have the common property of requiring only small pulses that can be supplied direct from a photo-multiplier avoiding all amplification. All make use of germanium crystal rectifiers which have a very low forward resistance even for pulses of 0.1 v. and small capacitive coupling from anode to cathode. These circuits may be divided into two main types - balanced circuits and circuits employing pulse equalization. In the former type (e.g. Baldinger

et al., 1947, Bay, 1951) the circuits are so arranged that a single pulse cancels itself, while due to the non-linear elements, a coincidence gives an output pulse. With the circuit of Bay, resolving times less than 10^{-9} s. may be achieved. Further, pulses of a large range of amplitudes may be applied to the circuits. The disadvantage of these circuits lies in the difficulty of matching crystal diodes over a wide frequency band, and also matching their temperature characteristics.

The two best known circuits of the second type are those of Bell et al. (1952) and DeBenedetti et al. (1952). The feature of both is that pulses from a photo-multiplier are applied to a pentode causing the current in the latter to be cut off. A standard pulse is thus produced. The standardised pulses from the two counters are added and amplified in a "slow" amplifier. A crystal diode at the input of this amplifier prevents the addition of pulses within its resolving time. The advantage of these circuits is in their simplicity and reliability, and as shall be shown below, the limiting action may be used to improve the resolving time. The limit to the resolving time is set by the rise time of the pulse at the anode of the limiting valves and is probably about 10^{-9} s.

Most of the above techniques and circuits were known in principle to the author before he began his experimental work, but it was not until some time later that any details of the important circuits such as those of Bell and DeBenedetti were published.

On the grounds of simplicity and stability, it was decided to build a circuit similar in principle to that of Bell and to use photo-multipliers as detectors.

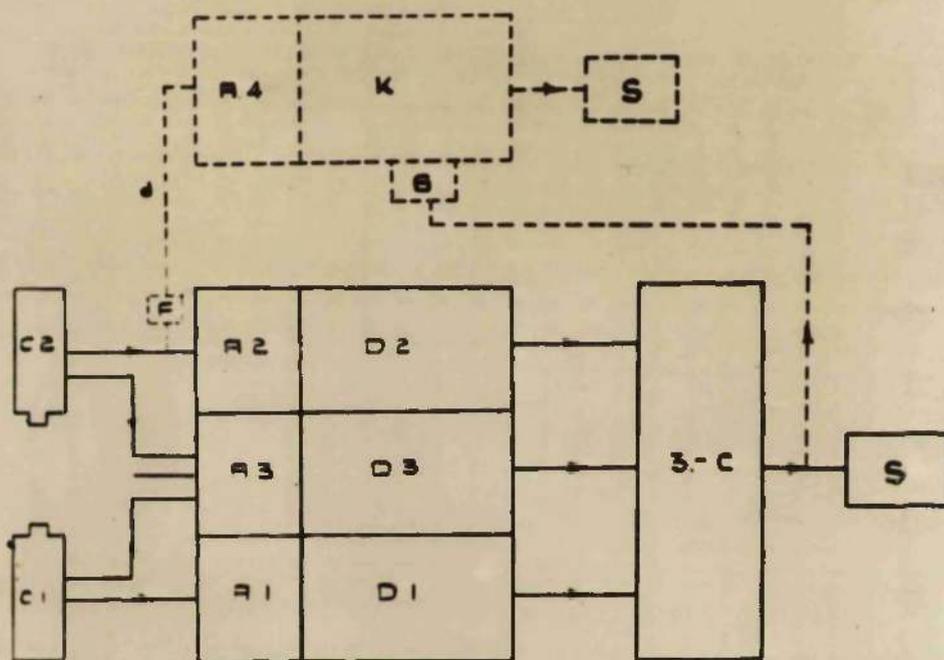


Fig. 2

Block diagram of apparatus. Full line shows basic apparatus. Dashed line shows later additions.

C 1, 2 - counters; A 1, 2, 3, 4 - amplifiers;
 D 1, 2, 3 - discriminators; K - kicksorter;
 3-C - triple coincidence unit; F - cathode
 follower;
 G - gate; d - 1.25 μ s. delay line.

CHAPTER VI

The development of a delayed coincidence apparatus of 2×10^{-9} s. resolving time apparatus

Introduction

In the preceding chapter, the development of delayed coincidence technique has been traced and some of the circuits used in this field discussed. From such a survey it appeared that for our purposes the circuit best suited was that of Bell, as it gave adequate performance with the greatest simplicity and reliability. This decision was further strengthened by the fact that an apparatus on similar principles had been built by Mr. G. M. Lewis of this laboratory. It had a resolving time of about 10^{-8} s. At that time, Bell had published only the broad principles of his circuits and so the writer's apparatus was developed rather from that of Lewis.

The Apparatus

The general principles of the apparatus were described in the previous chapter. A block diagram is given in fig. 2 (full line).

The two side channels which provided amplitude discrimination were not altered materially from those of Lewis. They consisted of a head amplifier - a cathode current fed back pentode, to which the pulse was applied via an attenuator. A_2 was a two-valve RC coupled amplifier without feed back. Inductive compensation was employed to increase

the bandwidth. D_2 was an AERE type 1028 discriminator, the wiring of which was modified to reduce the input capacity.

The triple coincidence unit was also after Lewis. It was of conventional Rossi type and had a resolving time (in conjunction with the discriminators) of $0.6 \mu\text{s}$.

The counters and fast coincidence channel were considerably modified. In the original design the counters consisted of E.M.I. type 5311 photo-multipliers with anthracene as phosphor. Pulses from the counters were applied directly to the limiting pentode. To avoid feedback, the cathode of the latter was decoupled. Its anode load was matched to the 95 Ohm Uniradio 31 coaxial delay cable along which the pulses are transmitted to be mixed in the shorted line which determines the duration of the pulse. The cables from C_1 and C_2 meet the shorted line at the "mixing point". Preceding the "diode clamp", a single valve gives further amplification. The "diode clamp" prevents further superposition of pulses within the resolving time of the subsequent "slow" electronics and also lengthens the pulses to be suitable for the latter. Beyond the diode clamp there was a further stage having a gain of 25 to bring the pulses to an amplitude suited to discrimination by the standard discriminator D_3 . Detailed circuits of this apparatus will not be given here. In Appendix II the circuits of the author's final apparatus are given.

Modifications

Conventional techniques of electronic development using pulse generator and oscilloscope could not be employed in this work because

no generator or oscilloscope of adequate speed was available. In the early stages deduction had to be made from the lengthened waveform at the output of A_3 and later from delayed coincidence resolution curves. Modifications were usually suggested from consideration of the general principles of the apparatus, so that it is proposed, before going further, to give a more detailed analysis of certain of its aspects and the modifications they suggested.

Pulse duration

The resolving time is primarily determined by the duration of the pulses at the diode clamp. It may be shown that this duration D may be written

$$D = 2L + T_A$$

L = time for a pulse to traverse the shorted line

T_A = rise time of the pulse

Further, T_A may be divided into two components, viz., the inherent rise time of the electronics, T_S , when a step pulse is applied to the limiting valve, and the time to cut off the limiting valve. If the rise time of the photo-multiplier pulse is T_r the cut-off time will be $\frac{T_r}{\eta}$. η is a function of the ratio

$$\frac{\text{amplitude of pulse}}{\text{amplitude required for cut-off}}$$

(If η can be made large, cut-off time can be made negligible. This was not the original purpose for which the limiting valve was introduced by Lewis, Bell and others, and it was the realisation of this property

that led the writer to hope to obtain short resolving times despite predictions as to the unsuitability, due to slow response, of the only available type of photo-multiplier.) We wish D to be a minimum.

If $2L$ is made less than T_A then it may be shown that pulse amplitude is rapidly lost with little improvement in D . This sets a lower limit to $2L$. Clearly to obtain an improvement it is the rise times which must be reduced.

T_r

The main factors affecting T_r , the rise time of pulses from the photomultiplier, are:-

1. Decay time constant of the phosphor.
2. Spread in transit time in the photomultiplier.
3. Time constant of the output circuit.

The statistical effects (random delays in emission of the first photo-electron) due to the excitation of a relatively small number of phosphor molecules (Post and Schiff, 1950) are not significant when energies such as are proposed are expended in phosphors, viz., more than 500-kev and light collection is good.

An analytical discussion of the pulses from a photomultiplier has recently (Lewis and Wells, 1954) been given, but for our purpose a more qualitative treatment seems adequate. The photomultiplier is essentially a current generator and the output time constant determines whether or not the pulse of current is integrated. If the time constant is short compared with the spread in transit time and phosphor decay time, the rise time depends almost entirely on the former. From the work of

Phosphor	Relative Energy Yield	Decay Time $\times 10^{-9}$ s.
Anthracene	100	23 ± 5
Napthalene	24	60
Stilbene	67	6 ± 1
Terphenyl in Toluene	55	2.2 ± 0.3

Allen and Engelder (1951) it would seem that rise times of about 10^{-8} s. could be expected with conventional accelerating voltages. With an integrating circuit, the rise time would be increased by of the order of the decay time of the phosphor. Decay times for a number of phosphors have been measured and are given in Table opposite. For stillbene, and solutions of terphenyl, the rise time would appear to be little greater than for the case of short time constant. The pulse amplitude would be greater. The advantage would seem to lie with the integrating circuit in these circumstances.

Note: The transit time spread mentioned above refers to the E.M.I. type photo-multiplier. For the electrostatically focussed type, e.g., R.C.A. type IP21, transit time spread is of the order of 10^{-9} s. (Morton 1949). The above conclusions would not then hold. However, to obtain the required gain with IP21 type photo-multipliers, it is necessary to greatly exceed the manufacturers' recommended voltages, and so the risk of failure is high. Since these tubes had to be imported, involving delays and dollar expenditure, it was felt that no risks could be taken with them. It was therefore decided to use the E.M.I. type tubes and attempt to overcome their disadvantages.

" "

η

Since the pulse width can be reduced by a large η we clearly wish to have as large output pulses as possible. The pulse amplitude is proportional to -

1. The number of photo-electrons emitted by the photo-cathode.

2. The gain of the photo-multiplier.
3. The reciprocal of the output capacity.

The first quantity depends on the efficiency of the phosphor for the conversion of the energy expended by the ionizing particle into light, and also on the efficiency of the photo-cathode for the conversion of this light into photo-electrons (see Table). Another factor involved is the fraction of the light emitted which actually reaches the photo-cathode. This fraction depends on the geometry and optical properties of the arrangement.

The gain of the photo-multiplier depends strongly on the applied voltage varying approximately as the seventh power. It is limited by the maximum voltage that can be applied to a stage. This limit may be set by onset of corona discharge from sharp points or regenerative positive ion effects and varies with individual tubes (Post, 1950). To obtain more gain the number of stages may be increased, though transit time spread changes with the square root of the number of stages (Morton, 1949).

From the above considerations, it was decided to use either still-bene or solutions of terphenyl in toluene as scintillator on the grounds of their short decay constant. Their scintillation efficiency is also not unfavourable. In the case of the solution, it was to be contained in cells of quartz with thin, plane ends cemented with Araldite to give good light transmission to the photo-cathode. A reflector of MgO was to be used.

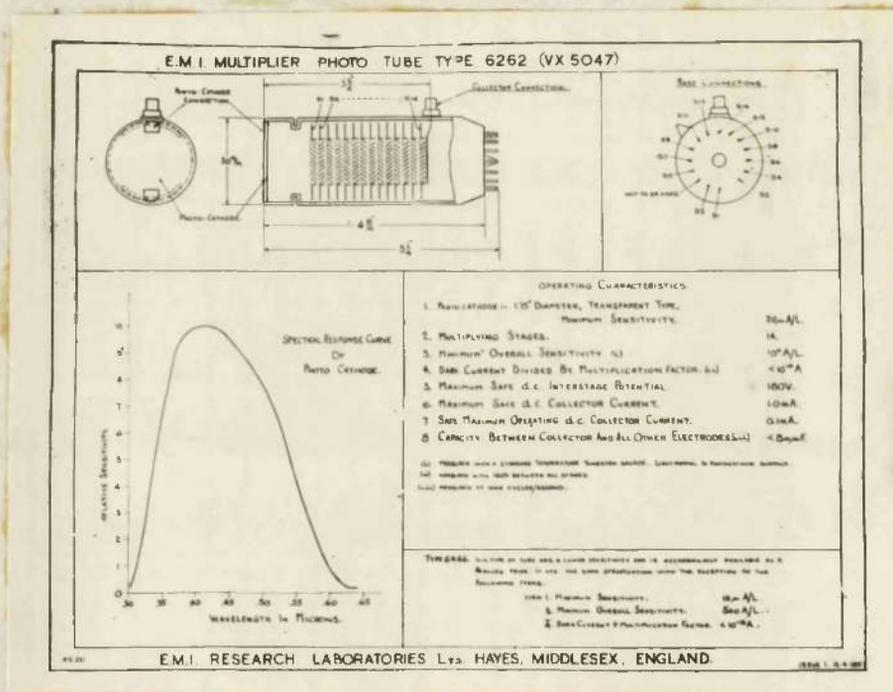


Fig. 3

E.M.I. Phototube Type 6262.

The 11-stage photo-multiplier was to be replaced by the new 14-stage E.M.I. type 6262 (see fig. 3). As well as a gain of 10^9 (160-v per stage) these tubes have a photo-cathode 2" in diameter so that light collection from large cells is possible.

To reduce output capacity a new mounting was designed in which the limiting valve was mounted on the photo-multiplier light tight box with its grid pin close to the output electrode. In these ways the writer attempted to reduce the quantity $\frac{T_r}{C}$

T_A

T_A is the rise time of a pulse which would reach the diode clamp if a step-pulse were applied at the grid of the limiting valve. It appeared that only minor alterations were possible in the limiting valve circuit, but it seemed desirable to eliminate the amplifying stage preceding the diode clamp. Some remarks about the latter seem pertinent. The diode clamp has the function of lengthening the pulse so that it may be amplified by conventional techniques, and of preventing superposition within the lengthened time. It is very important that the charging time through the diode should be less than the proposed pulse width. To facilitate this, the diode should have a low forward impedance for the amplitude of signal applied. Using electronic diodes, it appears that a signal approaching 1 volt is needed to obtain a sufficiently low forward impedance. Since the signal from the limiting valve is only 0.25 volts, the additional stage of amplification is required. To eliminate this, germanium crystals type GEX 44, some of which have a forward impedance

of about 200 Ohms for 0.25-v signals, were introduced. This reduced the charging time to about 2×10^{-9} s. Finally, two such crystals were used in parallel. These crystals were inserted between the mixing point and the first valve of A_3 . At the same time, the output circuit of the limiting valve was cleaned up by the use of miniature components, improved radio-frequency plugs, and better earthing.

Resolving time

The resolving time for two pulses may be said to be

$$\frac{1}{2} \left[\begin{array}{l} \text{duration of pulse A + duration of pulse B} \\ - \text{minimum overlap required} \end{array} \right]$$

Since the diode clamp is an integrating circuit of time constant about 10^{-9} s., the transmitted amplitude of the overlapping part of the superimposed pulses will depend on its width. Hence the bias of discriminator D_3 will affect the resolving time for a pair of pulses. Since the expression above (page 53) for the duration of a pulse contains the pulse amplitude (through α) the resolving time will be a function of the pulse amplitude. If the coincident pulses have a distribution of amplitudes, the experimentally measurable quantity will be the mean resolving time. This quantity is clearly dependent on the pulse spectrum as well as the circuitry.

Suppose the pulses from the two photomultipliers are identical. If delay is now inserted between either counter and the mixing point, the number of coincidences will be constant as a function of delay until the overlap is so small that D_3 does not trigger. The resulting

"coincidence resolution curve" will be a rectangle symmetrical about zero delay. Now let the pulses on one side all be increased in amplitude. Two effects will follow. Firstly, the duration will be decreased at the mixing point of pulses from the counter giving the increased amplitude. Hence the resolving time and width of the rectangle will be reduced. Secondly, the centroid of the rectangle will be displaced from zero. The significant point on the pulse for such circuits as these is the centroid. In the shorter pulse this will be nearer the true point in time of the nuclear event which is at a fixed time from the beginning of the rise of the pulse. Hence the pulse representing a low energy event will appear delayed with respect to one representing a higher energy event.

When the coincident pulses from both counters have a distribution of amplitudes, the resolution curve will be the superposition of an infinite set of rectangles of varying widths displaced relative to one another, giving rise to a smooth curve. The exact shape of the curve depends on the pulse spectrum. The position of its centroid depends on the mean pulse height from each counter.

The three bias levels of D_1 , D_2 , D_3 each affect both the resolving time and position of the centroid of the resolution curve. D_3 affects mainly the resolving time, while D_1 , D_2 change the mean accepted pulse height and produce displacements of the centroid of the resolution curve.

Initial tests

The main variables of the apparatus were the multiplier voltage, the length of the shorted line, and the settings of the three discrimina-

tors. The maximum voltage that may be applied to the tube is 2600. From the point of view of speed of operation it is desirable to operate as near this value as possible. The application of such high voltage raised two main difficulties.

Firstly, above about 1700-v the number of noise pulses was observed to increase considerably. This was taken to signify the onset of processes in addition to thermal emission from the photo-cathode, e.g., cold emission from sharp points. Secondly, at voltages greater than this, signal pulses from 500-kev γ -rays began to limit and thus signal to noise ratio was reduced. Pile up of the increased number of noise pulses in the side channels contributed to this effect. Pile up was minimized by reducing the output time constant of the photo-multiplier. A type C.V.1192 diode was inserted between the first and second valves to lengthen the pulses sufficiently to operate the discriminator while preventing pile up of successive small signals. Extra decoupling on the last few stages of the photo-multiplier was also found to be helpful in reducing the limiting effect. The effects of space-charge, however, could not be eliminated.

It was found very convenient to have the cathode of the phototube at earth potential and tests also showed a reduction in noise using this method of operating. As the tubes are surrounded by an earthed can, the latter has a defocussing effect in the important early stages when the photo-cathode is at a negative potential.

As the length of the shorted line was reduced below 5×10^{-9} s., the pulse height at the output of A_3 was found to decrease considerably.

This was thought to be due to the excessive rise time of the pulse. A_3 was redesigned using four valves to have much greater gain. The resolving time measured, however, decreased steadily, which would not have happened had the pulses risen as slowly as the fall in amplitude indicated. At resolving times of the order of 10^{-9} s. an amplifier of mid-band gain of about 10^4 was needed to give signals in the region from 10-20-v. The estimated bandwidth was about 5 M/cs.

In a paper giving detailed designs of a circuit very similar to this, Bell, Graham and Petch (1952) use an amplifier of gain 2×10^4 and bandwidth 2 M/cs without comment. An explanation of this effect has been suggested by F. H. Wells (private correspondence) who has examined the pulse at the grid of the first valve with a fast triggered oscilloscope. He finds that the crystal diode does not lengthen the pulse as much as D.C. measurements of its backward resistance lead one to expect.

The performance of the circuit

There is good reason to expect that the delay in emission of the second γ -ray of Co^{60} is very small. Coincidences between these γ -rays were used to measure the resolving time of the circuit. Solutions of terphenyl in toluene were used as scintillators, and 2000 volts applied to the multiplier. The shorted line was of length equivalent to 2×10^{-9} s., D_3 was set so as to just exclude single pulses, and D_1 and D_2 set at their minimum value. A resolving time of 4×10^{-9} s. was obtained. If the setting of D_3 was increased above the minimum value, the resolving time was reduced towards 2×10^{-9} s. However, the number

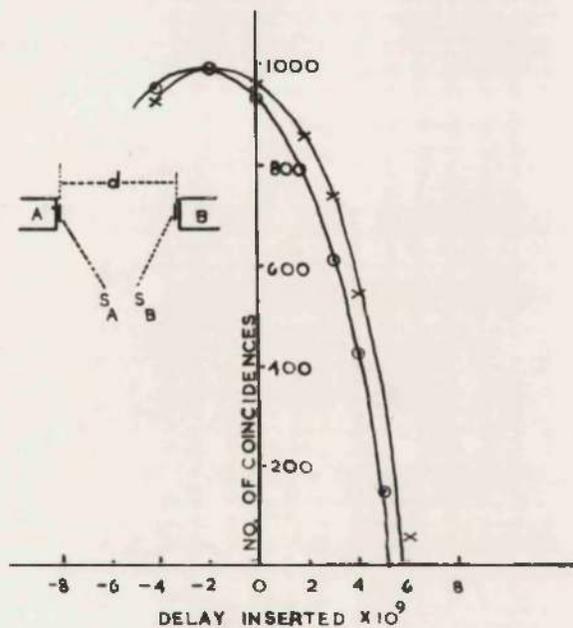


Fig. 4

Time of Flight of γ -rays:- Resolution curves through crosses and circles are those taken with source in positions A (S_A) and B (S_B) respectively. d was 7 cm.

In the inset, A and B are the phosphors; S_A , S_B the two positions of the source.

of coincidences also decreased. With a shorted line 1×10^{-9} s. long and minimum settings, a resolving time of 2.5×10^{-9} s. was achieved, and by increasing D_3 bias, this could also be reduced towards 1.5×10^{-9} s.

The effect of increasing the bias of D_1 and D_2 by similar amounts decreased the resolving time, but not to the same extent as corresponding increases in D_3 . If only one of D_1 or D_2 was increased, the resolving time decreased only slightly, but the resolution curve was displaced by about 1.5×10^{-9} s. for a ratio of about 2 in mean pulse heights in channels 1 and 2.

The above tests showed that a delayed coincidence apparatus with a resolving time of 2×10^{-9} s., capable of giving repeatable resolution curves, was available. They underlined most strongly the not unexpected dependence of the position of the centroid of the resolution curve on the relative energy bands accepted by discriminators D_1 and D_2 .

As a final test of the apparatus, an experiment to measure the time of flight of the γ -rays of Co^{60} was carried out. (1) The experimental arrangement is shown in Fig. 4. The source of about 20 mC is first placed in position A. The pulses in multiplier B, corresponding to those in A, are delayed by an amount $\frac{d}{v}$. Similarly, in position B they are delayed by $\frac{-d}{v}$. We therefore expect the centroid of the resolution curve to be displaced in the second case by an amount $\frac{2d}{v}$ with respect to the first. This was done for several values of d , the smallest being

(1) A somewhat similar technique has been applied by Cleland and Jastram (1951) in the measurement of the velocity of annihilation γ -rays.

7 cm. This corresponds to a separation of the curves of 5×10^{-10} s. (see Fig. 4). The curves are clearly resolved point for point and the estimated error in the determination of the centroid position was $\pm 1 \times 10^{-10}$ s.

These results showed that if suitable comparison curves could be obtained, delays of a few times 10^{-10} s. could be detected and measured in favourable cases.

SECTION III

THE LIFETIMES OF POSITRONS
IN SOLIDS

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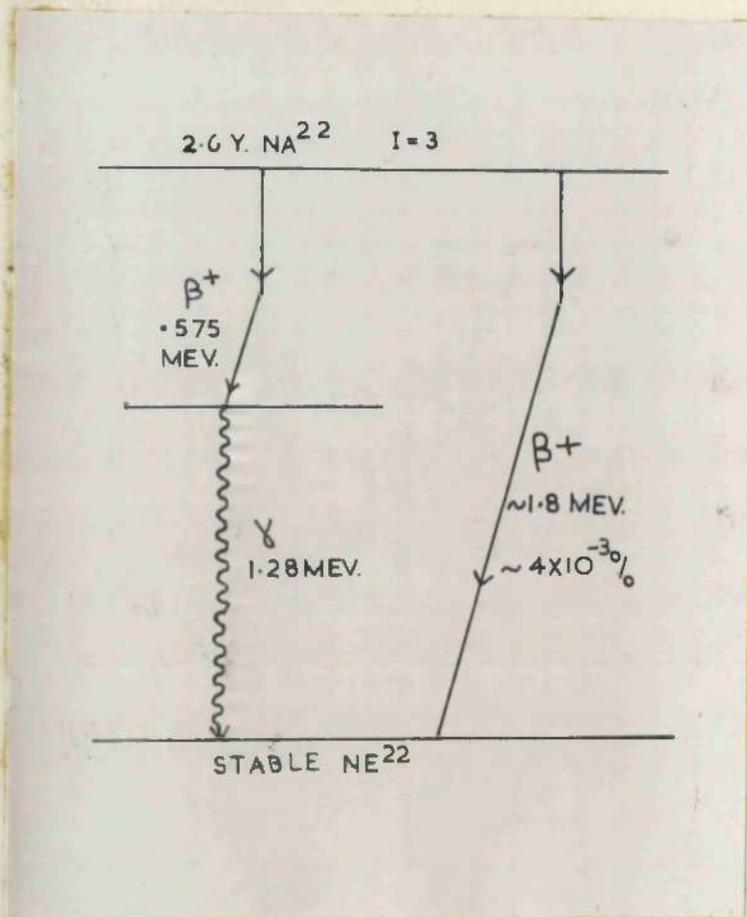


Fig. 5

Decay scheme of Na^{22} after

"NUCLEAR DATA"

CHAPTER VII

Some early experiments on the lifetimes of positrons in solids

Introduction

It had been shown by DeBenedetti (1951) that the difference between the lifetimes of positrons in lead and potassium was probably less than 10^{-9} s. His experiments consisted of allowing the positrons from a Cu^{64} source to pass through a thin crystal into the sample. A second crystal detected the annihilation radiation. The pulse initiated by the passing of the positron through the first crystal marked the time of entering the sample, while the pulse due to the annihilation radiation indicated the instant of its annihilation. This method has the disadvantage that, due to the continuous nature of the positron spectrum, and the poor geometry used, much annihilation takes place in the crystal and other foreign bodies. If, to avoid this, the crystal is made thin, the pulse height is reduced and the time resolution of the apparatus adversely affected.

An alternative method was suggested by the work of Deutsch (1951) in gases. He used Na^{22} as his source of positrons (see decay scheme fig. 5). If there is no appreciable delay in the emission of the 1.28 Mev γ -ray following the positron, this may be used to mark the instant of emission of the particle. This technique allows the source to be completely surrounded by the material in which one is attempting to measure the positron lifetime. Annihilation in the source itself can be kept small, as, with care, it can be obtained relatively free from carrier.

The Positron Source

Na^{22} decays by the emission of a positron to the 1.28 Mev excited level of Ne^{22} . The latter is an even-even nucleus and hence its ground state has spin 0 and even parity. From the shell model, the first excited state is expected to have spin 2 and even parity. The 1.28 Mev γ -ray would therefore be electric quadrupole and have a lifetime of the order of 10^{-12} s. This is negligibly small for our purposes here.

Na^{22} is prepared by the reaction $\text{Mg}^{24} (d, \alpha) \text{Na}^{22}$. The cross-section for this reaction rises rapidly with the bombarding energy of the deuterons. A magnesium target was bombarded in the Birmingham cyclotron and the Na^{22} chemically extracted at Harwell. The writer is greatly indebted to both establishments.

The source, of strength about 10 μC , was received in the form of a solution of NaOH . Tests showed that with chemically very weak solutions, aluminium was protected from further chemical attack by the sodium aluminate layer initially formed. The considerable delay in obtaining the source precluded risks and the aluminium upon which the active material was deposited was .005" thick. Two such pieces were used and the source material enclosed between them. It is estimated that about 20% of the positrons annihilate in this source and backing.

The Apparatus and Experiments

The apparatus used was that described in Chapter VI. The scintillators were bottles $2\frac{1}{2}$ " long and $1\frac{3}{4}$ " in diameter containing a solution of terphenyl in toluene.

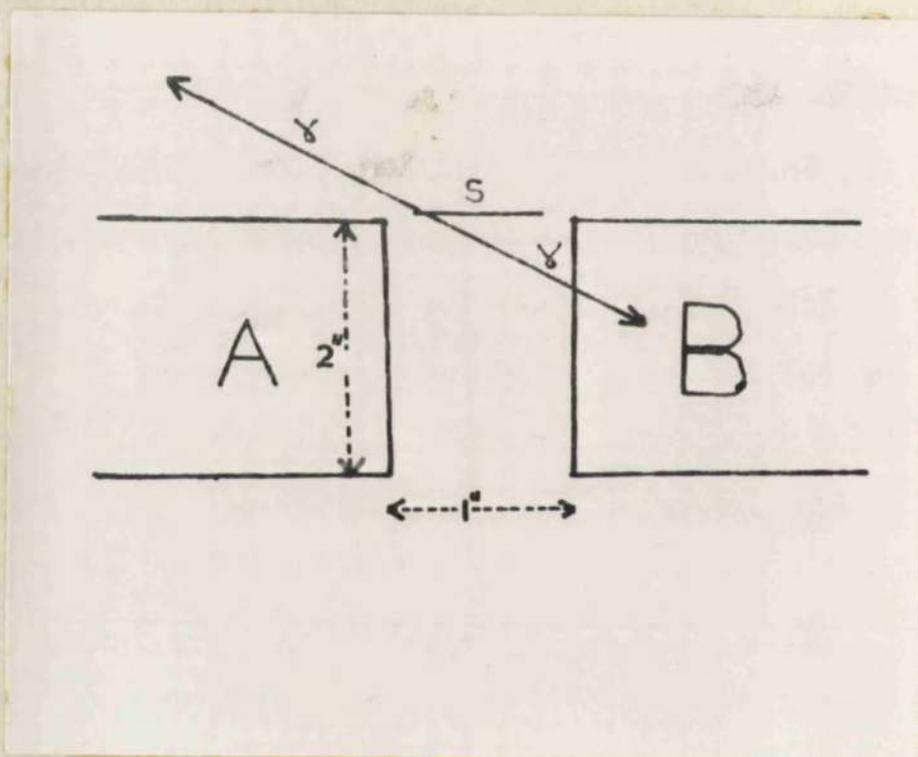


Fig. 6

Arrangements of source and counters for comparison experiments. A and B are the phosphors of counters A and B; S is the source, deposited on aluminium foil, surrounded by the materials being compared. Arrows show how only one of any pair of coincident annihilation radiations may be detected.

The geometrical arrangement of the source and counters is shown in fig. 6. The object of this was to avoid coincidences between the oppositely directed annihilation quanta. This was done for the following reason. With the source on the axis of the two counters, coincidences between annihilation quanta (which would be "prompt") would be much more frequent than coincidences between a nuclear γ -ray and one of the quanta. This would tend to mask delayed coincidences largely because of the 180° angular correlation of the annihilation quanta.

Because the side channel system was rather non-linear, to exclude all the pulses due to annihilation radiation in one of the counters would have necessitated such a high bias setting as to reduce seriously the efficiency of detection of the nuclear radiation. On the other hand, with the proposed geometry, a setting which passed say 5% of the "annihilation pulses" increased almost twofold the efficiency of the system, without adversely affecting the working of the apparatus.

An Upper Limit to the Lifetime of the Positron in Aluminium

If it is assumed that only the valence electrons contribute to the annihilation process, and neglecting the formation of positronium, lifetimes in metals of several times 10^{-9} s. might be expected. Lifetimes of this magnitude would give rise to a readily measurable exponential "tail" to the delayed coincidence resolution curve.

The source was therefore enclosed in sufficient aluminium to stop all the positrons and a resolution curve was obtained. From a logarithmic plot, a lifetime of about 1.5×10^{-9} s. was deduced. Repeating the

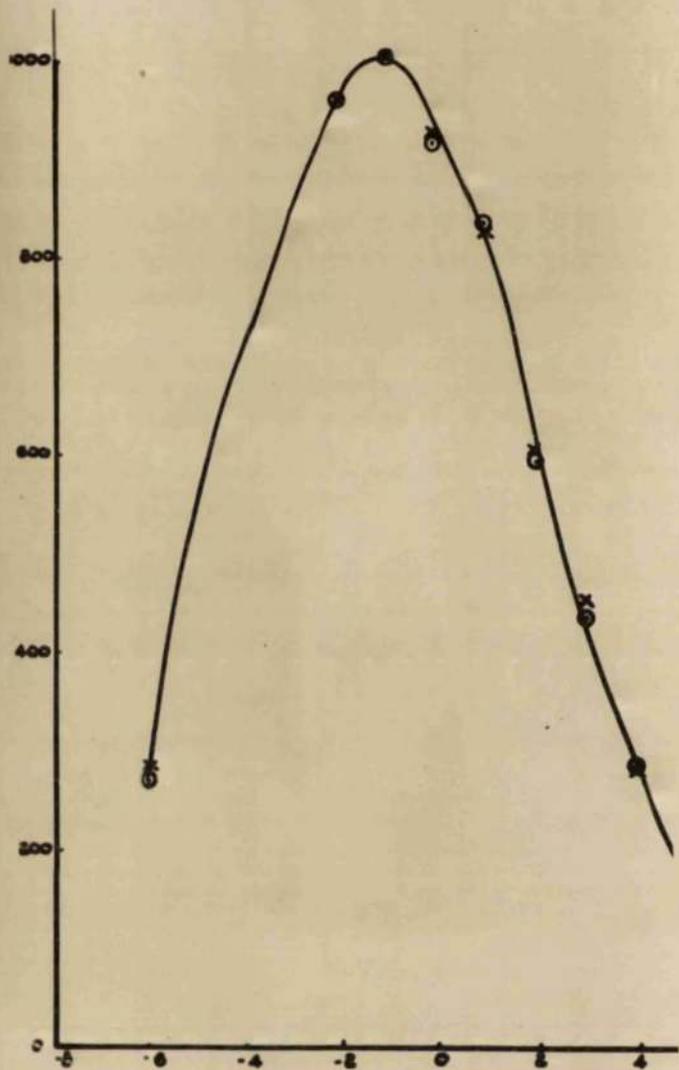
measurement with changed settings of the side channels gave a considerably different result. The introduction of a mass of scattering material was also found to influence the result. Change in the counting rate also produced changes.

Further experiments along these lines using a source of Co^{60} confirmed that a "tail" dependent on the apparatus and its settings might be observed.

With the highest settings of the discriminator bias of the side channels the tail seemed to correspond to a mean life of about 10^{-9} s. and we may therefore conclude that the lifetime of positrons in aluminium is less than this. Similar results were obtained with other metals.

From these preliminary experiments it was clear that the more refined techniques, in which the resolution curve for the suspected delayed events $[F(x)]$ is compared with a prompt resolution curve $[P(x)]$, were necessary. A search of the literature revealed no suitable source for the purpose of comparison. It was hoped to obtain some information therefore by careful comparison of the resolution curves for annihilation in different metals. Taking note of the lessons of the experiments described above, scattering material in the vicinity of the source was reduced to a minimum and when metal A was being compared with metal B both samples were present at once in the form of a sandwich — A,B,Source,B,A — in order that the residual scattering might be the same in both cases. Arrangements were also made for the accurate replacement of source and counters. Pairs of delayed coincidence curves were obtained, in the first of which A was next the source, and in the second, B. The relative displacement of the centroids of the two curves is equal to the difference in the mean

No. of coincidences



Delay $\times 10^{-9}$ s.

Fig. 7

Coincidence resolution curve of coincidences between the 1.28-Mev γ -ray of Na^{22} and 511-kev annihilation γ -rays.

- o - positrons annihilating in lead.
- x - positrons annihilating in iron.

life of the positron in the two materials. In this manner the positron lifetimes in lead, iron, magnesium and aluminium were compared. The curves obtained for the comparison of lead and iron are shown in fig. 7 and these are typical of the results obtained. The number of random coincidences was always negligible.

Clearly, from the figure, we can see that the difference between the lifetime of positrons in the metals investigated is very small. The results were analysed by the method of Bay assuming a single lifetime. The errors of the experiment arise from two sources. The statistical errors in estimating the position of the centroid are small for the following reason. G , the position of the centroid is given by

$$G = \frac{\sum x F(x)}{\sum F(x)} \quad \begin{array}{l} x = \text{delay inserted} \\ F(x) = \text{no. of coincidences} \end{array}$$

With each $F(x)$ is associated a probable error $\pm \delta F(x)$, but since the same terms appear in numerator and denominator, a positive error in the numerator corresponds to a positive error in the denominator. The errors in the quotient are therefore small.

The second source of error arises from instrumental drifts. If, for example, the gain or discriminator bias on one of the side channels varies, the resultant change in accepted pulse height will result in a change in the mean delay on that side. The magnitude of this error can be estimated from the shift of the point on the extreme left of the curve $F(x)$ over the period of the experiment.

The probable errors from these sources seemed to be about $\pm 5 \times 10^{-11}$ s. Within these errors no difference was found between the positron lifetime

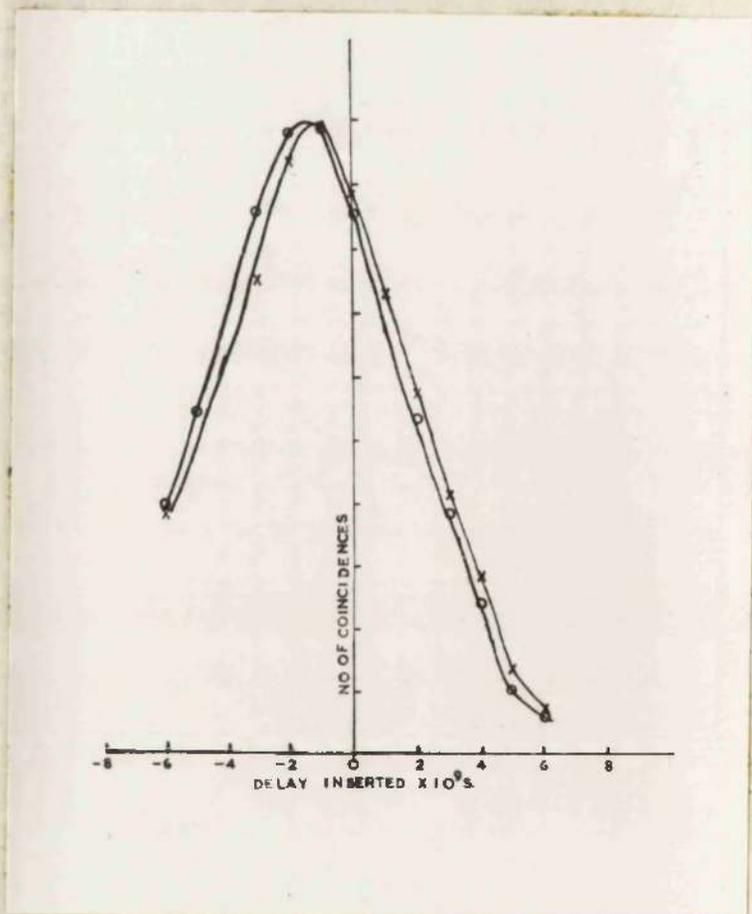


Fig. 8

Comparison of lifetime of positrons in wax and lead.

Resolution curve with wax next the source - X

Resolution curve with lead next the source - O

Displacement of centroid is $(2 \pm .5 \times 10^{-10} \text{ s})$

in any of the pairs of metals investigated. We may thus conclude that the difference between the lifetimes of positrons in any of the metals, aluminium, lead, iron and magnesium, is less than 5×10^{-11} s.

Comparison with Non-Metals

By similar experiments to the above, the lifetime in several non-metals was compared with that in aluminium. As it was felt that the presence of conduction electrons might enter into the explanation of the results in metals, the non-metals were chosen to be insulators, viz., paraffin wax, mica and Distrene. For these, a definite shift in the position of the centroid was observed. The curves for aluminium and wax are shown in fig. 8. It was not possible to be certain from the curves whether the assumption of a single exponential lifetime was justified.

Applying the analysis of Bay, assuming a single period, a mean delay of about 2×10^{-10} s. with respect to the lifetime in metals was observed for each of the materials tested. The errors in this determination are similar to those for the metals. We can therefore conclude that in these non-metals some positrons have mean lives longer than 2 or 3 times 10^{-10} s.

These results were confirmed by those of DeBenedetti and Richings (1952) which were published towards the completion of this work. By very similar experiments but using a different method of analysis, they studied a large number of both metals and non-metals.

Discussion

From the experiments of this chapter we have three main results, viz., that in the metals studied, positrons have mean lives which are equal to within 5×10^{-11} s.; that this mean life is less than 10^{-9} s.; that in non-metals some positrons have a mean life exceeding that in metals by at least 2 or 3×10^{-10} s.

Even when it is assumed that all Z electrons are available, a difference of at least 1.3×10^{-10} s. would have been expected between the lifetimes of positrons in lead and aluminium. However, all the indirect evidence (Du Mond et al. 1949; DeBenedetti et al. 1950), forces one to the conclusion that many times fewer electrons, viz., only the valence electrons, can take part in the process of annihilation from unbound states. This greatly increases the difference between the experimental and theoretical results. The conclusion is therefore suggested that bound states of the positron and electron may be rapidly formed in the material. To a first order, the lifetime observed would then depend on the properties of the positron-electron system and be independent of its environment. Particularly in metals, ~~the~~ the conversion of triplet states of these systems to the rapidly annihilating singlet states seems highly probable. (Thus one might expect a lifetime of about 10^{-10} s. in metals.)

If this process were slightly less effective in amorphous insulators such as wax or distrene, one would expect $\frac{3}{4}$ of the positrons to have a lifetime τ intermediate between 10^{-7} s. and 10^{-10} s. This would produce a centroid shift of approximately $\frac{3}{4} \tau$. For the insula-

tors studied, τ would appear to be of about $3-4 \times 10^{-10}$ s.

Another possibility might be that the basic theory gives a value for the cross-section per electron that is too low by several orders of magnitude. This would allow the variation of the lifetime in metals to be within the errors of the comparison experiment. The lifetime in lead would require to be not more than a few times 10^{-11} s.

The considerations set out in the discussion above show the importance of measuring the absolute value of the lifetime of positrons in a metal. Failing this, an experiment to set an upper limit of 10^{-10} s. or less would be of considerable value.

With the resolving time of the present apparatus it would be difficult to detect the possibly complex lifetimes of positrons in the non-metals examined. It was decided therefore to attempt to reduce the resolving time further with this end in view.

A D D E N D U M

Subsequent to the experiments described above, and while the apparatus was being developed, another experiment was performed which will be presented here.

The strong dependence of the annihilation probability on the spins of the positron and electron (cf. Chapter II) might possibly have been expected to produce a difference between the lifetimes of positrons in iron compared with other metals if the free annihilation picture was valid. This is because of the aligned spins of the d-shell electrons in the domains. No such difference was observed and, as a final check, the effect on the positron lifetime of applying a saturation magnetic field to soft iron was examined. This experiment was suggested by G. M. Lewis.

The sample was in the form of a hollow cylinder $\frac{3}{4}$ " diameter and $1\frac{1}{2}$ " long, whose walls were $1/16$ " thick. Threaded through the cylinder were a few turns of copper wire, through which a large current ~ 20 amps could be passed. Four turns of fine copper were connected to a fluxmeter. This instrument indicated a flux of some 7000 gauss per cm^2 .

The Na^{22} source described above was placed inside the cylinder less than 10% of whose surface was obscured by the copper wire. The experiment consisted of measuring the number of coincidences $F(x)$ at a fixed delay x ($x = 3 \times 10^{-9}$ s., see fig. 7) alternately with and without the field. If, due to the field there was a change in the mean life of the positrons, the resolution curve would be displaced by δx say. The counting rate at delay x would be approximately $F(x + \delta x)$, i.e., a

change of $\frac{dP}{dx} \delta x$. If $\frac{dP}{dx}$ is large, a small change δx will produce an appreciable change in counting rate. The procedure of taking alternate counts eliminates the effect of apparatus drift. The total number of counts with the field was the same as that without the field to within the statistical errors which were less than 1%. This corresponds to a change of less than 3×10^{-11} s. The low value of the limit set here arises from the fact that a cycling procedure can be applied without the risk of physical displacement, etc.

CHAPTER VIII

The Complex Behaviour of Positrons in Some Solid Materials

Introduction

One of the possible explanations of the phenomena described in the last chapter involved the formation in the material of positronium. The apparent absence of a long period associated with its triplet state was ascribed to the probable rapid conversion of triplet to singlet state by exchange processes. The most promising materials in which to observe these complex phenomena appeared to be in plastics. There being no apparent reason to choose one more than another, the most readily available, distrene, was chosen for study. Bell and Graham (1952), in an abstract, had reported a second long period in some materials which increased the interest in the investigation.

Before proceeding with these experiments, two major improvements in technique were made. The first was the preparation of a much thinner source in which less than 3.0% of the positrons annihilated. The second was a reduction in the resolving time by about 50% in actual use.

Preparation of the source

In order to reduce the amount of annihilation in the source and backing, a new and thinner source of Na^{22} was prepared. Turnings from a magnesium target bombarded with deuterons from the Birmingham 37-inch cyclotron contained the Na^{22} , estimated to be about 100 μc in activity. The standard method of extraction (Irvine and Clark, 1948) required a high

level of chemical technique, and particularly required the handling, and centrifuging, of relatively large volumes of active solution. The chief advantage of this method is the high efficiency of extraction of Na^{22} .

As this was not required, a more simple method was adopted. The turnings were boiled in distilled water which slowly converted the magnesium, and the sodium within it, to their respective hydroxides. This process was continued for several days until a sample of the solution showed an adequate activity. The solution was decanted off and slowly concentrated by evaporation. Due to its low solubility, the magnesium hydroxide came down as a precipitate, leaving the Na^{22} in solution. Evaporation was continued until less than 1 cc. of solution remained.

The Na^{22} was deposited on 0.0002 inch aluminium foil (free from pin holes) and covered with a similar foil. The total thickness of source and backing was 4 mg/cm^2 . Assuming a mean energy of 300 kev for Na^{22} positrons, the fraction stopped in the source would be about 3%.

The Coincidence Apparatus

Several modifications were made to the coincidence apparatus. These resulted in a decrease in resolving time from 2.4×10^{-9} s. to 1.4×10^{-9} s. for similar settings. The pulse height in the side channels was at the same time made more nearly a linear function of the energy expended in the phosphor.

To reduce the capacity coupled to the collector of the photo-multiplier, Bell et al. (1952) took the side channel signal from the last dynode. It was realised by us that the same purpose would be served by taking the

signal from an earlier dynode (D.11 in fact) where it would be more nearly proportional to the energy expended in the phosphor. The intermediate stages were heavily decoupled.

To obtain a sharper cut off of the limiting valve, Bell used zero bias, i.e. grounded cathode. In our apparatus this also eliminated the cathode decoupling condenser (2 μ f.). The latter had almost certainly an inductive component which would slow up the response of the circuit as a whole. A new miniature valve, type C.V.138, was used, with an H.T. voltage of 100 v. A negative signal of 0.8 volts applied to the grid reduced the current to 20% of its standing value. With the previous arrangement 1.5 volts were required. The valve could therefore be cut off in a much shorter time.

A new layout of the photo-multiplier and associated valves was designed to facilitate assembly and give a more compact earthing system. Parallel to the brass plate on which the head amplifier valves were fitted, the "photo-tube" was supported by two cradles of tufnal. The earth points of the photo-multiplier and limiting valve were grouped round the signal output plug which was soldered to the baseplate.

NOTE

The importance of the earth return system at these frequencies was brought home to the author when investigating a change in layout at the mixing point. Here the signal leads were brought much closer together at the expense of a much greater (\sim 3 inches) separation of the earth points. An increase of 50% in resolving time was observed, attributed to the loss of high frequency components of the signals. The resolving time returned

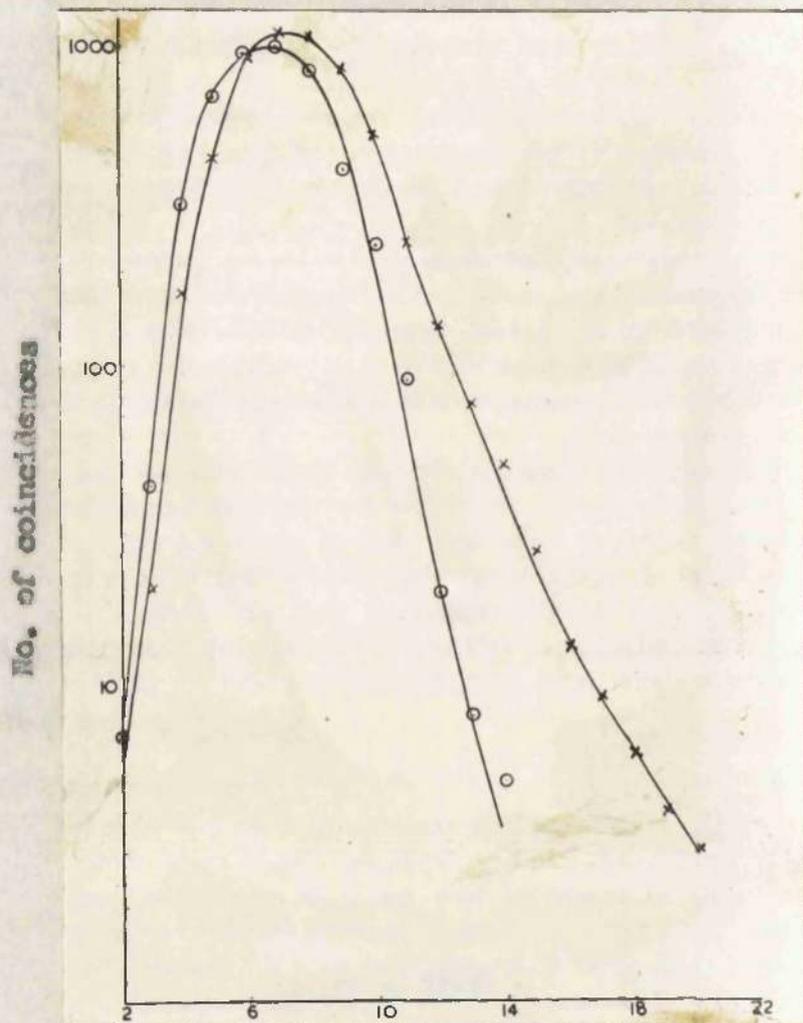


Fig. 9

Coincidence resolution curves of coincidences between the 1.28-Mev γ -ray of Na^{22} and 511-kev annihilation γ -rays.

x - positrons annihilating in distrene
 o - positrons annihilating in aluminium

to its previous value when the circuit was restored to its original state.

In the side channel a cathode follower replaced the head amplifier.

A stabilized 100-v. power pack was constructed to supply the new limiting valve and cathode follower.

The Lifetimes of Positrons in Distrene

The modified apparatus described above was used for these experiments. The phosphors were bottles of terphenyl 1 inch in diameter by 1 inch long. The side channel output was tested using a kicksorter and found to be approximately linear with γ -ray energy. A sandwich was prepared as previously, with two distrene plates 0.05" thick and two aluminium plates 0.02" thick. The distrene plates were machined from rod.

In order to achieve greater geometrical efficiency, the sandwich was placed on the axis of the counters. Due to the linearity of pulse height with energy, it was now possible to exclude the $\frac{1}{2}$ -Mev - $\frac{1}{2}$ -Mev coincidences without excessive reduction in the efficiency for 1.2-Mev - $\frac{1}{2}$ -Mev coincidences.

The experimental procedure was to take delayed coincidence resolution curves with alternately aluminium and distrene next the source. In taking these curves, a set of readings was first obtained for odd multiples of 10^{-9} s. delay, and then for even multiples. Any instrumental drifts would show up as departures from a smooth curve.

A pair of resolution curves obtained is shown in fig. 9 plotted on a logarithmic scale. Several qualitative features of the curves may be noted. Firstly, on the left hand side, both the aluminium and the distrene

curves rise from the same point, showing that no drift had occurred. Then we note that the distrene curve tends to a straight line for long delays only. There is a distinct concavity towards the right which is absent in the aluminium curve. This concavity is a certain indication that more than one period is present. If we compare the period measured from the slope of the tail with the centroid shift, we find that the latter quantity is only one-third of the former.

Repeating these experiments with different settings of the discriminators, the slope of the tail extending beyond the aluminium curve gave a consistent result. Also, when all pulses due to $\frac{1}{2}$ -Mev γ -rays were excluded from one of the counters, further increase in that discriminator had no effect on the displacement of the centroid. Small variations in the centroid shift with the position of the low energy discriminator were noticed until the latter was set so as to exclude γ -rays of energy less than 280-kev. This was thought to be due to the exclusion of coincidences between the 1.28-Mev γ -ray and its Compton scattered photons. These would give rise to prompt coincidences.

As points far out on the tail made ^{not} negligible contributions to the higher moments of the resolution curves, the number of random coincidences was determined. This was done directly by delaying the pulses from annihilation radiation by 9×10^{-9} s. (the point - 9 on fig. 9). No real coincidences could then occur and the total count was the random coincidence rate.

Analysis

From the tail of the curve a period of $2.5 \pm .5 \times 10^{-9}$ s. is measured. It is clear that there is another period which is much less than this. To apply the analysis of Bay, the simplifying assumption was made that the short period was close to that of aluminium. The analysis then gave the long period as $2.5 \pm 0.3 \times 10^{-9}$ s. for a fraction $0.27 \pm .03$ of the positrons. This agrees well with the lifetime as measured from the tail and supports the assumptions made. A further analytical check was made by applying the formula for a higher order of moments. In principle this should give a conclusive check. Unfortunately, the statistical errors, carried through the calculation arithmetically, increase with order of moments, and the check confirmed the assumptions with no greater weight than the comparison with the "tail". The magnitude and fraction of the long period has since been confirmed within the errors by the work of Bell and Graham (1953), who found the second period to be $2.3 \pm .2 \times 10^{-9}$ s. These workers were able to detect a short lifetime differing by 2×10^{-10} s. from that in aluminium. If this correction is applied to the results here it brings them very close to those of Bell and Graham.

The Variation of the Lifetime of Positrons in Distrene

Bell and Graham (1952) reported that in fused quartz there was a second long period, while in crystalline quartz this effect was not observed. A considerable influence of the gross structure of materials on the annihilation process seemed indicated. The magnitude of these effects might depend on the space parameters of the sample, so it was decided to investigate this point.

Some properties of plastics, e.g. density, vary from sample to sample, depending on its history, and method of preparation, and, in particular, strain effects due to rolling, extrusion, etc., may remain "frozen" in the plastic. Further evidence of this comes from variation of optical properties (R. C. Gray, Private communication). The samples used in the experiments described above were machined from a 2-inch diameter rod. The experiments were repeated using samples cut from the edge of a large sheet and also a sample made from many layers of 0.002-inch foil. No significant departures from the results obtained previously were noted.

The effect of temperature

For similar reasons, it was decided to investigate the effect of heating the specimen. In case there might be residual effects, the experiment was tried at a series of gradually increasing temperatures - 15°C, 30°C, 68°C, 100°C, 150°C. At the latter temperature the distrene softens.

In this experiment, the source sandwich was in an electrically heated oven. A thermo-couple, one junction of which was in contact

with the sample, was used to measure its temperature. Due to the proximity of the heated sample to the liquid phosphors, it was found necessary to cool the latter. They were surrounded by a thin copper can through which air was blown from the compressed supply.

Only a small effect was observed over this temperature range. At 150°C, the long period was found to have a value $2.8 \pm .3 \times 10^{-9}$ s. for a fraction $.25 \pm .03$ of the positrons.

The results at intermediate temperatures were:-

<u>Temperature °C</u>	<u>$\tau \times 10^{-9}$ s.</u>
15	$2.5 \pm .3$
30	$2.4 \pm .3$
68	$2.6 \pm .3$
100	$2.9 \pm .3$
150	$2.8 \pm .3$

Although the errors on individual results are large, there would appear to be a definite increase of the long period with time of the order of 10% for the 115° rise in temperature. Bell and Graham (1953) have reported a value 1.7×10^{-9} s. for this quantity at 196°C, a decrease of 25% for 220°C change in temperature, i.e. a similar rate of change at both high and low temperatures.

The radiation associated with the long period

In gases, (see Chapter XI) a long period is sometimes associated with three quantum radiation. DeBenedetti and Seigel (1952) had found

very little three-quantum annihilation from metals using three NaI scintillation counters in coincidence. The long period in distrene raised the possibility that three-quantum annihilation might be more frequent in that material. An experiment similar to that of DeBenedetti was attempted, using the apparatus available.

Three NaI scintillation counters were used. The phosphors were cylinders 0.75-inch diameter and 1.5-inch long. Having aluminium for reflector, these were mounted with paraffin in perspex containers. The pulses from the scintillation counters were amplified and discriminated by the side channels of the fast coincidence unit and a third similar amplifier and discriminator. The triple coincidence unit of the fast coincidence apparatus was also utilized.

The source of positrons was Cu^{64} in the form of .0002" copper foil irradiated in the Harwell pile. Source strengths of about 10^4 positrons per second were chosen. Some days after use, when the source had decayed to a suitable value, its activity was measured with a Geiger counter. The ratio of positrons to electrons given in Nuclear Data was assumed.

There are two main sources of spurious triple coincidences, viz., statistical coincidences and coincidences due to scattering. The former may be computed if the number of counts in each channel is measured and the resolving time known. In order to reduce the proportion of such counts, the source strength was kept small. In order to reduce the latter, the counters were raised on light stands 12 inches above the bench, and the discriminators were set so as to exclude radiations of energy less than 200-kev.

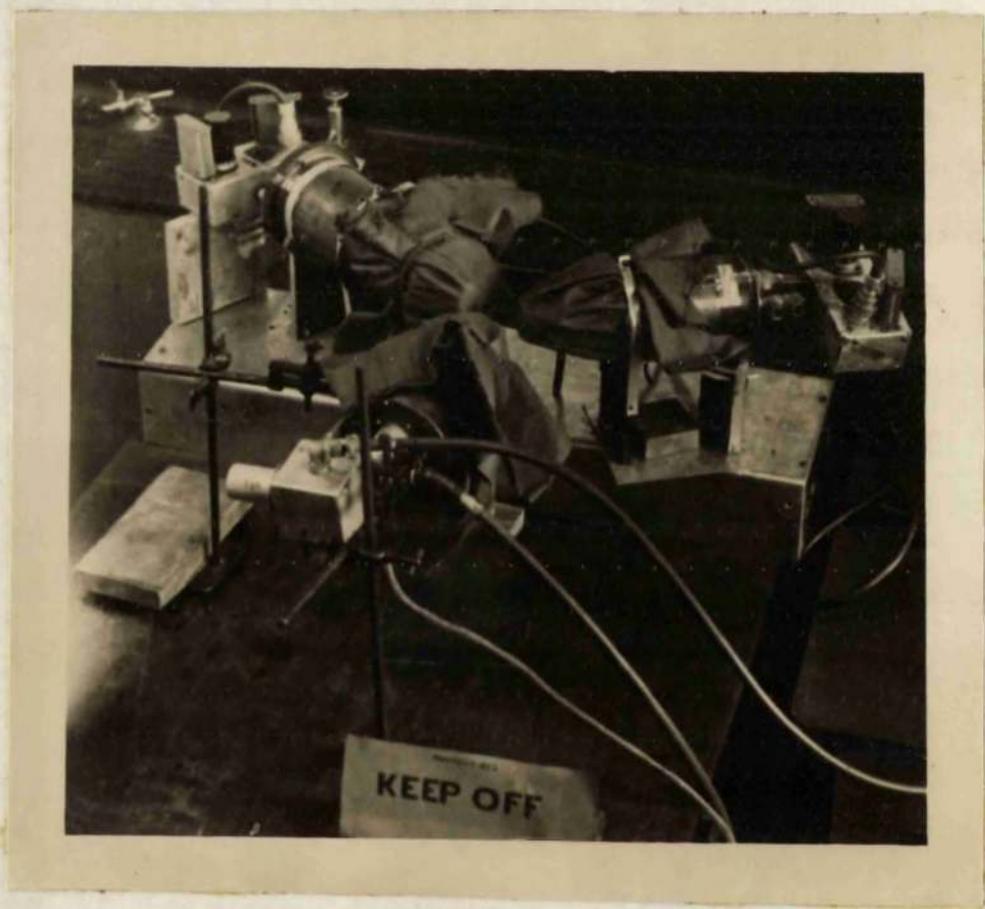


Fig. 10

Arrangement of counters for the
triple coincidence experiment.

The three counters were set up to be co-planar and making angles of 120° with one another. Fig. 10 shows a photograph of the arrangement. The source was placed in a slit in a distrene rod 0.25" diameter and raised into the plane of the counters. The number of triple coincidences was observed when set up in this way. The single counting rate in each channel was also noted. One of the counters was then lowered out of the plane, its axis making an angle of 45° with the plane. Allowing for the decay of the source with time, the counting rate in this counter was unaltered. Also its position relative to each of its neighbouring counters was unchanged. Therefore the scattering would be as before. Since the quanta from three-quantum annihilation are co-planar, no true coincidences would be observed. A run in this position gave the background. A typical hour's run gave (correcting for source decay)

$$\text{Counts co-planar} = 25 \pm 5$$

$$\text{Counts non-co-planar} = 13 \pm 3.5$$

Similar results were obtained from several such runs.

Analysis

The number of triple coincidences T_c is given by

$$T_c = 3 N_3 e_1 e_2 e_3$$

where

N_3 = No. of three-quantum annihilations

$e_{1,2,3}$ = efficiencies of counters 1, 2, 3

N = total number of positrons emitted

From a rough calculation, it is clear that N_3 is small compared with N .

The single counts in the scintillation counters were therefore almost entirely due to two-quantum annihilation. From the known source strength and the single count rate, the efficiency of the counters for 511-kev radiation was found. Assuming that the γ -ray spectrum of three-quantum annihilation is similar to that observed in gases (Chapter XI), and remembering that radiation of energy less than 200-kev was excluded, an estimate was made of the efficiency for three-quantum detection. This estimate may be in error by at least 20%.

The calculation gave the fraction of positrons annihilating by three-quanta to be 1.5%. This figure may be in error by a factor of two. Even at its lowest value it was three times the value suggested by DeBenedetti et coll. (1952) for copper. In this calculation the effect of angular correlation has not been taken into account as the data did not warrant this refinement.

An attempt to compare the amounts of three-quantum radiations in distrene and aluminium

Another attempt to investigate the three-quantum annihilation used a method suggested by the experiments of Pond and Dicke (1952) in their study of positronium in gases. This method uses two counters set up far apart so that the detection of the highly correlated two-quantum annihilation photons is favoured.

We wish to find the difference in the number of three-quantum annihilations in distrene and aluminium. If the number of "triplets" in distrene

exceed the number in aluminium, the number of two-quantum annihilations will be less by this amount. The experiment therefore consists of finding the difference in two-photon coincidence rate when distrene and aluminium respectively are next to the source.

Two of the counters of the previous experiment with the same amplifiers, etc., were connected in coincidence. Two inputs of the triple coincidence unit were connected together so that it operated as a simple coincidence unit. The counters were set up 20 cm apart.

The discriminators were set on the least rapidly varying part of their bias curves, corresponding to a point just below the photo-electric peak on the differential pulse height curve. This was done in order to obtain maximum stability.

Scattering of one of the quanta even through a small angle would result in the loss of a coincidence. In order that the probability of scattering should be the same when the annihilation is in distrene as when it is in aluminium, the sandwich technique was again used.

The experimental procedure was to take alternate runs with aluminium and distrene next to the source. Six pairs of runs, each of 60 minutes' duration, were taken. While statistics could have been improved by further runs, the deviations due to gain changes in the electronics did not warrant this. The results obtained were:-

Distrene 371 ± 3 coincidences/minute

Aluminium 370 ± 3 coincidences/minute

From this we can only set an upper limit on the proportion of three-quantum annihilations. This upper limit would seem to be $\sqrt{18}$ in 370, i.e. not more than 1.25%.

To measure a difference of, say 1%, to an accuracy of $\pm 10\%$ would have required an apparatus of very high stability. The efficiency of detection would then have had to remain constant to $\pm .05\%$ over a period of several hours. The major effort required to produce such an apparatus was considered beyond the scope of the writer's work.

NOTE:

The triple coincidence experiment gave a result that 1.5% of the positrons decayed by the emission of three-quanta. The errors in this quantity were at least a factor of two. It may therefore be taken to set a lower limit of, say 0.5%. The two counter experiment sets an upper limit of 1.25% compared with aluminium. It would seem therefore that the result must lie in the vicinity of one percent.

The results of this chapter will be discussed in Chapter X.

CHAPTER IX

The Mean Life of Positrons in Aluminium

Introduction.

In the discussion of Chapter VII the importance was stressed of obtaining an absolute measurement of the lifetime of positrons in metals. The experiments of that chapter showed that this lifetime was less than 10^{-9} s. If the hypothesis that annihilation in metals was associated with positronium were correct, a lifetime in the region of 10^{-10} s. might be expected. The technical preparations for the experiment were based on the assumption that the lifetime was of the order of 10^{-10} s. The root of the problem appeared to be that of providing a suitable prompt resolution curve with which to compare the delayed curve for coincidences between the 1.28-Mev Nuclear γ -ray of Na^{22} and the delayed annihilation radiation. Several instructive attempts to overcome this difficulty were made before the final solution was found. These will now be mentioned briefly.

Artificial Source

The ideal solution of this problem would have been to find a radioactive source giving two γ -rays in prompt coincidence, having energies of 1.28-Mev and .511-Mev respectively. A search of the literature revealed no such source nor even anything approximating to it. The source Co^{60} has two γ -rays of energy (1.17 and 1.33-Mev respectively)

not very different from that of the Na^{22} γ -ray. If the γ -rays of Co^{60} were to fall on a scatterer, the secondary γ -rays would, by the Compton effect, have a continuous distribution of energies. By suitable collimation it would be possible to select those of approximately 500-kev. Coincidences would then be taken between the direct γ -rays and selected γ -rays. While sound in principle, this method breaks down on the grounds of intensity. The double collimation, viz., from source to scatterer, and from scatterer to crystal, greatly reduces the efficiency of the low-energy counter. The maximum permissible counting rate in the direct counter set an upper limit to the source strength that could be used. Even with this source strength ($\sim 10 \text{ mCi}$) the coincidence counting rate was unacceptably low. The ratio of real to random counts was also adversely affected. For these reasons the method was abandoned.

The next attempt made use of the "method of reversals". This method was used by Bell, Graham and Petch (1952) in their studies of the delay in emission of the γ -ray of Au^{198} . The method may be described as follows. With the announcing radiation in Counter A and the decaying radiation in counter B, a delayed resolution curve $F(x)$ is obtained. The apparatus is then readjusted so that the announcing radiation is detected in counter B and the decaying radiation in counter A. This gives a curve $F(-x)$. In the absence of delays due to pulse height, the centroids of these curves will be separated by a distance 2τ (where τ is the mean life for emission of the decaying radiation).

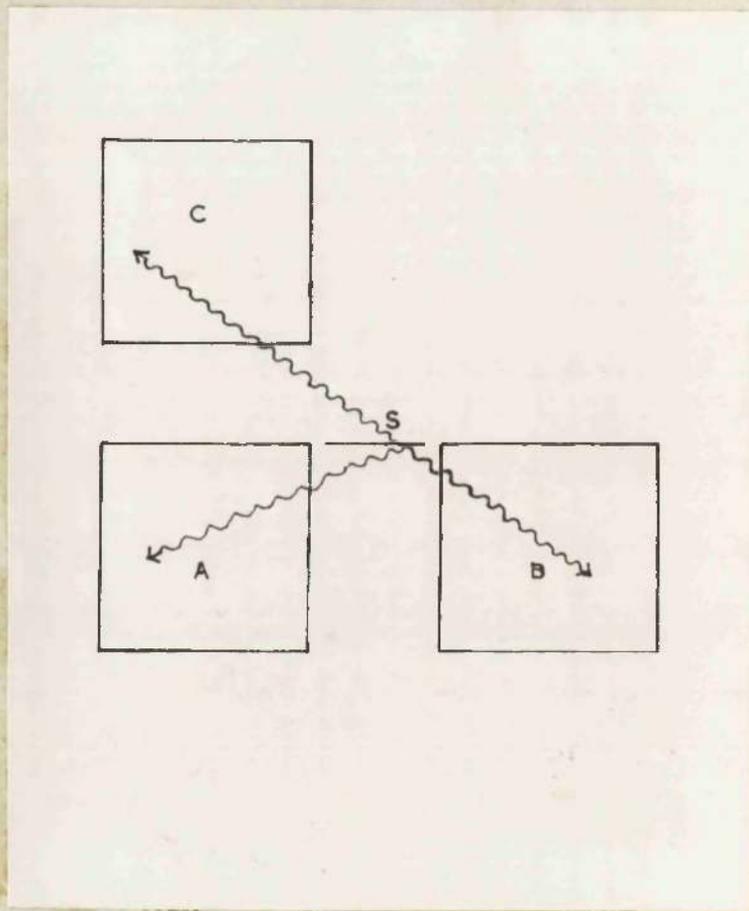


Fig. 11

Arrangement of phosphors for
proposed absolute determination
of lifetime of β^+ in a metal.

The method eliminates any fixed delays in transit time, etc., but not delays due to pulse height.

It was realized by the writer that the Compton electron spectrum of both the nuclear and annihilation γ -rays was a continuous distribution of electrons down to zero energy. By the use of a kicksorter in each channel, pulses of equal amplitude could be selected from both the announcing and decaying radiations. In order to be certain that the annihilation radiation only was detected in one counter, a third "slow" coincidence counter was to be introduced to detect the second annihilation quantum. The geometrical arrangement is shown in fig. 11. Due to the fact that the annihilation quanta^{are} emitted at exactly 180° , if a nuclear γ -ray is detected in B, (see fig. 11), and a corresponding annihilation photon in A, the second annihilation photon cannot possibly be detected in the third counter. Only if the desired detection of a nuclear γ -ray in A, and annihilation quanta in B, takes place, can the second quantum be detected in C. The experiment then consists of obtaining a curve $F(x)$ with C in the position shown. By moving C to the corresponding position with respect to counter A, the curve $F(-x)$ is obtained. The pulse heights are unaltered in the two experiments, so that the distance between the centroids would be 2τ .

The coincidence counting rates in this experiment were estimated to be less than those of, say the experiments of Chapter VII, by a factor of 10 or 20. An increase of source strength by a factor of four was expected to give the minimum adequate counting rate. This source was

ordered from Harwell and was expected to be ready by the time the apparatus was prepared. Delivery took place in fact one year later. In the meantime a simpler though less elegant method had been devised using much of the apparatus for this proposed experiment, and the method was successfully carried out.

The basis of this method was to compare the coincidence resolution curve for the Na^{22} nuclear γ -ray and annihilation γ -rays with a similar curve obtained using Co^{60} . A kickscaler replaced the discriminator on the low energy channel so that the same pulse amplitude was picked out from the Compton recoil spectra of both the annihilation γ -ray and the γ -ray of Co^{60} . Since the mean energy of the Co^{60} γ -rays does not differ greatly from that of the Na^{22} nuclear γ -ray, the discriminator excluding annihilation radiation was retained.

This method is similar in principle to the first method described. The latter required two collimations - source to scatterer, and scatterer to crystal; and, further, the efficiency of the scatterer was small. Here only one effective collimation, viz., the selection of pulse amplitudes, is required, and there is no intermediate scatterer. The efficiency is therefore at least two orders of magnitude higher than the previous method. For the Na^{22} source it is only less in efficiency than the comparative annihilation experiments by a factor of about 5.

If we suppose that the proposed technique successfully overcomes the difficulties due to pulse height delays, to what extent does the experiment provide an absolute measure of the positron mean life or

lives? From the discussion of Chapter VII, page 65, it was concluded that the delay in emission of the 1.28-Mev γ -ray with respect to the instant of emission of the positron was of the order of 10^{-12} s. or less.

An uncertainty of this order is therefore introduced into the measurements.

The intermediate state of Co^{60} was shown by an angular correlation experiment (Metzger and Deutsch 1950) to have spin 2 and even parity. The ground state is known to have spin zero and even parity. The second γ -ray from Co^{60} must therefore be emitted in an electric quadrupole transition. The mean delay in its emission with respect to the first will be of the order of 10^{-12} s. But since both counters are sensitive to both the announcing and decaying radiations, the centroid of the coincidence resolution curve for Co^{60} will coincide with that of $P(x)$. Any small delays will appear as a broadening of the distribution only. It would therefore appear that in principle the measurement will differ from the absolute value only by uncertainty in the delay of the Na^{22} nuclear γ -ray. This error is likely to be much smaller than the experimental errors, so that the measurement may be considered absolute within these errors.

The Apparatus

A block diagram of the apparatus is shown in fig. 12. The basic apparatus, shown in full line, does not differ from that described in Chapter VIII. Discriminator D_1 was set as before to exclude annihilation

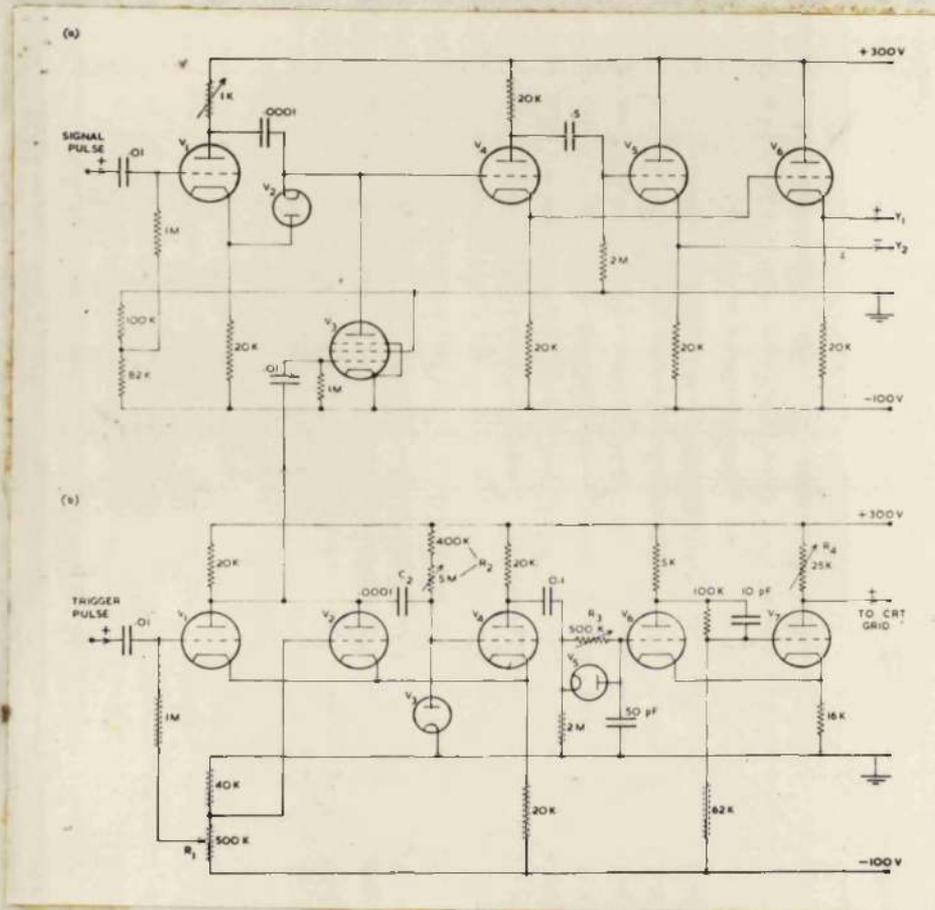
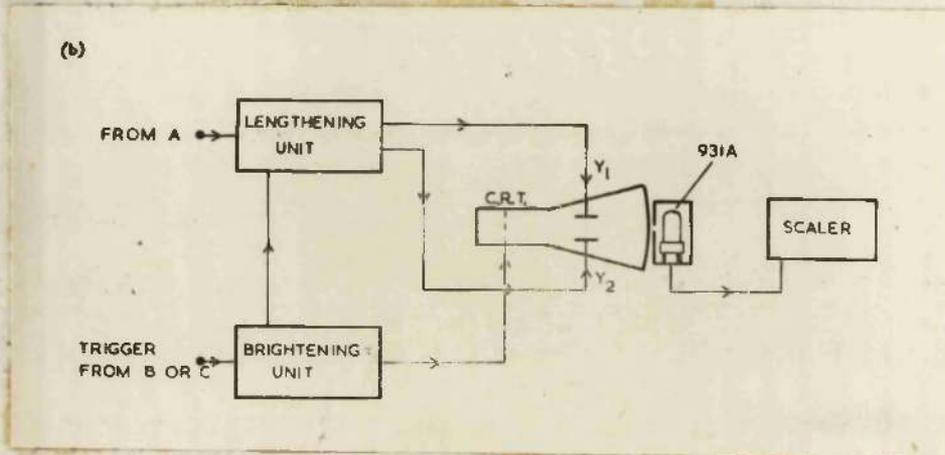


Fig. 13

Block diagram and circuits of
 single channel C.R.T. kicksorter
 Circuit (a) - Pulse lengthening unit
 Circuit (b) - Brightening unit

radiation. At first, an additional discriminator, D_4 , was connected in parallel with D_2 . The outputs of this and the triple coincidence unit 3-C were fed to an anticoincidence unit. The triple coincidence unit gave a pulse when a coincidence occurred in which the pulse in C_2 exceeded the bias of D_2 . If, however, it also exceeded the bias of D_4 , this coincidence was rejected. Thus, only pulses from C_2 having amplitudes between the bias levels of D_2 and D_4 could give rise to acceptable coincidences. The two discriminators and the anti-coincidence unit acted as a kicksorter. Later, the full apparatus shown in fig. 12 was used. There, discriminator D_2 was set below the lower limit of the kicksorter channel and so did not affect this aspect of the working of the apparatus. A pulse from the triple coincidence unit opened the gate of the kicksorter and allowed it to accept pulses within its channel. Due to the delay introduced by travelling through the foregoing electronics (particularly the discriminators), the output pulse from 3-C was delayed by about 1.25 μ s. To compensate for this, the pulses from F were delayed artificially by introducing a delay cable (type Z.14M, characteristic impedance 1000 Ω) between there and A_4 .

The same piece of apparatus served both as a kicksorter here and as an anti-coincidence unit in the earlier method. This very versatile piece of equipment, of which several are in use in the laboratory, was supplied by Mr. G. H. Lewis. Essentially it consisted of a C.R.T. As a simple kicksorter, pulses from the amplifier were fed to a lengthen-

ing unit and a brightening unit in parallel. These units supplied flat-topped pulses such that the brightening pulse lay entirely under the flat portion of the lengthened pulse. The former was applied to the biased off grid of the C.R.T. and the latter to the Y deflecting plates. The pulse was represented on the screen by a bright spot whose position was a measure of its amplitude. In practice, the amount of D.C. bias required to bring the spot to a fixed position was measured. A photo-multiplier followed by a scaler, counted the spots occurring between the edges of a slit which was placed against the screen. The channel width was determined by the width of the slit.

Since the brightening unit and lengthening unit could be actuated by separate pulses, this allowed the spectrum to be gated (as was done above by pulses from S-C). On the other hand, with zero bias, pulses entering the brightening channel can be counted. Then, a pulse applied to the deflecting plates acts as a rejector signal.

The advantage of the second method over the first was that in the first, the width of the channel depended on two completely independent biases and thus was subject to considerable variations. These gave rise to changes in the counting rate and therefore erratic points in the coincidence resolution curves. It served well for preliminary work, though the final results were obtained by the second method. The discussion of the remainder of the chapter will refer to the second method.

Preliminary Tests

Using the kicksorter with its normal narrow channel width, the pulse spectrum of Co^{60} from amplifier A_1 was examined: a cathode follower was used. Similarly the pulse spectrum due to the nuclear γ -ray of Na^{22} was looked at. These spectra were found to be very close to one another. By gating the kicksorter with the output of D_1 , the lower energy parts of the spectra were cut off and only the spectrum of pulses passed by D_1 was observed. From a number of runs like this it was possible to measure the mean pulse height accepted by D_1 as a function of discriminator bias. The spectrum of annihilation γ -rays from Cu^{64} and the γ -rays of Co^{60} in channel 2 was also examined.

The use of a kicksorter in place of a discriminator in channel 2 reduced the efficiency of this channel for the detection of annihilation γ -rays.

With Na^{22} in aluminum and the settings of D_1 and D_2 used in previous experiments, a counting rate of 300 per minute at the output of S-C was obtained. The width of the channel was adjusted to give about 100 c.p.m. at its optimum position. From the known width of this channel and the measured spectra, the mean pulse height accepted in the channel from annihilation γ -rays and from Co^{60} was measured as a function of channel position (i.e. kicksorter back bias) over the possible working range. When the centroid of the distribution was plotted as a function of the ratio of the pulse heights, there appeared to be a linear relationship between them. If $\Delta \lambda$ is the displacement in time of the

centroid of the coincidence resolution curve with ratio H_2 with respect to that with H_1 , it was found that

$$\Delta \lambda = (H_2 - H_1) (1 \pm 0.1) \times 10^{-9} \text{ s.}$$

i.e., if the ratio $\frac{\text{pulse height channel 1}}{\text{pulse height channel 2}}$ for Co^{60} differs from that for Na^{22} in aluminium by, say 5%, an error of 5×10^{-11} s. would be introduced.

Similar experiments were carried out with the Na^{22} source in aluminium and good agreement with the above was obtained.

These results enable corrections to be made, for e.g. the small difference between the Co^{60} and Na^{22} γ -rays. This amounts to 10^{-11} s.

Final Measurement

When these preliminary experiments were completed, the position of the channel was adjusted so that it occurred at a part of the annihilation γ -ray spectrum similar in shape to the Co^{60} spectrum, and particularly so that the mean pulse height in the channel was the same. With this setting, several runs were made, each consisting of first a resolution curve for the Co^{60} γ -rays and then a curve for the Na^{22} source placed between aluminium plates. A typical pair of curves is shown in fig. 14. It is not possible to say from these whether or not only one period is present, but from comparison of the tails it would seem unlikely that any partial period of more than 3 or 4×10^{-10} s. is present. If we analyse the results on the hypothesis that only one period is present, the following results are obtained. Five measurements in all were made. Runs 1 and 2 used the two discriminators, runs 3, 4 and 5 the kicksorter.

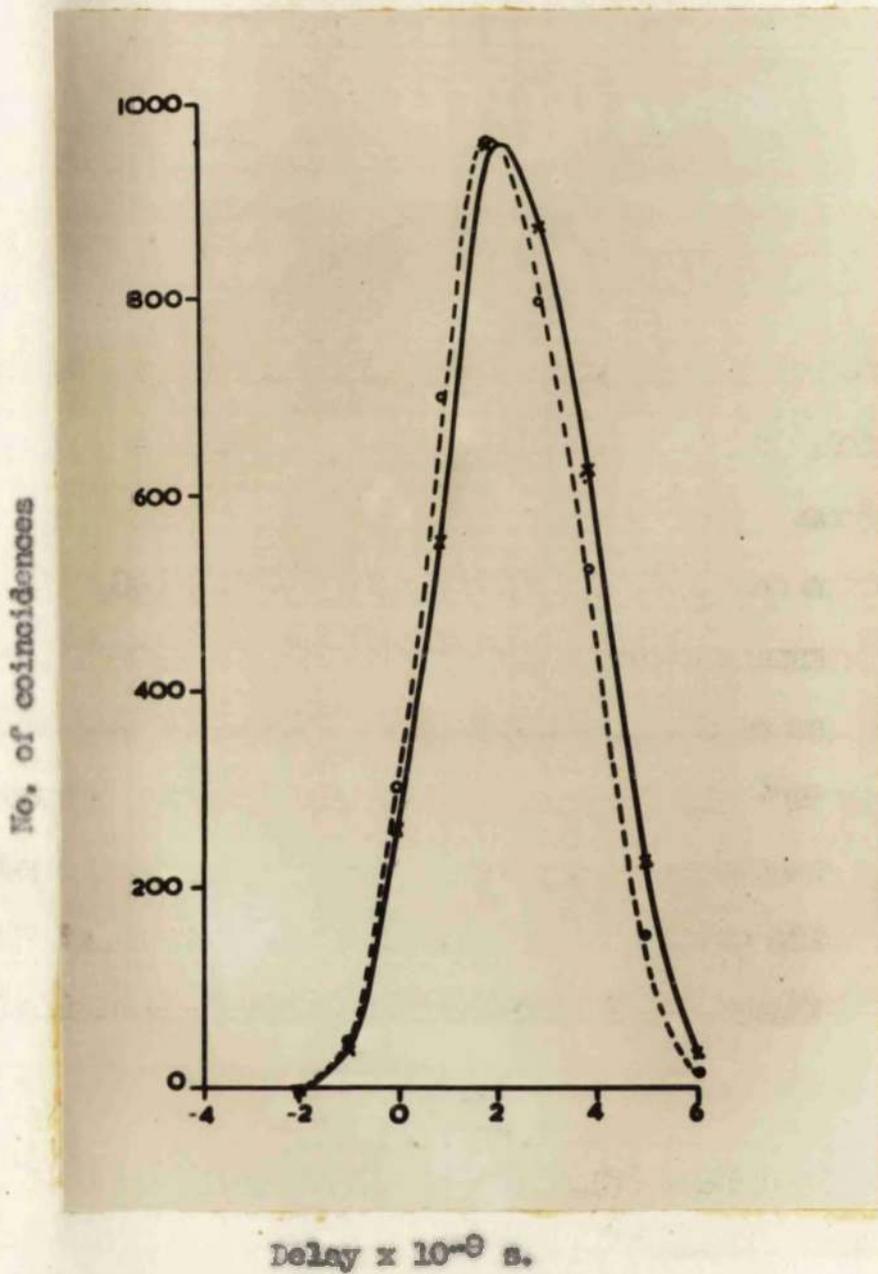


Fig. 14

Coincidence resolution curves of coincidences between -

- x — 1.23-Mev γ -ray of Na²² and 511-kev annihilation radiation from aluminium
- o — 1.17 Mev and 1.33-Mev γ -rays of Co⁶⁰

<u>Run</u>	<u>τ</u>
1	1.5×10^{-10} s.
2	1.9×10^{-10} s.
3	0.7×10^{-10} s.
4	2.0×10^{-10} s.
5	1.9×10^{-10} s.

Since aluminium was the source backing, the only foreign matter was the source material, in which, it was estimated, less than 1% of the annihilations took place. Errors arose from two main sources: statistical errors, and those due to drifts in apparatus. The statistical errors on individual measurements are small $\sim 2 \times 10^{-11}$ s. The error from pulse height drifts was estimated from the displacements of successive resolution curves of Co^{60} (or Na^{22}) to be about 4×10^{-11} s. Thus the error on individual measurements was 6×10^{-11} s. In the paper published on this subject, this error was quoted along with the mean of the above. A more correct figure would be the standard deviation giving the final result.

$$\tau = (1.6 \pm .4) \times 10^{-10} \text{ s.}$$

This result will be discussed in Chapter X.

Delay of Co^{60} γ -rays

In Chapter III, when discussing the relationship between $P(x)$ and

$P(x)$, it was stressed that both curves should cut the x axis at the same point on the left when the decaying radiation was detected in one counter only. For Na^{22} this latter condition was satisfied. The point at which it cut the x axis on the left was therefore the same as that for a true comparable $P(x)$. If there was delay between the emission of the first and second γ -rays of Co^{60} , both the right and left hand sides of the curve would be displaced outwards from those of a true $P(x)$. In particular, the left side would be displaced to larger -ve. values of x near the x axis. This would be noticeable as a displacement with respect to the Na^{22} in this region. No such systematic displacement was noticed in the five pairs of curves taken. A lower limit to the mean life of the intermediate state of Co^{60} may be set by considering the R.M.S. of the random displacements of the resolution curves. The mean displacement error of a single measurement quoted above is 6×10^{-11} s. For five measurements we may therefore set a limit of about 3×10^{-11} s.

Technical Note

A further development of this apparatus is suggested by selecting the pulses in channel 1 with a kicksorter, as well as those in channel 2. This technique may be generalized to study the lifetime of any γ -emitting state that is reached by the emission of a γ -ray from a higher level.

No such general method has previously been devised for use when the "exponential tail" method breaks down. The method of Bell et al., using two β -ray spectrometers, is applicable only where internal conversion is strong. Indeed the method imposes no restriction on the nature of the announcing and decaying radiation which could for example be neutrons.

In order to use the first moment analysis of Bay, which is by far the most powerful due to the small statistical errors, it is necessary to distinguish the announcing from the decaying radiations. When these are both neutrons or γ -rays, they must differ sufficiently in energy to allow this.

One possible comparison source of γ -rays which commends itself, is coincidences between the 510-kev γ -rays from positron annihilation. Here, no doubts as to possible delays need be entertained. A restriction on the use of this is that one of the pair of radiations with which it is being compared must have an energy sufficiently less (by at least a channel width) than 510-kev. This is in order that the upper channel which must be on a suitable portion of the 510-kev spectrum may not accept any pulses from the lower energy γ -ray.

For the reasons mentioned before, Co^{60} would be a suitable comparator. Here, a similar restriction on the energy of the γ -rays would apply. At still higher energies, coincidences between positron-electron pairs might be suitable.

The accuracy attainable by these methods is quite high. The errors in the positron measurement come largely from apparatus drifts. Since

the amplifiers of the fast coincidence unit were not stabilised by feed back, this procedure could greatly increase the stability. Further, new E.H.T. power supplies for the phototubes (Type 1055) of an order of magnitude of greater stability than those used (Type 1007) are now available. It might therefore be possible to increase the accuracy of measurement by this apparatus to ± 1 or 2×10^{-11} s.

These recommendations have now been adopted by Mr. G. M. Lewis and others in their studies of the excited states of nuclei.

CHAPTER X

A Discussion of the Experimental Results

In Chapter II, several possible hypotheses regarding positron annihilation in solids were put forward. They may be summarised as follows.

1. That annihilation takes place in free collisions between thermalised positrons and the valence electrons in the material.
2. That positronium is rapidly formed in the material and the lifetime is that of positronium. Mechanisms such as exchange, or interaction with lattice vibrations, would convert the long-lived triplet state into the short-lived singlet state.
3. Compound formation, where the positron forms a bound state with a negative ion, might take place. There is also a strong possibility that more than one of these processes might take place.

The writer's main results may also be enumerated.

- (1) In metals, e.g. aluminium, iron, lead, magnesium, the mean life is constant to within $\pm 5 \times 10^{-11}$ s. and is about 1.6×10^{-10} s.
- (2) In certain amorphous materials such as distrene, there are two distinct lifetimes, one of which is close to that in metals, the other, for about one-third of the positrons,

is 2.5×10^{-9} s. The latter lifetime increases slowly with temperature.

- (3) In distrene, the proportion of three-photon annihilation is between 0.5% and 1.5% of the total number of annihilations.

These results have been extended to a great many more materials by the work of Bell et al. (1953) and DeBenedetti and Richings.

It is possible from the above information to rule out the first hypothesis. In the case of aluminium for example, if we assume all three valence electrons are available, the theory predicts a mean life of 7.5×10^{-10} s. - five times greater than the value observed by us. The mean life in metals should be inversely proportional to

$$\frac{A}{dN}$$

A = atomic weight

d = density

N = number of available electrons per atom

Over the metals investigated by the writer and others, this quantity varies by more than a factor of 10, while only a factor of 2 is possible within the extreme limits of error. Finally, the hypothesis is unable to predict two independent periods such as are found in distrene and many other amorphous materials. It would seem therefore that this hypothesis cannot by itself account for the experimental results.

The evidence for the second is much stronger. On this, we would definitely expect to observe more than one period, though a high

probability for conversion of triplet to singlet positronium would make the two periods indistinguishably close. Thus, the failure to observe a long period in metals and some crystalline solids would not constitute a serious objection to the proposal.

Another piece of evidence lies in the proportion of three-quantum annihilation. If the 2.5×10^{-9} s. period in distrene were due to the conversion of triplet positronium to the singlet state, we should expect a fraction of three-quantum annihilations given by

$$\frac{1}{3} \frac{2.5 \times 10^{-9} \text{ s.}}{\text{unperturbed lifetime of the triplet state}}$$

Taking the triplet lifetime in free space, viz., 10^{-7} s. (Ore and Powell, 1949), we obtain 0.8%. This falls between the limits of 0.5% - 1.5% suggested by our experiments. A recent experiment by Pond gives the result 0.88%, agreeing even more closely.

The fact that $\frac{1}{3}$ of the positrons are associated with the long period raises some problems. Before the results by Bell were published, showing that this fraction occurred in all cases where a second long period was observed, the author suggested that this was the fraction of positronium molecules that found their way into holes in the material where interaction with other components of the system was reduced. The expansion of such holes with increasing temperature, it was suggested, reduced the interaction with the surroundings further and gave rise to the increased mean life. It would seem, however, in view of the constancy of the fraction over a wide variety of solids

and liquids, that a more general explanation is required.

If only one-third of the positrons form positronium in the triplet state, a further ninth must form it in the singlet state, i.e, approximately half of the positrons form positronium. Suppose the remainder annihilate in free collisions. Let us consider, e.g., Teflon for which Bell has measured two periods. The mean life for free annihilations, assuming all the valence electrons are available, would be about 8×10^{-10} s. Due to competition with positronium formation, this would be halved. The small fraction annihilating from the singlet state direct would have a mean life indistinguishable experimentally from the free annihilation. Thus there would appear to be a short period of about 4×10^{-10} s. together with the longer period of those formed in the triplet state.

The experimental results described above, while not giving by any means a complete picture of the phenomena of positron annihilation in condensed matter, appear to form a sound basis for future work in this field. On the experimental side such work might include experiments of higher precision with shorter resolving times. More particularly, the quenching effect of strong magnetic fields on the long periods in distrene and similar materials. As a test of the expansion hypothesis, the effect of mechanical compression might be studied. The great lack in this field is detailed theoretical calculations. The experiments described above would seem to point out particular problems in need of theoretical attention.

SECTION IV

THE SPECTRUM OF ANNIHILATION RADIATION

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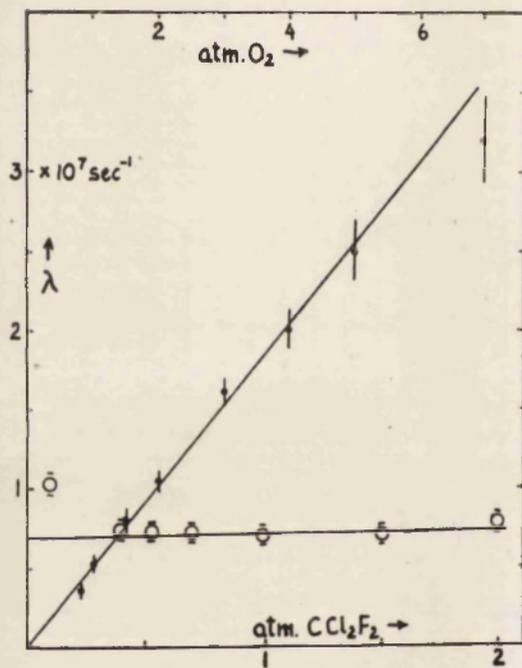


FIG. 1. Rate of decay of annihilation radiation in CCl_2F_2 (open circles) and in O_2 (full circles). The pressure scales are adjusted so that the abscissa corresponds to the same electron density in both gases.

Fig. 15

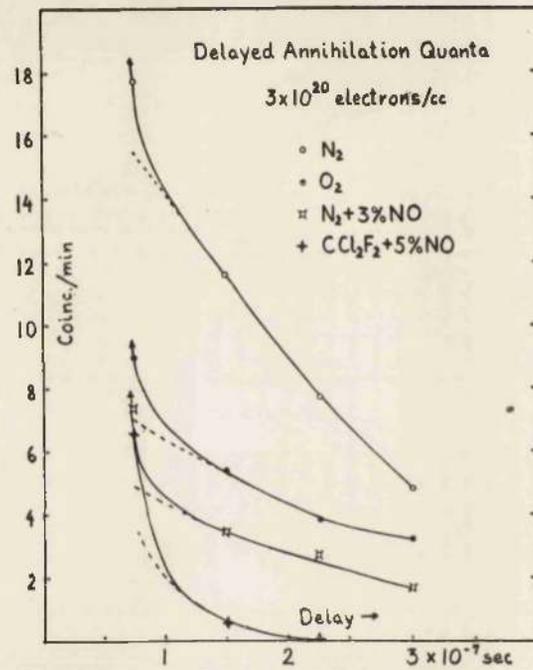


FIG. 1. Decay curves of positrons in several gases. The dotted lines are corrected for time resolution of the instrument.

Fig. 16

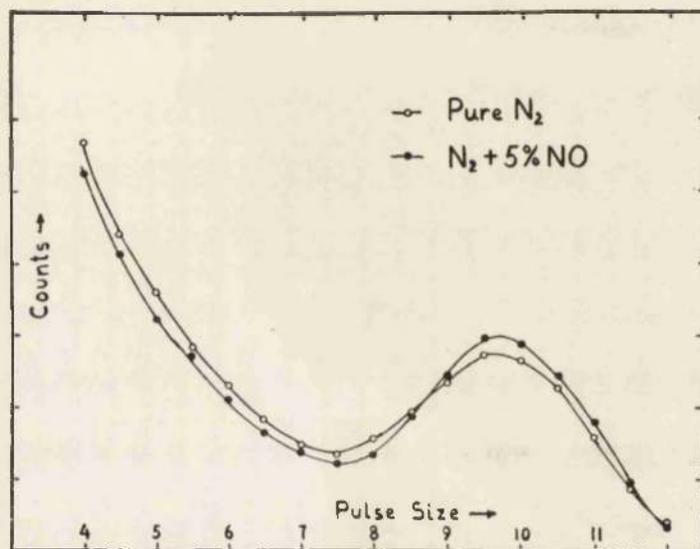


FIG. 2. Part of the scintillation spectrum of two-quantum (solid circles) and two-quantum plus three-quantum (open circles) annihilation. The uncertainty of individual points is indicated by the size of the circles.

Fig. 17

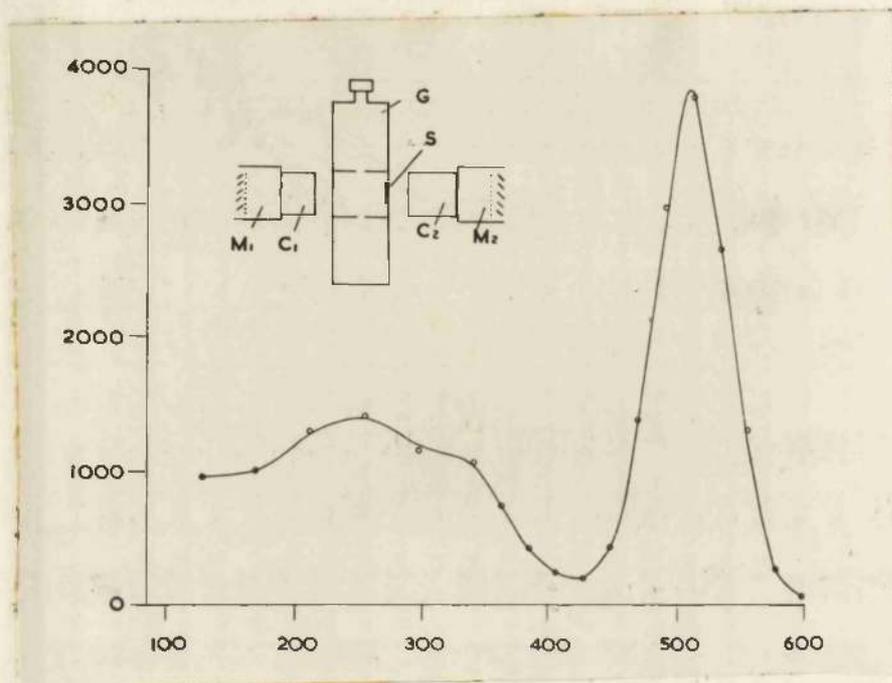
CHAPTER XI

The Spectrum of Annihilation Radiation from Positrons Annihilating in Freon and Oxygen

It was found by Deutsch (1951) that in freon, a lifetime for positrons was observed that was close to that predicted by Ore and Powell (1949) for positronium in its triplet state, and further, the lifetime varied only slightly with pressure (see Fig. 15). The addition of certain gases in concentrations of 5% or less caused the disappearance of this period (see Fig. 16). He concluded from this that in freon, positronium was abundantly formed, attributing the disappearance of the long period to the conversion of the triplet to the singlet state by an exchange process. This view received confirmation by many subsequent measurements, e.g., of the h.f.s. of positronium, quadratic Zeeman effect (Deutsch, 1951c, Deutsch and Brown 1951).

DeBenedetti and Siegel (1952) showed the presence of three simultaneous annihilation quanta from freon, in numbers considerably in excess of the fraction $\frac{1}{370}$ expected as the free annihilation hypothesis. Deutsch and Dulit (1951) showed that there were radiations of energy less than 510-kev (see Fig. 17) and that these were diminished in intensity by the addition of NO in the same manner as the number of annihilations with the long period.

Ore and Powell (1949) calculated the distribution of γ -ray energies to be expected in three-quantum annihilation. It was therefore decided to examine those annihilation quanta definitely associated with the long period in freon and compare the energy spectrum with that predicted by theory.



Energy in kev.

Fig. 18

Pulse height distribution obtained for the annihilation radiation of Cu^{64} positrons in copper.

Inset: experimental arrangement; S - source; G - gas container; C_1 C_2 - scintillators; M_1 M_2 - multipliers.

Oxygen stood out among the other gases as one in which the variation of positron mean life with pressure (Deutsch 1951a) obeyed the expected inverse law. It was therefore decided to investigate the spectrum from oxygen also.

The method proposed was that of delayed coincidences. Na^{22} was the source of positrons, the source being provided by the writer. This technique enables the delayed annihilation γ -rays to be selected from amongst the quanta from positrons annihilating in the source, container, etc., and from those, if any, having a short lifetime in the gas.

The Apparatus and Experimental Arrangement

The source of Na^{22} was mounted on, and covered by, aluminium foil .005" thick. It was placed, as shown in fig. 1 & inset, inside a cylindrical gas container 2" diameter, the wall of which was .012" copper. Two perforated phosphor bronze plates .005" thick, 1.5" apart, restricted the positrons to the central region of the cylinder.

The counter near the source had terphenyl in toluene as scintillator, occupying a volume $1\frac{1}{2}$ " diameter and 2" long (C_2). The other counter, which was to act as γ -ray spectrometer, had a block of NaI as scintillator 1.25" cube (C_1). This block was composed of two pieces cemented together along a common plane with silicone grease. The pieces were sawed from a random block to give the maximum size of crystal. The faces to be joined were ground plane with carborundum under paraffin. The scintillators were both packed in MgO as reflector and mounted on 14-stage E.M.E. photo-multipliers, Type 6262.

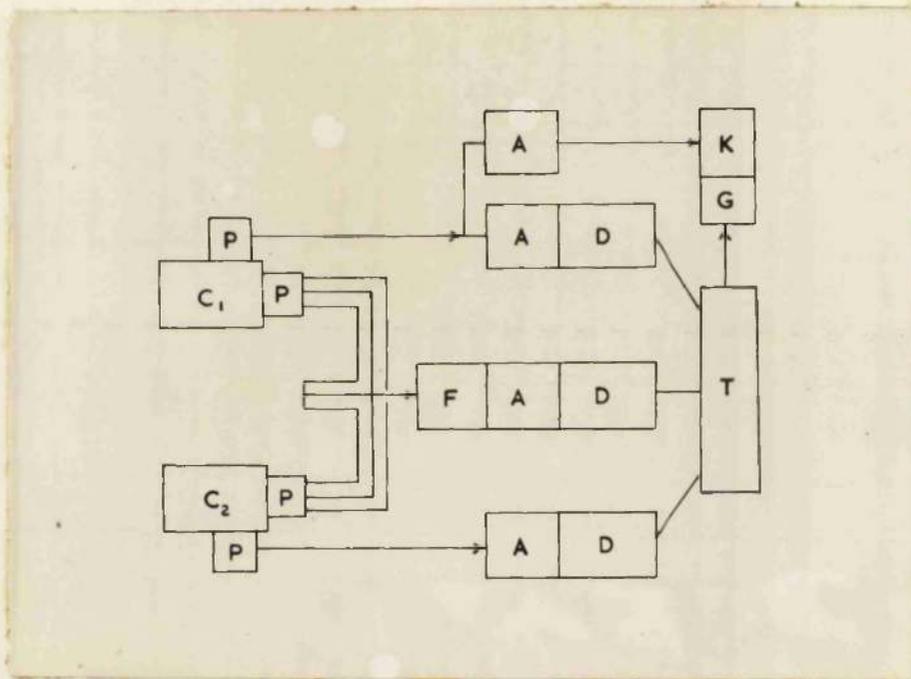


Fig. 19

Block diagram of apparatus.

C_1 C_2 - counters; P - preamplifier;
 F - unit receiving fast coincidences;
 A - amplifier; D - discriminator;
 T - triple coincidence unit; G - gate;
 K - kicksorter

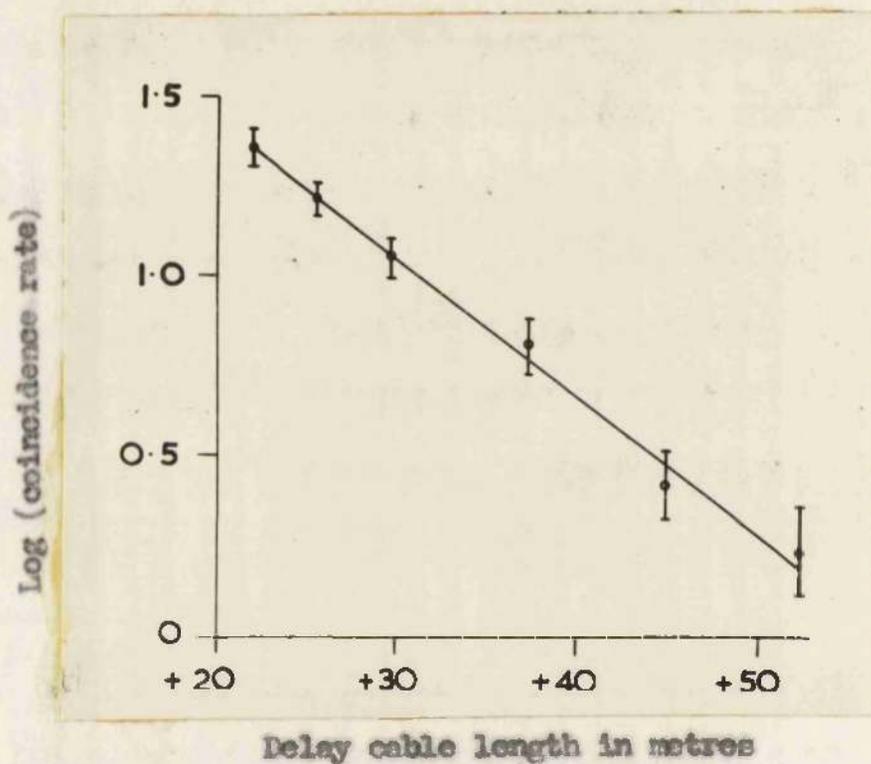


Fig. 20

Variation of coincidence rate
with delay for positrons in freon
at 4.8 atmospheres.

The ends of the iodide and terphenyl scintillators were respectively situated 1 inch and 0.5 inch from the cylinder. The system was set up on a light plywood table to minimise scattering. The electronic arrangements are shown in fig. 19. This consists of a fast coincidence unit of resolving time (in this application) 5×10^{-8} s. and a linear amplifier and kicksorter. The kicksorter is gated by pulses from T and the spectrum displayed there consists only of those pulses which have a delay with respect to a corresponding pulse in C_2 .

The fast coincidence unit is very similar in design to those described previously in this thesis. To obtain linearity, the side channel pulses were taken from D.10 of the photo-multiplier and the general layout was based on that of the apparatus of Chapter IX. The gated kicksorter is that described in Chapter IX.

Delay Times

Using only the fast coincidence unit, a measurement of the number of coincidences as a function of delay inserted was made when the cylinder was filled with freon. The latter was at a pressure of 4.8 atmospheres, which is close to its saturation vapour pressure at room temperature. From the slope of the tail of this distribution plotted on a logarithmic scale (Fig. 20) the lifetime found was

$$\tau = 1.25 \pm 0.10 \times 10^{-7} \text{ sec.}$$

This time is only a little shorter than that obtained by Deutsch (1961)

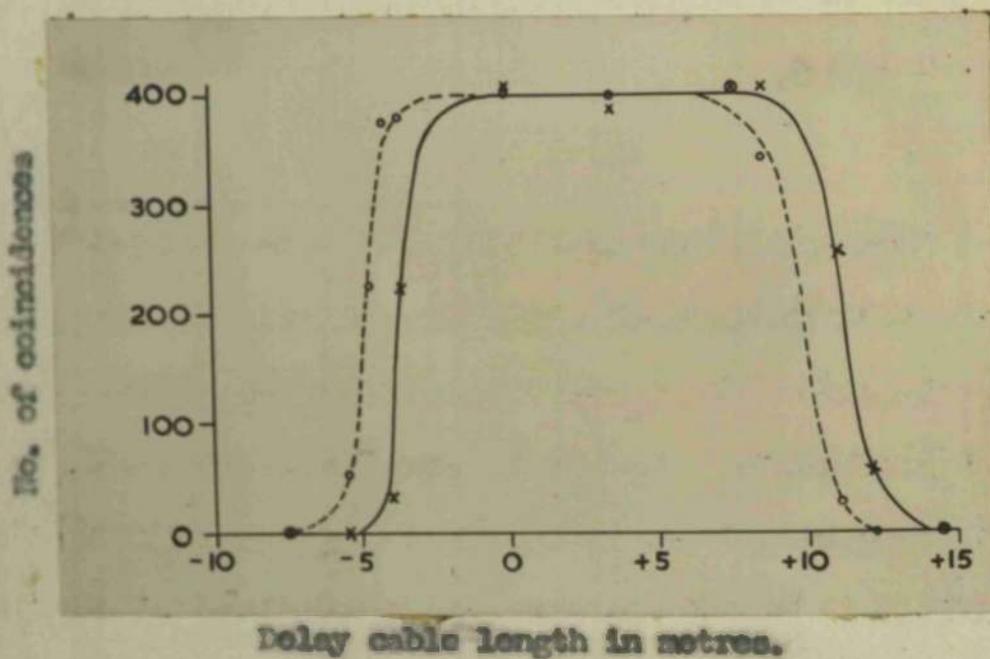


Fig. 21

Coincidence resolution curve between prompt pulses of a given height in the sodium iodide counter and 1.23-Mev γ -pulses in the other counter.

- x - electron energy 200-kev
- o - electron energy 400-kev

at lower pressures, confirming further that the variation of lifetime with pressure is small. The lifetime in oxygen at 2.4 atmospheres, found by Deutsch to be 8×10^{-8} s., was confirmed approximately by us.

As was pointed out in earlier chapters, spurious apparent delays between pulses may be introduced by differences in their amplitudes. The same effect may introduce distortion in the pulse spectrum selected to correspond to a particular delay. For example, the experimental delay x of a pulse is the sum of the delay in annihilation T and the delay due to pulse height d .

$$x = T + d$$

Large annihilation pulses correspond to a small value of d . Hence, for a fixed x , only those large pulses corresponding to a large value of T will be accepted. But the number of events with delay T decreases exponentially with T . Therefore fewer large pulses will be accepted than smaller ones and the low energy end of a spectrum will be emphasized. This effect will be serious only if d is not small compared with x . The value of x is limited by considerations of intensity and the ratio of real to random coincidences.

To investigate the delay due to pulse height, the kickster was used to select a narrow band of pulses. The cylinder was evacuated so that no delayed radiation was present.

Coincidence resolution curves were obtained for a number of pulse heights. Two of these are shown in fig. 21, viz., those corresponding to 200 and 400-Mev respectively. The delay here is 7.5×10^{-8} s. This

is small compared with the decay times in the gases chosen. From these results a small correction was made to the spectra obtained later.

Spectroscopy

In order to interpret a continuous γ -ray spectrum, it was first necessary to examine the spectrometer characteristic of the NaI crystal for line spectra in the energy region below 510-kev. Fig. 18 shows the spectrum of the annihilation radiation from Cu^{64} positrons annihilating in copper. It was observed with the NaI counter and kicksorter only. The low energy bulge in the multiple Compton distribution can be explained by backscattering of γ -rays from the rear parts of the crystal mounting. A similar spectrum was obtained from Na^{22} in the evacuated container with the two counters in prompt coincidence. The liquid filled counter detecting the Na^{22} γ -ray was positioned to minimize coincidences from its back-scattered Compton radiation.

The effect which the position of the positron at annihilation would have on the observed spectrum was examined. Curves of the fig. 18 type were taken with a Cu^{64} source enclosed in Cu situated at various points in the region 1-inch-3 inches from the iodide crystal. Similar investigations were made with Hg^{203} (280-kev γ -ray) and Cs^{137} (660-kev γ -ray) sources, screened where necessary from conversion X-rays. These spectral curves were little affected by positioning, the variation in the ratio of peak counts to total counts being less than 5%.

Counts per energy interval in arbitrary units

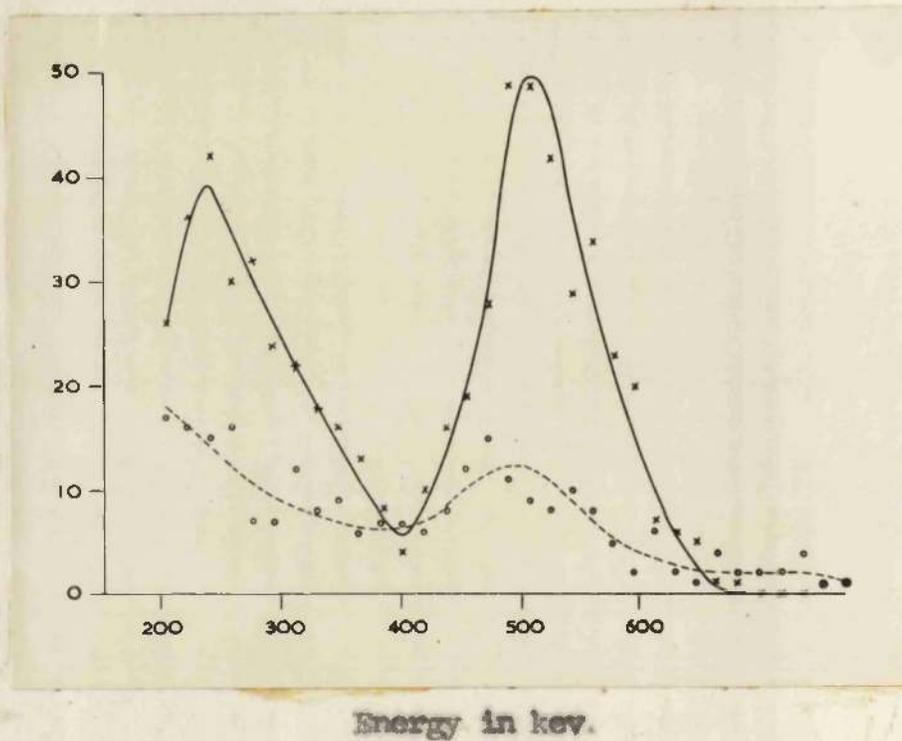


Fig. 22

Pulse height distribution obtained for
delayed annihilation radiation of posi-
trons in oxygen.

x — real counts, after subtracting randoms
o — random counts

Number of delayed coincidences per unit
energy interval.

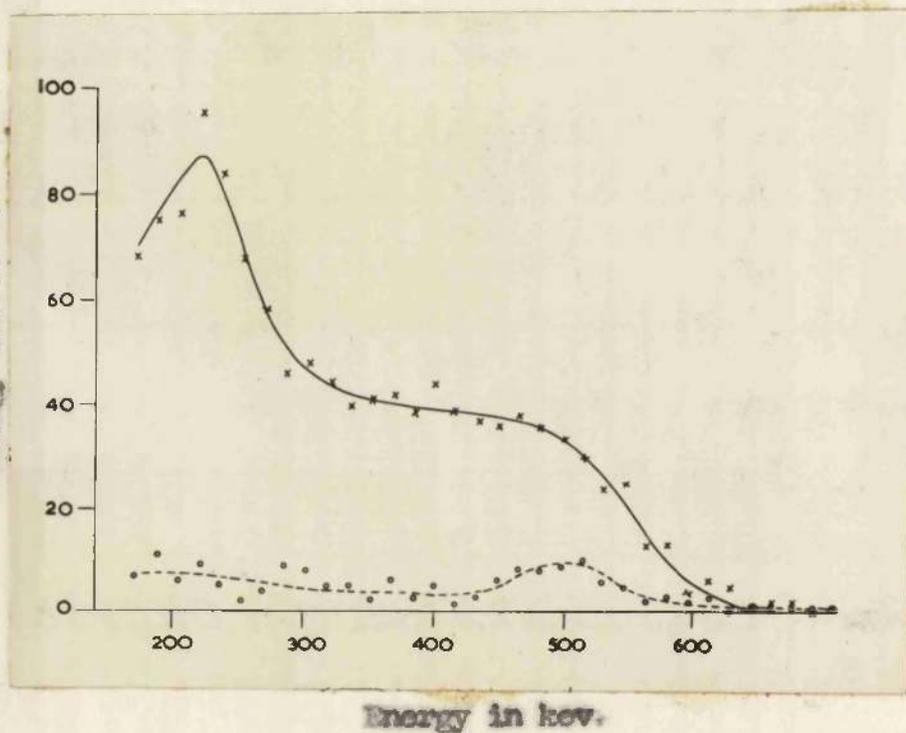


Fig. 23

Pulse height distribution for the
delayed annihilation radiation of
positrons in freeon.

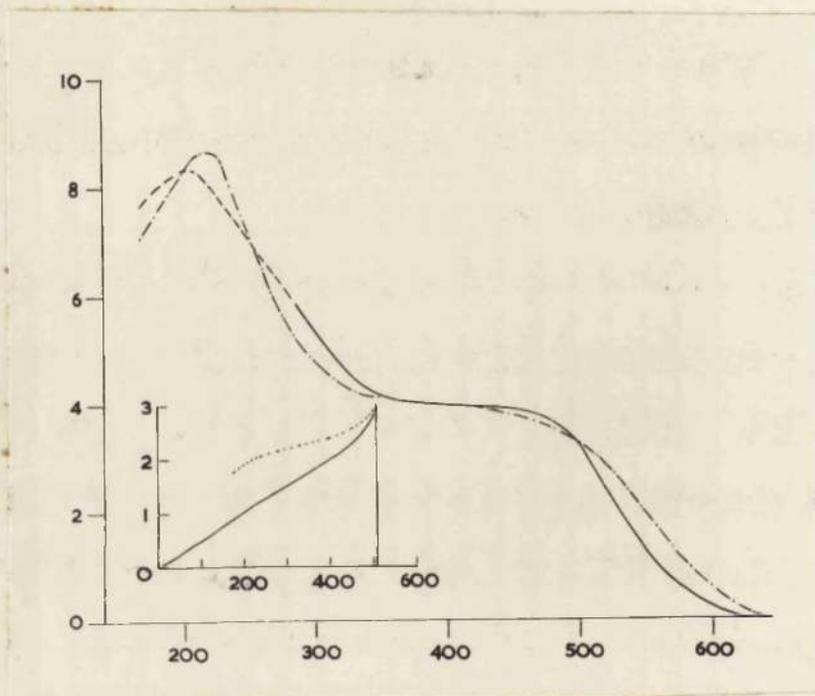
x - real counts after subtracting
randoms
o - random counts

Delayed Coincidence Spectra in Freon and Oxygen

In these experiments the pulses from the 1.23 Mev γ -ray channel (G_2) were delayed by 25 metres of cable (cf. fig. 1, the point + 25). The discriminator in the iodide channel was set at 140-kev to ensure satisfactory operation of the fast coincidence unit, and that in the liquid scintillator channel set at 600 kev to exclude annihilation radiation. The whole spectrum was recorded continuously by photographing the screen of the C.R.T. kickester. The spectrum of random coincidences was obtained directly by inserting 25 metres of cable in the iodide channel (the point - 25 on fig. 1, produced back). The results obtained are shown in figs. 21 and 22. The broken line shows the spectrum of random coincidences. This shows a distinct peak at 510-kev, providing a useful calibration point. The full line shows the "real" counts and is the difference between the observed spectrum and the random spectrum, the latter being subtracted point by point.

Fig. 21 for oxygen at 2.5 atmospheres exhibits clearly the presence of a strong 510-kev photo-peak, together with a low energy bulge corresponding to that in fig. 18 for copper. In the case of oxygen the main peak at 510-kev is less pronounced in height compared with the low energy bulge than it is in fig. 18. This is due in great measure to the larger width at half height of the main peak (20% compared with 14%). The increased width is in turn due to a variation in gain over the several hours of running time involved in the measurement and to a lesser extent to a slight gradual deterioration in the quality of the crystal since the

Counts per energy interval in arbitrary units



Energy in kev.

Fig. 24

Main curves ————— pulse height distribution expected
 - - - - - experimental curve of fig. 23

Inset: - - - - - Ore-Powell theoretical curve for three-quantum annihilation
 expected response with particular NaI crystal

experiments first began. Further, a 10% increase is to be expected in the low energy pulses due to the displacement of the resolution curve (cf. p 107 above). Lastly, measurements on scattering would suggest a rise of $\sim 5\%$ at the lower energies. It seems clear, therefore, that over the range investigated, the annihilation radiation from positrons in oxygen consists of a single line at 510-kev.

The results for freon at 5.2 atmospheres appears in fig. 23. Here the 510-kev peak is absent and the broad plateau exhibits the inhomogeneous nature of the annihilation radiations.

In fig. 24 inset, the theoretical Ore-Powell spectrum is shown. From this, the pulse spectrum to be expected from a scintillation counter was computed. Using the γ -ray absorption data given by Heitler (1944), the relative efficiency of the spectrometer crystal for γ -rays in this energy region was calculated and the modified curve shown by the dotted line (inset fig 24). From the experimentally observed line spectra at 280-kev, 510-kev and 680-kev the ratio of electrons in the peak to the total number energized was determined for these energies. The expected electron spectrum (not corrected for varying peak widths) is shown by the main curve of fig. 24. The lower energy portion shown by dashes involved a slight extrapolation but this part of the curve is essentially correct in shape and the errors there are estimated at less than 5%. The possibility of two quanta registering simultaneously has not been introduced here as two quanta running close together are not favoured in the theory on phase space grounds. Further, most of the positrons

annihilate close to the source (at this pressure of freon) and there the counter subtends a relatively small angle.

For comparison, the experimental curve of fig. 23 is also shown in fig. 24 and is seen to be in close agreement with the theoretical expectation over the range investigated.

Discussion.

In the case of freon, this experiment provides further very strong evidence that the long period for positrons annihilating in that gas is almost entirely associated with the triplet state of the positron and electron. For abundant annihilation from this state to be observed, it is necessary that the pair form a bound system, since the process is of the third order and could not otherwise compete with two-quantum annihilation. The work therefore gives good support to the theory of Deutsch that in freon, positronium is formed in great abundance.

There is not, however, a clear explanation of the results in oxygen. The effectiveness of this gas as a quenching agent for positronium (Deutsch) suggested a possible explanation, viz., that positronium was formed in the gas and then the triplet state converted to the singlet state.

It is safe to conclude from the spectral measurements that not less than 90% of the annihilation radiation associated with the 8×10^{-8} s. period is two-quantum radiation. We should then expect a lifetime

one-tenth of that for unperturbed positronium, viz., 10^{-8} s. In fact a mean life of 8×10^{-8} s. is observed. This theory cannot therefore be maintained.

The mean life agrees fairly closely with that to be expected on the simple picture of annihilation in collisions and the variation of lifetime with pressure (Deutsch) is in agreement with this view.

A P P E N D I X

The γ -rays of Lutetium

The investigation of the γ -rays of Lu^{176} was carried out as part of the work of Dixon, McNair and Curran, a full account of which may be found in their paper (Phil. Mag. 45, 683).

Two γ -rays of 270-kev and 180-kev respectively, together with one strongly converted γ -ray of 89-kev, were reported by Scharff-Goldhaber (Unpublished). A decay scheme was proposed by Goldhaber and Hill (1952) in which the β -rays and all three γ -rays were in cascade. Arnold and Sugihara (1953), found the γ -rays to have energies of 200 ± 20 -kev and 300 ± 20 -kev. A more accurate measurement of the γ -ray energies was of particular interest in view of their agreement with the predictions of the Bohr-Mottelson model of the nucleus. A precise estimate of the relative intensities of the γ -rays would test the validity of the decay scheme proposed by Goldhaber and Hill.

The technical problems in this investigation arose from the low specific activity of the source material. A NaI scintillation spectrometer was used and the pulses displayed and photographed on the screen of the C.R.T. kicksorter described in Chapter IX. The background due to cosmic rays and internal radioactive impurities decreases as the dimensions of the NaI crystal are reduced. Increasing the dimensions of the crystal, the detection efficiency approaches a constant value asymptotically. There is therefore an optimum size for the best ratio of signal to background. This was found to be about 0.75" diameter and 0.75" long.

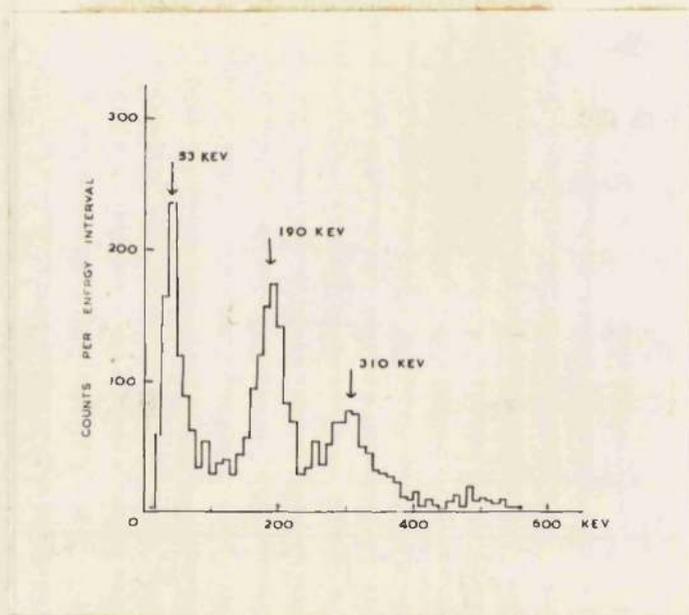


Fig. 25

γ -ray spectrum from Iu^{176}
obtained with a scintillation
counter (NaI)

To avoid uncertainties in the position of the crystal and in attenuation of the incident γ -rays, thin aluminium foil was used as a reflector instead of MgO, and the crystal and reflector mounted in a thin perspex container. Provision was made in the latter for accurate positioning of the γ -ray source. The latter was in the form of Lu_2O_3 (.49 mg/cm²) in a thin aluminium capsule. This aluminium, together with the perspex, was just sufficient to stop the most energetic β -rays from the source. The crystal and photo-multiplier were shielded by 8" of lead on all sides, which reduced the background counting rate from about 2000 c.p.m. to 25 c.p.m. The spectrometer was calibrated with the 280-kev γ -rays and k x-rays from Hg^{203} .

The γ -ray spectrum of Lu^{176} obtained is shown in fig. 25. From this, γ -ray peaks at 310 ± 10 -kev and 190 ± 10 -kev stand out clearly. The x-ray peak at 53-kev is thought to arise principally from the absorption of the γ -rays in the thick source.

The writer computed the detection efficiency of the crystal for γ -rays of the above energies, allowing for attenuation in the source, aluminium, and perspex. From this the intensity of the γ -rays was found. From the proportional counter data of Dixon et al., the internal conversion coefficient was estimated. When this was taken into account, it appeared that both the high energy γ -rays had the same intensity, which agreed well with the number of β -rays, thus supporting the decay scheme of Goldhaber and Hill. *

The agreement between experimental measurement and theoretical

prediction of the energy levels is shown in the table below.

Experimental	89-kev	279 ± 10 -kev	589 ± 20 -kev
Theoretical	(89-kev)	296-kev	623-kev

- * The I.C. coefficients are also in agreement with the multipole orders of transition which the scheme suggests.

Appendix II

The Circuits of the Fast Coincidence Unit.

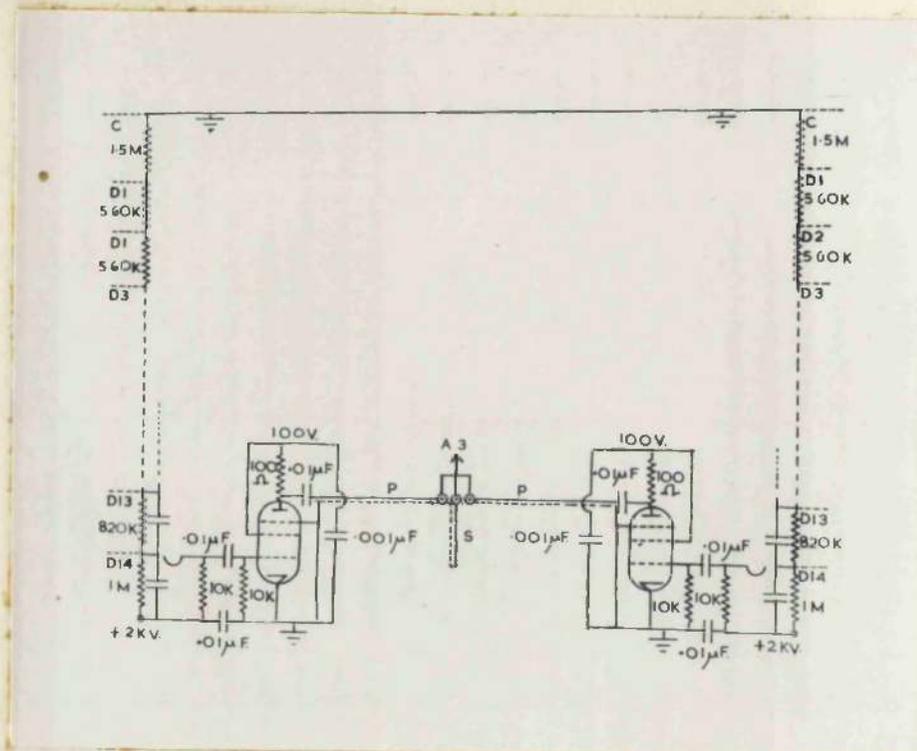


Fig. 26

Fast Coincidence Circuitry

- C - Photo-cathode of photomultiplier type 6262
- D1 - 14 - Dynodes of photomultiplier
- Decoupling condensers on resistance chain are 50 P.F.
- Valves are type CV. 133
- P - Uniradio 31, 95 coaxial cable
- S - Shorted line 45 coaxial cable

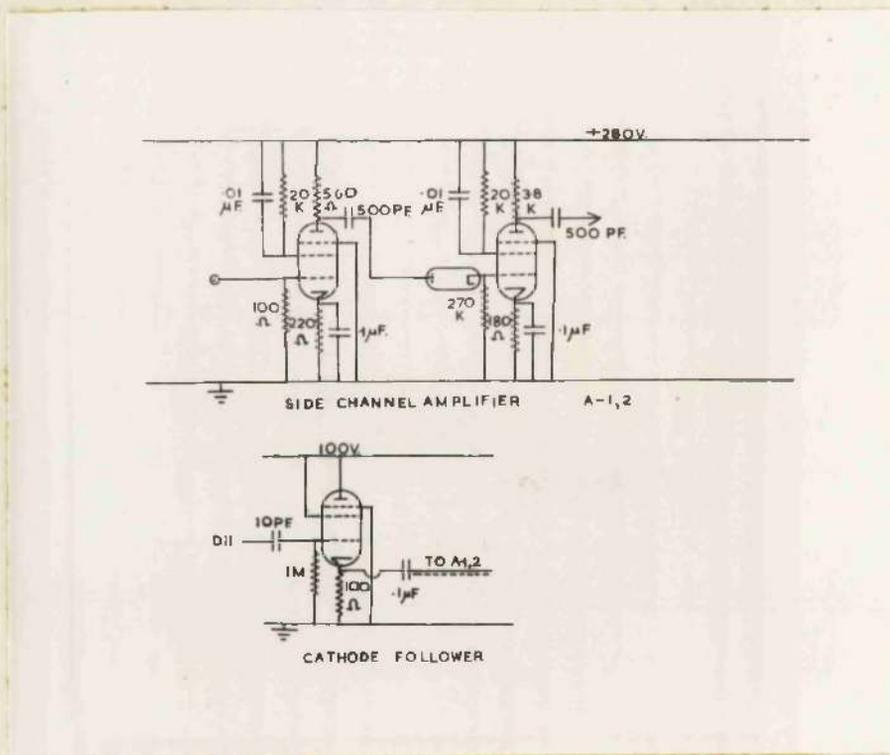


Fig. 27

Cathode follower and side channel
 amplifier A-1, 2
 Cathode follower valve is type
 CV.138
 Valves in A-1, 2 are CV.1136

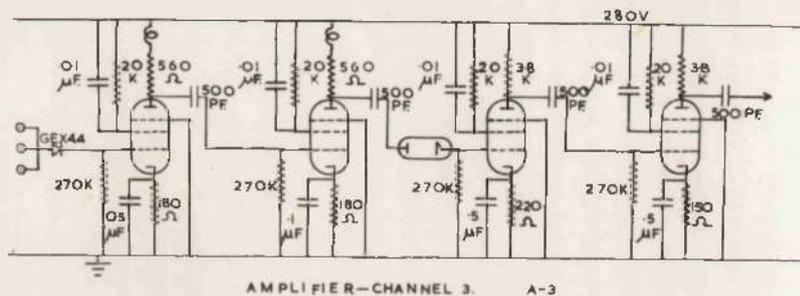


Fig. 26

Amplifier A. 3

Valves are type CV.1136

Chokes are made up of 15 turns on
a former 2 cm diameter and 2 cm long.

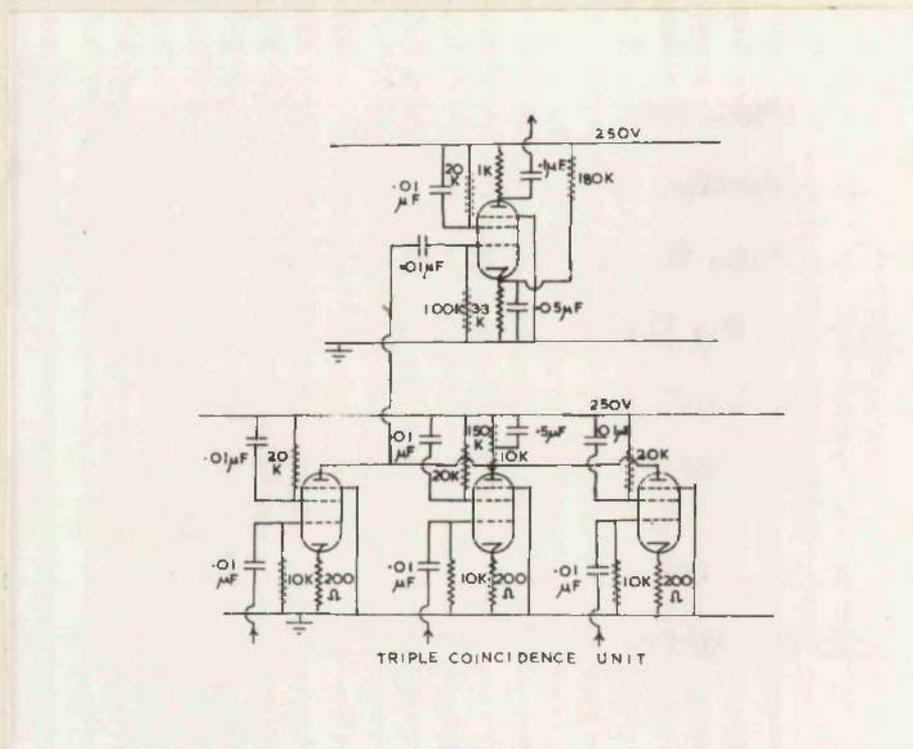


Fig. 29

Triple coincidence unit
Valves are type CV.1136

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