THE UNIVERSITY OF BRITISH COLUMBIA
FACULTY OF GRADUATE STUDIES

PROGRAMME OF THE
FINAL ORAL EXAMINATION FOR THE DEGREE
OF DOCTOR OF PHILOSOPHY

of

ROBERT MICHAEL PEARCE
B.Sc. (McGill) 1947
M.A. (British Columbia) 1949

THURSDAY, JUNE 12th, 1952, at 2:30 P.M.
IN ROOM 301, PHYSICS BUILDING

COMMITTEE IN CHARGE:
Dean W. H. Gage, Chairman
Professor F. A. Kaempffer  Professor H. Adaskin
Professor K. C. Mann  Professor D. Derry
Professor W. Opechowski  Professor B. Savery
Professor J. B. Warren  Professor F. Noakes
GRADUATE STUDIES

Field of Study: Physics

- Nuclear Physics—Professor K. C. Mann
- Quantum Mechanics—Professor G. M. Volkoff
- Special Relativity—Professor W. Opechowski
- General Relativity—Professor M. Wyman
- Electronics—Professor A. van der Ziel
- Chemical Physics—Professor A. J. Dekker
- Quantum Theory of Radiation—Professor F. A. Kaempffer
- Spectroscopy—Professor A. M. Crooker
- Cosmic Rays—Professor J. B. Warren
- Theory of Measurements—Professor A. M. Crooker
- Electromagnetic Theory—Professor G. L. Pickard

Other Studies:

- Differential Equations—Professor T. E. Hull
- Group Theory—Professor D. C. Murdoch
- Topics in Applied Mathematics—Professor E. Leimanis
THESIS

I

THE SUPPRESSION OF COMPTON ELECTRONS IN SOME PHOTOELECTRON SPECTRA

A new method has been used to suppress the undesirable Compton electrons ordinarily present in photoelectron spectra. This is accomplished by electronic cancellation of the individual Compton electron counts. The new method has been used with a thin-lens type of spectrometer, and has made possible the detection of new gamma rays in Ra (B + C), Ta$^{182}$, and Sn$^{124}$. No new gamma rays were found in Co$^{60}$.

II

THE DOUBLE BETA DECAY OF Sn$^{124}$

A research has been made for double beta decay in Sn$^{124}$ using an energy dependent coincidence technique particularly suited to the detection of double beta events according to Majorana’s neutrino theory. No events attributable to double beta decay were found. From this result, an upper limit of $0.3 \times 10^{17}$ years was set on the half-life for the process.
PUBLISHED PAPERS


I
THE SUPPRESSION OF COMPTON ELECTRONS
IN SOME PHOTOELECTRON SPECTRA

II
THE DOUBLE BETA DECAY OF Sn\textsuperscript{124}

by

ROBERT MICHAEL PEARCE

A THESIS SUBMITTED IN PARTIAL FULFILLMENT OF
THE REQUIREMENTS FOR THE DEGREE OF
DOCTOR OF PHILOSOPHY
in
PHYSICS

We accept this thesis as conforming to the standard required from candidates for the degree of DOCTOR OF PHILOSOPHY.

Members of the Department of Physics

THE UNIVERSITY OF BRITISH COLUMBIA
MAY, 1952
ABSTRACT

PART I

A new method has been used to suppress the undesirable Compton electrons ordinarily present in photoelectron spectra. As much as 90% of the Compton electron intensity was removed. This was accomplished by electronic cancellation of the individual Compton electrons. The method has been used with a thin lens type of spectrometer and has made possible the detection of new gamma rays at .391, .857 and 1.00 Mev. in Ra(B+C), at 1.01 Mev. in Ta_{182}, and at .472 and .843 Mev. in Sb_{124}. No new gamma rays were observed from Co_{60}. 
ABSTRACT

PART 2

A search for double beta decay in Sn$^{124}$ has been made using a coincidence technique particularly suited to double beta decay under the Majorana form of neutrino theory. Negative results were obtained and a lower limit of $0.3 - 0.7 \times 10^{17}$ years has been set on the half-life of the process.
TABLE OF CONTENTS

PART 1

THE SUPPRESSION OF BACKGROUND IN
SOME PHOTOELECTRON SPECTRA

I INTRODUCTION

II EXPERIMENTAL PROCEDURE
A. The Detector Counter
B. The Source Counter
C. The Anticoincidence Circuits
D. The Suppression of Compton Electron Counts as
   a Function of Energy
E. Loss of Photopeak Intensity

III RESULTS OF THE GAMMA RAY STUDIES
A. Ra (B + C)
B. Ta$^{182}$
C. Co$^{60}$
D. Sb$^{124}$

IV SOME ASSOCIATED STUDIES IN Ta$^{182}$
A. The Beta Spectrum of Ta$^{182}$
B. Gamma-Gamma Coincidences in Ta$^{182}$

V CONCLUSIONS

APPENDICES

2. Internal Reflection in Lucite.
PART 2
ON THE DOUBLE BETA DECAY OF Sn^{124}

I  INTRODUCTION

II  SOME PREVIOUS WORK
   A. The Triplet \( \text{Sn}^{124} \rightarrow \text{Sb}^{124} \rightarrow \text{Te}^{124} \)
   B. The Triplet \( \text{Pa}^{110} \rightarrow \text{Ag}^{110} \rightarrow \text{Cd}^{110} \)
   C. The Triplet \( \text{Te}^{130} \rightarrow \text{I}^{130} \rightarrow \text{Xe}^{130} \)
   D. The Triplet \( \text{U}^{238} \rightarrow \text{Np}^{238} \rightarrow \text{Pu}^{238} \)

III  EXPERIMENTAL PROCEDURE
   A. The Principle of the Experiment
   B. The Experimental Arrangement
   C. Calibration of the Kicksorter
   D. The Optimum Source Thickness
   E. Obtaining the Data
   F. Shape of the Background Spectrum
   G. Comparison with Previous Work

IV  RESULTS AND CONCLUSIONS
TABLE OF ILLUSTRATIONS

FIG. 1 SCHEMATIC DIAGRAM OF THIN LENS SPECTROMETER.
FIG. 2 GENERAL SHAPE OF A PHOTOELECTRON SPECTRUM.
FIG. 3 SCHEMATIC DIAGRAM OF CONVENTIONAL SOURCE - HOLDER.
FIG. 4 INTEGRAL BIAS CURVES AT VARIOUS ELECTRON ENERGIES.
FIG. 5 SCHEMATIC DIAGRAM OF SOURCE - HOLDER USED WITH COMPTON SUPPRESSION.
FIG. 6 SOURCE - HOLDER & ANTICOINCIDENCE PHOTOMULTIPLIER.
FIG. 7 BLOCK DIAGRAM OF ANTICOINCIDENCE ARRANGEMENT.
FIG. 8 DETECTOR HEAD AMPLIFIER.
FIG. 9 SOURCE HEAD AMPLIFIER.
FIG. 10 FEEDBACK AMPLIFIER USED IN THE DETECTOR CHANNEL.
FIG. 11 DISCRIMINATOR AND DIFFERENCE AMPLIFIER.
FIG. 12 SOURCE COUNTER AMPLIFIER.
FIG. 13 CANCELLATION AS A FUNCTION OF ENERGY FOR Ta & Ra.
FIG. 14 PHOTOELECTRON SPECTRA FROM GAMMA RAYS Ra(B+C).
FIG. 15 PHOTOELECTRON SPECTRA FROM GAMMA RAYS Ta$^{182}$.
FIG. 16 PHOTOELECTRON SPECTRA FROM GAMMA RAYS Co$^{60}$.
FIG. 17 PHOTOELECTRON SPECTRA FROM GAMMA RAYS Sb$^{124}$.
FIG. 18 KURIE PLOT OF THE Ta$^{182}$ BETA GROUP.
FIG. 19 COINCIDENCE MIXER.
FIG. 20 THE MASSES OF AN ISOBARIC TRIPLET.
FIG. 21 BLOCK DIAGRAM OF COINCIDENCE ARRANGEMENT.
FIG. 22 COINCIDENCE SPECTRA OBTAINED ON KICKSORTER.
FIG. 23 DIFFERENCE BETWEEN COINCIDENCE SPECTRA SHOWN IN FIG. 22.
ACKNOWLEDGEMENTS

The work described in this thesis was supported by a Grant-in-Aid-of-Research allotted to Dr. K.C. Mann by the National Research Council of Canada.

I am indebted to Dr. Mann for invaluable suggestions and discussions during the course of the research.

National Research Council awards to the author made possible the continued effort from 1949 to 1952.

The work described in Part II of the thesis was done in collaboration with Dr. E.K. Darby.
PART 1

THE SUPPRESSION OF BACKGROUND IN SOME PHOTOELECTRON SPECTRA
I

INTRODUCTION

It is to be expected that a knowledge of the energy levels of radioactive nuclei will provide the means of testing theories of nuclear forces, since any successful theory must predict energy level sequences. For this reason, the "decay schemes" of radioactive nuclei have been the subject of considerable research.\textsuperscript{1,2} By "decay scheme" is meant a complete determination of the nuclear levels as to energy, angular momentum (spin), and parity together with transitions between these levels. In most cases a knowledge of the energy levels can come only from a study of the radiations from the nucleus, which result from transitions from one state to another. The decay of a nucleus by primary electron emission to the daughter nucleus is usually followed by the de-excitation of the daughter nucleus by gamma rays or internal conversion electrons. It is customary in these investigations to measure the intensities and energies of all detectable gamma rays. This may make possible the assignment of a self-consistent decay scheme. However it may be possible to postulate more than one sequence of energy levels which would lead to gamma-ray transitions consistent with the experimental evidence. Because of experimental difficulties, it is highly probable that low intensity gamma rays which are actually present have escaped detection. A knowledge of these would undoubtedly make easier the choice of a unique decay scheme. For this reason, an apparatus has been designed and constructed in this laboratory to increase the probability of detection of low intensity gamma-rays.
Evaluated tube
Baffles
Detector
Source

Current in coil
focuses one energy.

**FIG. 1**

SCHEMATIC DIAGRAM OF THIN LENS SPECTROMETER.

**FIG. 2**

GENERAL SHAPE OF A PHOTOELECTRON SPECTRUM.
The shape desired is shown below.
Gamma-ray energies are usually measured by the photo-electric technique, whereby photoelectrons are ejected from thin lamina by the gamma-rays. Such a lamina is called a "radiator" and usually consists of some high atomic number material, since the probability of photo-electron emission increases rapidly with the atomic number Z. In this method, a beta-ray spectrometer is used to study the photoelectrons.

The thin-lens type of spectrometer used in this work has been described in detail elsewhere.\textsuperscript{3,4,5} Its operation depends upon the focussing of electrons by the magnetic field due to a current in a large iron-free coil. The electrons pass down an evacuated tube which lies on the axis of the coil and concentric with it (see Fig. 1). Baffles are arranged within the tube to define a path for the electrons. The momentum of the electrons so focussed at the detector is determined by the coil current. In this way the intensity of focussed electrons is studied as a function of coil current or momentum and a spectrum is obtained.

If photoelectrons are focussed at an energy $E$ by the spectrometer, the energy $h\nu$ of the gamma ray is given by

$$h\nu = E_k + E,$$

where $h$ is Planck's constant,
\nu is the gamma-ray frequency,
and $E_k$ is the K-shell binding energy of the radiator.

In favourable cases photoelectrons from the L-shell of the radiator may be detected, although the cross-section for this process is smaller. The L-photopeaks are found at slightly higher energies than the corresponding K-photopeaks because of the smaller L-shell binding energy.
Fig. 3
Schematic Diagram of Conventional Source - Holder
For an electron describing a circular path of radius $\rho$, the momentum in a magnetic field $H$ is given by

$$p = \frac{e}{c} H \rho.$$ 

Thus $p$ is directly proportional to $H \rho$ in gauss-cm and therefore to $H$, since $\rho$ is a constant of the spectrometer. Furthermore $H$ varies directly with the magnet current, $I$, since no iron is present, with the result that the momentum varies linearly with the magnet current. Thus singlepoint calibration of the momentum in terms of coil current is possible and some well known line is measured to calibrate the instrument. Conversion from momentum $p$ to the energy, $E_\beta$, may be easily shown to follow from

$$\frac{c}{e} p = H \rho = \frac{10^4}{3} \sqrt{E_\beta (E_\beta + 1.02)},$$

where $H \rho$ is in gauss cm, and $E_\beta$ is in Mev.

In actual practice, such a conversion is made easier by published tables which give $p$, $E$, and $H \rho$ for a large spread of energies.

In the conventional photoelectric technique, the radiator is placed close to the source of gamma rays. The primary beta particles are found to be much more intense than the photoelectrons produced in the thin radiator and must be removed to make the detection of photoelectrons feasible. The primaries are therefore absorbed in a low atomic number ($Z$) material placed between source and radiator, (see Fig. 3). A low $Z$ absorber is chosen because the photoelectric cross-section varies as $Z^5$, and only the radiator is desirable as a photoelectron source.
Unfortunately, many gamma-rays suffer Compton encounters in this absorber, and some of the resulting Compton electrons are focussed and recorded. The distribution from a single gamma-ray appears as a smooth, almost symmetrical, spectrum from zero energy to a maximum which approaches
\[ h\nu' \approx 0.255 \text{ Mev.} \]
for \[ h\nu \gg 0.51 \text{ Mev.} \]

The Compton electron distribution is much less intense than the primary beta-ray spectrum, but is of the same order of magnitude as the photoelectron line intensity. The photoelectrons appear in the spectrum as peaks on the smooth Compton electron distribution as shown in the upper curve of Fig. 2.

Because of this background, a peak and its immediate vicinity has to be more carefully measured than if no background were present. Detection of a peak becomes possible only when the Compton distribution intensity has been measured with an uncertainty which is less than the intensity of the peak. Since the spectrum is obtained by the recording of random events, the uncertainty of the intensity can be made small only by recording a large number of events. Such a process is time consuming, and instrumental instability imposes a limit on the accuracy possible. In the case of short lived radiations, long counting times are not available.

To improve this situation, an apparatus designed to suppress this Compton background has been constructed for use in a thin-lens spectrometer. It consists, in part, of a counter at the source end of the spectrometer which detects Compton electrons. This counter is arranged in anticoincidence with the counter at the detector end where the particles are focussed and recorded. Thus
a focussed beta particle is not recorded when a simultaneous event is observed in the source end counter. That is to say, a Compton electron produces a coincident pulse in the counters at each end of the spectrometer, and a circuit is arranged to reject this event.

The ideal effect of this procedure upon a typical photoelectron spectrum is shown in Fig. 2. The "signal-to-noise ratio" has been improved, and photopeak detection has been made easier.

Part 1 of this thesis describes the operation of the apparatus and reports on measurements obtained on Ra (B + C), Ta\(^{182}\), Co\(^{60}\) and Sb\(^{124}\). In addition, other measurements taken on Ta\(^{182}\) are reported.

It is to be expected that with a better method of detection of weak gamma-rays, the uncertainties in the assignment of decay schemes should be reduced. However, even then uniqueness is not always possible in the assignment and other supporting evidence must be utilized. Coincidence measurements to determine which gamma-rays are in cascade may clarify the situation. Such experiments have been performed with Ta\(^{182}\). The beta spectrum of Ta\(^{182}\) has also been measured.
II

EXPERIMENTAL PROCEDURE

A. The Detector Counter

For this work on Compton suppression, the spectrometer was changed over from Geiger counters to scintillation counters. This change was necessary because a rapid response was needed in the mixing circuit, (see Section II C.)

Beta particles focussed in the spectrometer are detected by a type RCA 5819 photomultiplier used with an anthracene crystal. The magnetic field of the spectrometer is as large as 50 gauss at the photomultiplier position and was found to defocus the electrons in the multiplier tube. To shield the photomultiplier from this field, it was housed in a mild steel tube. Tests showed that this arrangement provided adequate shielding and did not appear to interfere with the focussing action of the spectrometer. Endplates to the tube were made "light-tight" by thin neoprene gaskets. One endplate carries the head amplifier and cable connections. The other endplate forms the vacuum seal to the detector end of the spectrometer. Focussed electrons pass through a hole in the endplate and strike the detector crystal. This consisted of a flake of anthracene mounted on lucite. The photocathode of the multiplier tube is held against the lucite and adequate optical coupling is ensured by a layer of transparent jelly between the lucite and glass surfaces. Without this precaution, internal reflections of the scintillations occur at the lucite-air and at the glass interfaces, (see Appendix 2).
FIG. 4
INTEGRAL BIAS CURVES TAKEN AT VARIOUS ELECTRON ENERGIES.
Counts from cosmic rays and local contamination are kept to a minimum by using an anthracene crystal of small volume. A crystal 0.4 mm. in thickness and approximately 7 mm. in diameter is used. It was found to be 100% efficient for beta particles, while having a cosmic ray background of three counts per minute. The efficiency was determined by comparing the efficiency of the crystal with that of a geiger counter with a collimated beta source of RaE. The low background compared favourably with the usual background of, say, 25 counts per minute in a geiger tube.

The evaporation in vacuo of the anthracene is such that the crystal had to be replaced after six weeks operation.

The counter performs satisfactorily in the energy region above 150 Kev. At low energies, the pulse height is no larger than thermal noise in the multiplier. However, the usefulness of the spectrometer itself does not extend to energies much below this limit, due to scattering in the long electron path, absorption in the source and from other causes.

The proportionality of pulse height to electron energy can be seen in Fig. 4. The separate curves are integral bias curves from the scintillation counter taken with the spectrometer set to focus electrons at the different energies indicated. The pulse size does not increase above 850 Kev. in this particular case since the electrons then pass completely through the crystal.

B. The Source Counter

In order that the Compton electrons be suppressed and the photoelectrons be unsuppressed, the counter at the source end must count Compton electrons but not photoelectrons, as was explained in the Introduction. This was arranged in the manner
Fig. 5
Schematic diagram of source-holder used with Compton suppression.
FIG. 6

SOURCE - HOLDER & ANTICOINCIDENCE PHOTOMULTIPLIER
shown schematically in Fig. 5. The anthracene crystal is placed between the low $Z$ absorber and the radiator so that all Comptons accepted by the spectrometer must pass through it. Photoelectrons on the other hand, originate beyond the crystal and hence leave no record in the source counter. Thus Comptons produce anti-coincidence pulses while photoelectrons do not.

Fig. 6 shows how the arrangement described above was physically realized. The radiator and anthracene crystal are mounted with Canada Balsam on the end of a lucite rod which protrudes into the spectrometer vacuum system. The rod also serves to hold the source and to provide optical coupling from the crystal to the photomultiplier. The source is contained in a small hole drilled along a diameter of the lucite rod close to the crystal. The rod is fixed to the spectrometer end-plate with Apiezon wax to form a vacuum seal. The glass envelope of the photomultiplier is pressed against the end of the rod away from the vacuum system. The lucite rod is polished in order to provide an optical path for the scintillations in the lucite—making use of internal reflections off the walls of the rod. Unfortunately, the opaque body of the source was found to interfere somewhat with the optical path. This difficulty was overcome by putting sloping shoulders on the lucite (see Fig. 6) which reflected scintillations around the source. A transparent jelly is kept between the lucite and the glass envelope of the photomultiplier to ensure efficient light transfer.

Enough lucite is left between source and crystal to absorb the primary betas. If this is not done, the counting rate in the crystal becomes excessive because of the high intensity of the primary betas. Furthermore, many of the photoelectrons would be cancelled by truly coincident primary beta particles.
FIG. 7
BLOCK DIAGRAM OF ANTICOINCIDENCE ARRANGEMENT
same argument it might be expected that some photoelectrons would be lost because of truly coincident gamma rays. However, the gamma-ray efficiency of the crystal (intrinsic + geometrical) was estimated to be much less than 0.1 percent, so that this effect may be ignored.

As in the case of the detector photomultiplier, a mild steel cylinder and end plates protected the tube from the spectrometer field.

C. The Anticoincidence Circuits

The next step was to use the pulses from the source counter to cancel coincident detector pulses. The conventional method of doing this would be to feed the output pulses from the two counters through amplitude "discriminators" to reject thermal noise pulses. A mixing stage would follow, and would be so arranged that pulses from the detector counter would be blocked when in coincidence with source counter pulses. However, a difficulty arose due to the proximity of the source to the crystal (see Fig. 6). Although the gamma ray efficiency of the crystal is very low, as was pointed out above, the favourable geometry produces a very high gamma counting rate in the anticoincidence channel. This was estimated to be $2 \times 10^5$ counts per second for a typical source, $\frac{1}{2} \text{mc Ra(B + C)}$. This rate is of the order of $10^5$ times larger than the rate in the detector counter. Since a conventional discriminator remains insensitive for several microseconds after passing a pulse, such a discriminator could not be used in the anticoincidence channel without serious counting losses.

Hence it was necessary to adopt the unconventional arrangement shown in the block diagram in Fig. 7. The principle
FIG. 8
DETECTOR HEAD AMPLIFIER

FIG. 9
SOURCE HEAD AMPLIFIER

OUTPUT INTO 100 Ω COAX.

OUTPUT INTO 1000 Ω COAX.

Type RG 65/U
Delay 0.4 µs/ft.
FIG. 10

FED-BACK AMPLIFIER USED IN THE DETECTOR CHANNEL.
FIG. II

DISCRIMINATOR AND DIFFERENCE AMPLIFIER.

6AG5  6J6  6AK5  6AG5  6AK5  6AG5  6AG5
1/2  6AL5

DISCRIMINATOR AND DIFFERENCE AMPLIFIER.
of operation is as follows: the detector pulses are shaped by a discriminator and brought to the difference amplifier. The many source counter pulses are amplified and their whole spectrum subtracted from the shaped detector pulses. Any detector pulse which loses amplitude in this subtraction is rejected by a second discriminator. Thus the anticoincidence circuit is arranged in such a manner that neither of the two discriminators run faster than does the slower channel.

A more detailed description of the separate circuit follows. The circuit diagram of the detector head amplifier is shown in Fig. 10. The latter is a feed-back amplifier with an observed rise-time of 0.1 $\mu$ sec. and a maximum output of 120 volts. It is provided with an input attenuator. The pulses are led from the output cathode follower to the discriminator shown on the left of Fig. 11. The shaped pulses from the discriminator then go to the difference amplifier also shown in Fig. 11. The spectrum of source pulses is also fed into this stage where they are subtracted from the discriminator pulses (from the detector counter). Those output pulses from the difference amplifier which retain their full amplitude are then accepted by the discriminator of a commercial scaling unit and recorded.

The pulses in the source channel are delayed 0.1 $\mu$ sec. (see Fig. 7) to allow the discriminator of the detector channel to fire. This delay is provided by three feet of RG 65/U cable. The head amplifier of the source channel is shown in Fig. 9. The plate load matches the 1000 ohm characteristic impedance of the delay cable in order to stop reflections in the cable.

The source or anticoincidence amplifier is shown in
FIG. 12
SOURCE COUNTER AMPLIFIER
CANCELLATION AS A FUNCTION OF ENERGY FOR Ta & Ra.

Fig. 13

CANCELLATION AS A FUNCTION OF ENERGY FOR Ta & Ra.
Fig. 12. It has a gain of 50 which is necessary in that some of the useful pulses are small (see section "D"). All pulses are limited to 2 volts by cutting off the final tube. In this way the difference amplifier which follows can not be overloaded. To stop the source amplifier itself from blocking, crystal diodes are arranged as shown in Fig. 12. The rise time of the source amplifier is 0.04 \( \mu \) sec., and the insensitive time after receiving a pulse as large as 1 volt was estimated to be 0.2 \( \mu \)s.

D. Suppression of the Compton Electron Counts as a Function of Energy.

Suppression of the Compton electron counts is not 100% effective over the spectrum. The percentage of Compton counts not cancelled is shown as a function of momentum for \( \text{Ta}^{182} \) and \( \text{Ra}(B + C) \) in Fig. 13. Suppression efficiencies are as high as 90% in the central region of maximum Compton intensity where cancellation is most needed. However, the suppression is poor in the high and low momentum regions and some explanation should be given for the energy dependence of the suppression.

Obviously, the most energetic Comptons in a spectrum can have lost no energy to the source crystal by the very fact that they retain their full possible energy. If no energy has been lost to the source crystal, no cancelling pulse can have been produced. Thus the most energetic Comptons cannot be cancelled in principle. A certain minimum energy loss in the crystal is necessary to produce a cancelling pulse, depending on the sensitivity of the source counter. The magnitude of this minimum energy loss determines how far the poor cancellation extends down from
the top of the spectrum. In this investigation the source counter can detect a minimum energy of about 50 Kev., the limit to the sensitivity being thermal noise in the photomultiplier. So the top 50 Kev. of the spectrum is not cancelled in this case.

Actually some Comptons as much as 150 Kev. below the maximum energy are not cancelled, and an extension of the above argument may be used to explain this.

Let us consider the Compton distribution from a single gamma transition of 1.2 Mev. It may be said that the most energetic Comptons arise from encounters in the outermost layer of the source holder which consists of the radiator and a portion of the anthracene crystal. In fact the top 50 Kev. of the spectrum must come from a layer of thickness 30 mg/cm$^2$ since the rate of energy loss, $\frac{dE}{dx}$ is 1.7 Kev/mg/cm$^2$ at 1 Mev.$^{11}$ Compton encounters in this layer will not provide cancelling pulses of sufficient magnitude. To explain the changes in cancellation efficiency over the spectrum by the concept of this insensitive layer, we must consider the various angles, $\theta$, which occur between the direction of the incident gamma and the scattered electron. The energy of a scattered electron is given$^{12}$ by

$$E(\theta) = \frac{2 m_0 c^2 r^2 \cos^2 \theta}{1 + 2 r + r^2 \sin^2 \theta},$$

where $r = \frac{\hbar \nu}{m_0 c^2}$.

We have already dealt with the head-on ($\theta = 0$) encounters in the last layer. Comptons from $\theta = 10^0$ encounters in the last layer have a good chance of acceptance by the spectrometer baffles. Since $E(10^0)$ is about 100 Kev. below the
maximum, $E(0)$, this means that some of the electrons at 150 kev. below the maximum of the spectrum come from the last layer and are therefore not cancelled.

The insensitive-layer concept is also consistent with the good cancellation at intermediate energies. This can be understood by considering the origin of these electrons. They may come from deep within the sourceholder, in which case they will be detected in the anticoincidence crystal. They may not come from the last insensitive layer since the angle $\theta$ necessary in a collision giving rise to an intermediate energy electron is too large for the spectrometer baffles to accept the electron.

Thus a qualitative picture built on the one assumption of the origin of the uncancelled electron explains the shape obtained in the intermediate and upper regions of the spectrum. There is also the possibility that the scintillations from the high energy Comptons are not properly optically coupled to the photocathode because of the opaque body of the source. This might be the case since the high energy Comptons come from head-on collisions which tend to occur, therefore, in the plane of the source hole and the spectrometer axis. To test this explanation of the poor suppression at high energies, a seemingly perfect optical system was constructed. The source was mounted in the lucite rod by a pressure mold, thus abolishing the source hole. This was done at $145^\circ$C. at a pressure of 3000 lbs. per sq. in. The source used was $\text{Co}^{60}$ wire of 0.01 inch diameter in the form of an open lattice located at the source position. The source was believed small enough so as not to interfere with the scintillations. However, the behaviour of this arrangement was identical to that of holders having large holes to carry the
source. The latter type of source holder is therefore to be preferred since the molding process is time consuming.

To quantitatively account for the shape of the curve, it would be necessary to determine the origin of the electrons in detail for all energies. This is impossible because of such complications as the large range of possible collision angles, the presence of multiple scattering, and the different transmission of the spectrometer for different points on the source.

A tentative explanation may be given for the poor cancellation at low Compton energies (see Fig. 13), the explanation being consistent with the insensitive layer concept used above in the high energy region. In the case of a single 1.2 Mev. gamma-ray it was stated that an intermediate energy electron could not arise from a large angle encounter in the very last layer of the crystal, because the baffles would not accept it. There is a chance of its being accepted, however, if it is multiply scattered in the last layer of the source holder. Furthermore, this chance of acceptance increases for low energy electrons since the root mean square angle of scattering is proportional to \( \frac{113}{E} \). Hence low energy electrons have a better chance of originating in the insensitive layer and being focussed than do Comptons of intermediate energy.

Thus the main features of the \( \text{Tal}^{182} \) cancellation curve in Fig. 13, which resembles that of the hypothetical 1.2 Mev. gamma-ray, have been explained. Finally, we must pass to the case of \( \text{Ra}(B + C) \) with its many lines (see Table I) extending to 2.4 Mev. At 1 Mev. in the Ra spectrum the preponderance of Compton electrons comes from the highest energy gamma rays. So the poor cancellation of the Comptons from lines whose energies are just above 1 Mev. is
not noticed. For this reason the form of the cancellation curve of Ra resembles that of $^{182}$Ta (see Fig. 13), except of course for a shift in energy.

E. Loss of Photopeak Intensity

The use of the anticoincidence method results in a small loss of photopeak intensity. Almost all of this loss is due to chance coincidences, i.e. a photoelectron is focussed but not recorded because of an accidentally simultaneous event in the source counter. With a gamma-ray source of $\frac{1}{2}$ mc. strength, the photopeak loss was measured to be 5%. An almost negligible loss of photoelectrons results from events in the source counter which are truly coincident with the photoelectrons, i.e. Compton events from gamma-rays which are in cascade with the gamma-ray causing the photoelectron. This loss is estimated to be much less than 0.1%, as was mentioned in Section IIB.

It might be thought that another cause of photopeak intensity loss would be the somewhat larger source-to-radiator distance necessitated by the presence of the source crystal. This distance is about 2.5 mm. instead of say 1.5 mm. in the conventional source holder without the crystal. However, the larger distance does not lower the photoelectron intensity since both the source and radiator are not points but are extended. Furthermore, the angular distribution of the photoelectron leans so far forward at the range of energies in question that the source-to-radiator distance tends to lose its importance, i.e. if the source touched the radiator, many electron trajectories would be at too large an angle to the spectrometer axis to be accepted by the baffles.
The loss of photoelectron intensity is considered in the statistical treatment of Appendix 1.
FIG. 14 PHOTOELECTRON SPECTRA FROM GAMMA RAYS Ra(D+C).
III

RESULTS OF THE GAMMA RAY STUDIES

A. Ra(B + C)

The photoelectron spectrum of a 0.5 mc. source of Ra(B + C) with Compton electrons suppressed is shown in the lower curve of Fig. 14. The upper curve is a conventional spectrum taken previously in this laboratory also using a thin-lens spectrometer. The radiator used in the present work was a uranium foil of 24 mg/cm² thickness. In the earlier work, a lead foil of 40 mg/cm² thickness was used.

The energies of the gamma rays detected are listed in Table 1, together with the results of Mann and Ozeroff, of Latyshev and co-workers, and of Ellis. New lines have been found at 0.391, 0.857 and 1.00 Mev. In addition, the lines at 0.450 and 0.781 Mev. reported by Mann have been confirmed. A line has been detected at 1.55 Mev. which may be the unconfirmed line reported to be at 1.52 Mev. by Latyshev.

Substantial agreement is found between the separate investigators on the majority of lines.

No photoelectrons were found for the line listed at 2.41 Mev. This line is calculated from the Compton endpoint of 2.15 Mev.

B. Ta

The gamma ray spectrum of a 1 mc. source of Ta with Compton suppression is shown in the lower curve of Fig. 15. The upper curve is the spectrum taken in the conventional manner in the same geometry, i.e., with the anticoincidence counter turned off.
TABLE 1

GAMMA RAYS OF RADIUM B+C IN Mev.

<table>
<thead>
<tr>
<th>Present Investigation</th>
<th>Ellis(^{17})</th>
<th>Latyschev(^{16})</th>
<th>Mann(^{15})</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.00</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1.10</td>
<td>1.12</td>
<td>1.120</td>
<td>1.12</td>
</tr>
<tr>
<td>1.22</td>
<td></td>
<td>1.21</td>
<td>1.22</td>
</tr>
<tr>
<td>-</td>
<td>1.238</td>
<td>1.234</td>
<td></td>
</tr>
<tr>
<td>1.37</td>
<td>1.379</td>
<td>1.370</td>
<td></td>
</tr>
<tr>
<td>-</td>
<td></td>
<td>1.390</td>
<td>1.40</td>
</tr>
<tr>
<td>1.55</td>
<td></td>
<td>1.52</td>
<td></td>
</tr>
<tr>
<td>-</td>
<td>1.62</td>
<td>1.75</td>
<td></td>
</tr>
<tr>
<td>2.15</td>
<td>2.193</td>
<td>2.20</td>
<td>2.21</td>
</tr>
<tr>
<td>2.41</td>
<td></td>
<td>2.40</td>
<td>2.40</td>
</tr>
</tbody>
</table>
FIG. 15 PHOTOELECTRON SPECTRA FROM GAMMA RAYS. HP (Gauss-cm.)

Conventional photoelectron spectrum

Spectrum taken with Compton electrons suppressed

Counts per minute

Lead Radiator

Uranium Radiator

1200 1697 2400 3396 4800 6787 H_\beta (Gauss-cm.)
Tantalum$^{182}$ was included in the present investigation because of considerable disagreement in reported energy measurements above 300 kev. O'Meara$^{18}$ has reported fourteen lines in the region from 300 kev to 1.1 mev which were not found by other investigators.$^{19,20,21}$ In this study, the three well known$^{19,20,21}$ lines at 1.23, 1.24 and 1.13 mev have been identified. In addition, a new line at 1.01 mev has been found. None of the lines reported by O'Meara are present.

The smooth prominence in the spectrum at approximately 1.0 mev results from the sudden loss of Compton suppression discussed in Section II,D. This occurs where the Compton distribution has the steepest slope and gives rise to a characteristic hump. This hump is quite different from the narrow, triangular form typical to a photopeak. Unfortunately, the new peak at 1.01 mev is superimposed on this prominence, (see Fig. 15). To make certain that the 1.01 mev peak was not spurious, a lead radiator was substituted for the uranium radiator. The peak in question shifted by exactly the difference in K-shell binding energy between lead and uranium.

C. Co$^{60}$.

The photoelectron spectrum of a 0.5 mc. source of Co$^{60}$ taken with and without Compton suppression is shown in Fig. 16. The same source-holder geometry was used in each spectrum. Only the two well-known$^{22,23}$ lines at 1.17 mev and 1.33 mev were found. No evidence appeared for any weak gamma transitions. A smooth hump occurs in the Co$^{60}$ spectrum as it did in the case of Ta$^{182}$ which has a similar high energy spectrum. It is interesting that
FIG. 17 PHOTOELECTRON SPECTRA FROM GAMMA RAYS $^{51}$Co.
FIG. 16 PHOTON SPECTRA FROM GAMMA RAYS Cc-60.
there is no hump in the upper part of the Ra(B + C) spectrum. Presumably this spectrum is the sum of the spectra from the many Ra gamma-rays. Each of these single spectra may itself have a hump, but the composite spectrum may well be smooth.

D. Sb\textsuperscript{124}.

Fig. 17 shows the spectrum of photoelectrons ejected from a uranium radiator by the gamma rays from 1 mc. of Sb\textsuperscript{124}. Only the spectrum with Compton electrons suppressed is shown, the ordinary spectrum being omitted for the sake of clarity. The percentage suppression is similar to that of the previous work, except that the gamma rays from Sb\textsuperscript{124} are so distributed in energy that smooth humps appear at both 2400 gauss-cm. and 5800 gauss-cm. A radiator of thickness 40 m/cm\textsuperscript{2} was used to take the spectrum above 4000 gauss-cm. and 24 m/cm\textsuperscript{2} was used below.

The gamma ray energies so obtained are given in Table 2 together with the values of some other workers. New gamma rays have been found at 0.472 Mev. and 0.843 Mev. There is some evidence for the existence of a gamma ray at 0.609 Mev., because of the large width of the photopeak from the 0.600 transition. The remaining energy values are in good agreement with those of previous investigators.
<table>
<thead>
<tr>
<th>Present Investigation</th>
<th>Kern et al\textsuperscript{24}</th>
<th>Cook et al\textsuperscript{25}</th>
<th>Iowa State\textsuperscript{26}</th>
</tr>
</thead>
<tbody>
<tr>
<td>.472</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>.600</td>
<td>.603</td>
<td>.608</td>
<td>.598</td>
</tr>
<tr>
<td>.609</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>.650</td>
<td>.650</td>
<td>.654</td>
<td>.645</td>
</tr>
<tr>
<td>.714</td>
<td>.714</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>-</td>
<td>-</td>
<td>.732</td>
<td>-</td>
</tr>
<tr>
<td>-</td>
<td>-</td>
<td>-</td>
<td>.817</td>
</tr>
<tr>
<td>.843</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>1.71</td>
<td>1.708</td>
<td>1.708</td>
<td>1.67</td>
</tr>
<tr>
<td>2.04</td>
<td>2.056</td>
<td>2.04</td>
<td>2.07</td>
</tr>
<tr>
<td>-</td>
<td>2.072</td>
<td>-</td>
<td>-</td>
</tr>
</tbody>
</table>
FIG. 18
KURIE PLOT OF THE Ta$^{182}$ BETA GROUP.
IV
SOME ASSOCIATED STUDIES IN Ta$^{182}$

A. The Beta Spectra of Ta$^{182}$

Comparison of observed beta spectra with theory is usually made with the aid of a Kurie plot. The function
\[ \left( \frac{N(p)}{Fp^2} \right)^{\frac{1}{2}} \]
is plotted against the energy \( E \), where \( N(p) \) is the intensity of electrons of momentum \( p \), and \( F \) is a factor to correct for the Coulomb field of the nucleus. From the Fermi relation\textsuperscript{27}
\[ \left( \frac{N(p)}{Fp^2} \right)^{\frac{1}{2}} = \frac{E_{\text{max}} - E}{E_{\text{max}}} \]
the plot should be a straight line intercepting the axis at \( E_{\text{max}} \).

The beta spectrum of Ta$^{182}$ was taken in a thin lens spectrometer with a source of thickness of $9 \text{ mg/cm}^2$. The Kurie plot for the spectrum obtained above 250 Kev. is shown in Fig. 18. A single beta group with an endpoint at $530 \pm 3$ Kev. can be seen. This endpoint is in agreement with the value of 525 Kev. given by Beach et al\textsuperscript{20}. Many internal conversion lines occur below 250 Kev., making a Kurie plot at low energies extremely difficult.

In this plot, the approximation to the Coulomb factor \( F \) which is valid for large \( Z \) was used. It is given by\textsuperscript{28}
\[ F = \frac{1}{p} - 0.355 \]
where \( p \) is in units of \( m_0c \).
FIG. 19
COINCIDENCE MIXER.
B. **Gamma-gamma Coincidences in Ta\textsuperscript{182}**

The gamma rays of Ta\textsuperscript{182} are seen to fall into two energy groups, one below 0.33 Mev. and one above 1.00 Mev. This made it possible to observe the high energy group by itself. This was done by using large anthracene crystals with photomultipliers and accepting only the largest pulses, which were presumably caused by the high energy gammas.

Approximately 10 microcuries of Ta\textsuperscript{182} was placed between the two anthracene crystals. The pulses from each of the photomultipliers were lead from a headamplifier identical to that shown in Fig. 8 to an amplifier shown in Fig. 10. The output from the two amplifiers went to the coincidence mixer shown in Fig. 19. The coincidence mixer had a resolving time of 0.136 usec. No genuine coincidences were observed between the high energy gamma rays of Ta\textsuperscript{182}.

It was possible to place an upper limit on the number of high energy gammas in cascade in the following way: coincidences were observed from the two gamma rays of a Co\textsuperscript{60} source in a similar geometry. The Co\textsuperscript{60} gamma rays at 1.17 and 1.33 Mev.\textsuperscript{4,22,23} are in cascade and have energies close to that of Ta\textsuperscript{182}. More than 100 times as many coincidences per gamma were observed in Co\textsuperscript{60} than in Ta\textsuperscript{182}. Thus less than 1% of the high energy gammas from Ta\textsuperscript{182} are in cascade. Presumably the three intense high energy gammas are three parallel modes of decay.
APPENDIX 1

AN ANALYSIS OF THE IMPROVED STATISTICAL ACCURACY

It has been mentioned above that not all the Compton electrons are suppressed and that the photoelectrons suffer a slight suppression by the use of the anticoincidence method. The question then arises as to whether the method makes any improvement in the sensitivity of photopeak detection. Obviously it might make no improvement in the case of a very large photopeak on a very small Compton background.

Let the events recorded in unit time with the conventional arrangement be \(N_T\), \(N_P\) and \(N_C\) for the total count, photoelectron count and Compton electron count respectively. Let the primed symbols refer to the counts obtained using the Compton suppression method.

Since the photoelectron count is obtained from
\[N_P = N_T - N_C,\]
we may write for the standard deviation\(^{29}\) of \(N_P\)
\[\sigma = \sqrt{\sigma_T^2 + \sigma_C^2},\]
where \(\sigma_T\) is the standard deviation of \(N_T\) and \(\sigma_C\) is the standard deviation of \(N_C\). Since \(\sigma_T = \sqrt{N_T}\) etc. for random events, we have
\[\sigma = \sqrt{N_T + N_C},\]
\[= \sqrt{2N_C + N_P}.\]

Taking the usual figure of merit\(^{30}\), \(\frac{\sigma}{N_P}\), we have
\[\frac{\sigma'}{N_P'} < \frac{\sigma}{N_P},\]
as the condition that the anticoincidence device represents an
improvement over conventional operation.

Or

$$\sqrt{\frac{2N'_C + N'_p}{N'_p}} < \sqrt{\frac{2C + N_p}{N_p}}.$$  \hspace{1cm} (1)

Let us define the ratios of the new to the old counting rates for photoelectrons and Comptons respectively by

$$p = \frac{N'_p}{N_p}, \hspace{1cm} (2a)$$

$$c = \frac{N'_C}{N_C} \hspace{1cm} (2b)$$

The condition (1) then becomes

$$\frac{N'_p}{N'_C} < \frac{2(p^2 - c)}{p(1 - p)}.$$  \hspace{1cm} (3)

Typical experimental values of $p$ and $c$ are 0.95 and 0.12 respectively. Using these values in (3), the method represents an improvement in counting for all cases where $\frac{N'_p}{N'_C} < 30$. This is always the case in practice since such large peaks do not occur. It follows that the anticoincidence method is an improvement in all situations.
APPENDIX II

INTERNAL REFLECTION IN LUCITE

The critical angle $\Theta$, for total internal reflection of light at an interface is given by

$$\Theta = \sin^{-1} \frac{1}{\mu},$$

where $\mu$ is the index of refraction.

For lucite $\mu = 1.50$.

Hence the critical angle $\Theta = 42^\circ$.

Thus the shoulders of the lucite light pipe (see Fig. 6) which are at an angle of $45^\circ$, are adequate to reflect most scintillations from the crystal to the photocathode of the photomultiplier.
PART 2

ON THE DOUBLE BETA DECAY OF $^{124}_{\text{Sn}}$
INTRODUCTION

The emission of single electrons from radioactive nuclei has been the subject of many researches during the past fifty years. It is now well established that in single beta-decay, the atomic number of the nucleus changes from $Z$ to $Z \pm 1$ depending upon whether a negative electron (negatron) or a positive electron (positron) has been emitted. If, however, the process involves only the emission of a single electron, it can easily be shown that the laws of conservation of energy and angular momentum are violated. To avoid this impasse, Pauli proposed his neutrino hypothesis which describes any single beta-decay process as being really the simultaneous emission of two particles, the electron and a particle of negligible mass and no charge. Then negatron decay is described as the conversion of one of the neutrons in the nucleus into a proton according to

$$N \rightarrow P + (\beta^- + \nu)$$

where $N$ and $P$ represent neutron and proton respectively, $\beta^-$ represents the emitted negatron and $\nu$ represents the "neutrino" with no charge and small mass. On the other hand positron decay is described as the conversion of a nuclear proton into a neutron according to

$$P \rightarrow N + (\beta^+ + \nu^*)$$

where $\beta^+$ represents the positron and $\nu^*$ represents the "anti-neutrino" also with no charge or mass. By virtue of the properties assigned to both $\nu$ and $\nu^*$ they are essentially undetectable.

In the ordinary form of the neutrino theory, neutrinos
FIG. 20

THE MASSES OF AN ISOBARIC TRIPLET.
are normally assigned to negative energy states which are almost always completely filled. A particle in such a state is called a neutrino. The few vacancies in this negative energy "sea" are called antineutrinos, a formalism quite analogous to that of Dirac's hole theory of the positron. Thus in this form, the neutrino and the antineutrino are distinguishable and represent two distinct particles. On the other hand, the Majorana form\(^3\) of the neutrino theory makes no distinction between \(\nu\) and \(\nu^*\) and states that the two particles are indistinguishable. Both forms of the neutrino theory make the same predictions for single beta-decay processes.

The condition whereby single beta-decay is energetically possible is simply that the nuclear mass of the parent atom be at least equal to the nuclear mass of the daughter atom plus the mass of the emitted electron. If the nuclear mass difference be greater than this lower limit, then the excess mass is converted into kinetic energy of the electron-neutrino pair. Expressed in terms of atomic mass units (whereby the orbital electron masses are included), the criteria may be summarized by stating that single negatron decay is possible if the mass of the whole parent atom is greater than the mass of the daughter atom.

Fig. 20 shows the atomic masses which are predicted for certain triplets of isobars (same mass number A but different Z) by the semi-empirical mass formula, a formula proposed by Weizsächer\(^3\) and others which predicts atomic masses with reasonable accuracy.

Nucleus 2 will certainly decay by negatron emission to nucleus 3. It may or may not go by positron emission to nucleus 1, depending upon the mass difference between 2 and 1. Nucleus 1 can-
not decay to 2 since mass considerations preclude this. To decay to 3, it would have to change from $Z$ to $Z + 2$ which would correspond to the simultaneous emission of two negatrons. Such a double beta process has not been definitely observed, although it is apparently energetically possible in several cases.

The calculated half-life for double beta-decay depends upon all the factors which affect the half-life of the single beta-decay process. These are the energy available (mass difference), the spins of the initial and final states and the parity change involved, the latter being a mathematical term which describes the symmetry properties of the wave functions describing the initial and final states. The calculations are based upon second-order perturbation theory. Since the transition probabilities calculated by the use of perturbation theory for second order processes are very small, the half-life of the double beta-decay process is predicted to be extremely large. If we assume that such calculations are even approximately correct, then should double beta-decay exist at all, its activity would be very weak. Hence it is not surprising that previously published work\textsuperscript{36-41} describing searches for this process quote conflicting results and that the existence of the process itself is in doubt.

Two different attacks have been made on the calculations of the probability of double beta-decay. In the ordinary theory, two neutrons decay in a double beta-process and the emission of two neutrinos is to be expected since

\[
\begin{align*}
N &\rightarrow P + (\bar{\nu}^+ + \nu^-), \\
N &\rightarrow P + (\bar{\nu}^+ + \nu^-), \\
2N &\rightarrow 2P + \bar{\nu}^+ + 2\nu^-.
\end{align*}
\]

\[
N \rightarrow P + (\bar{\nu}^+ + \nu^-), \\
N \rightarrow P + (\bar{\nu}^+ + \nu^-), \\
2N \rightarrow 2P + \bar{\nu}^+ + 2\nu^-.
\]
Thus, the double beta-decay process results in the simultaneous emission of four particles, (two negatrons and two neutrinos). Goeppert-Mayer\textsuperscript{33} has calculated the half-life of this process to be of the order of $10^{24}$ years.

The other neutrino theory, the Majorana form\textsuperscript{32}, makes no distinction between neutrino and antineutrino. Thus $\nu$ is the equivalent of $\bar{\nu}$. Now consider the single process

$$N \rightarrow P + (\bar{\nu} \rightarrow \nu)$$

This corresponds to the simultaneous emission of a negatron and a neutrino. But on the ordinary theory, an anti-neutrino, $\bar{\nu}$, is just a vacant negative energy state of a neutrino, so that the emission of a neutrino is exactly the same as the absorption of an antineutrino. Therefore the process could be written

$$\nu^* \rightarrow N \rightarrow P + (\bar{\nu} \rightarrow \nu)$$

and according to Majorana this is the same as

$$\nu \rightarrow N \rightarrow P + (\bar{\nu} \rightarrow \nu)$$

In the Majorana picture, the first neutron decays emitting a virtual neutrino which is absorbed by the second decaying neutron. Thus

$$2N \rightarrow 2P + 2(\bar{\nu})$$

No neutrinos are emitted and the process involves the emission of two particles only (two negatrons). As might be expected the calculated probability of this event is greater than for the four particle theory. Furry\textsuperscript{34} has calculated the half-life of the two particle process to be of the order of $10^{16}$ years.

A $10^{24}$ year activity (four particle concept) is incapable of detection using ordinary techniques, but $10^{16}$ years may be just within reach.
Thus double beta decay offers a means of deciding between the ordinary and the Majorana form of the neutrino theory. It very probably offers the only means since the direct detection of $\sqrt{2}$ and of $\sqrt{2}^*$ appears to be equally impossible.

Another consequence of double beta decay would be that the electric charge of the electron would be shown equal that of the proton.*

---

*Remark by Oppenheimer
II
SOME PREVIOUS EXPERIMENTS

A. The Triplet $^{124}\text{Sn} \rightarrow ^{124}\text{Sb} \rightarrow ^{124}\text{Te}$

Several earlier workers selected $^{124}\text{Sn}$ as a source. Double beta decay has been reported by Fireman$^{36}$ as observed in $^{124}\text{Sn}$ with a half-life of $0.4 \times 10^{16}$ years. The process allegedly observed was

$$^{124}\text{Sn} \rightarrow ^{124}\text{Te} + 2\beta^{-}.$$  

Fireman used two thin window Geiger counters on each side of a thin flat tin source. Only coincident events were recorded in order to identify the double beta process and in order to lower the background from cosmic rays and local contamination.

The background rate was further reduced by the use of lead shielding and by banks of Geiger counters connected in anticoincidence. The background rate reported was 14 counts per hour. With the tin in place a slightly larger rate was obtained. Fireman interpreted this difference to be caused by double beta decay, and calculated the half life from the mass of the source and the solid angles subtended by the counters. Absorption curves showed the "beta particles" had a maximum energy of 1.5 Mev.

Libby and Kalkstein$^{37}$ performed a similar experiment on $^{124}\text{Sn}$ and obtained a negative result. They were able to place a lower limit of $1.7 - 2.4 \times 10^{17}$ years on the half-life.
B. The Triplet \[ ^{46}\text{Pa}_{110} - ^{47}\text{Ag}_{110} - ^{48}\text{Cd}_{110} \]

Recently Winter investigated the reaction,
\[ ^{46}\text{Pa}_{110} \rightarrow ^{48}\text{Cd}_{110} + 2 \beta^- , \]
in a cloud chamber and obtained negative results. A lower limit of \(0.6 \times 10^{18}\) years was placed on the half-life of the decay.

C. The Triplet \( ^{52}\text{Te}_{130} - ^{53}\text{I}_{130} - ^{54}\text{Xe}_{130} \)

An investigation of the process,
\[ \text{Te}_{130} \rightarrow \text{Xe}_{130} + 2 \beta^- , \]
was made by Inghram and Reynolds. A mass spectrometer was used to search for the presence of \(\text{Xe}_{130}\) in telluride ores. An excess of \(\text{Xe}_{130}\) was found and was attributed to the double beta decay of \(\text{Te}_{130}\). From a fairly reliable estimate of the age of the ore, a half-life of \(1.4 \times 10^{21}\) years was found.

D. The Triplet \( ^{92}\text{U}_{238} - ^{93}\text{Np}_{238} - ^{94}\text{Pu}_{238} \)

A negative result has been obtained by Seaborg et al. in an investigation of the process,
\[ ^{92}\text{U}_{238} \rightarrow ^{94}\text{Pu}_{238} + 2 \beta^- . \]
The available energy is known to be 1.1 Mev. from the decay schemes of neighbouring isotopes. A search was made for the presence of 90 day \(\text{Pu}_{238}\) in pure \(\text{U}_{238}\). This was done by chemical separation of the \(\text{Pu}_{238}\) followed by a search for the 5.51 Mev. alpha particles from \(\text{Pu}_{238}\). No such activity was found, and a lower limit of \(6 \times 10^{18}\) years was set on the half-life.
Since double beta decay was reported in Sn\(^{124}\), this isotope was chosen for our investigation. Recently Duckworth\(^{42}\) has measured the Sn\(^{124}\) - Te\(^{124}\) mass difference to be \(1.5 \pm 0.4\) Mev. by an accurate mass spectrometer.

Our experiment was quite different in nature from the experiments described above, and we feel, should lead to a more reliable identification of double beta decay, should it exist.
III
EXPERIMENTAL PROCEDURE

A. The Principle of the Experiment

If double beta decay is observable at all, it follows from the half-life considerations that it will very probably be a two particle process. For this reason, the experiment was designed to make fuller use of the properties of the two particle process than was made in previous work.

A distinguishing feature of the two particle process is that the sum of the energies of the two beta particles has a constant value equal to the total energy available for the process. This is because the two beta particles receive all the energy carried away. This is not the case when neutrinos are emitted.

This unique feature was used in an experiment in which the sum of the energies of coincident events in Sn$^{124}$ was recorded on a pulse amplitude analyser or "kicksorter". The energy spectrum of the double beta process thus displayed should consist of a sharp peak at an energy corresponding to the sum of the two beta energies. The background spectrum, however, should appear as a smooth distribution.

B. The Experimental Arrangement

The block diagram of the experimental arrangement is shown in Fig. 21. A source consisting of a 200 mg, tin foil was placed between two scintillation counters in a light-tight box. The source was on loan from the Oak Ridge National Laboratories and was enriched to 95% Sn$^{124}$ by electromagnetic separation. Coincident beta particles were detected in thick anthracene crystals...
(1" x 1" x 1"). which faced the tin foil. The pulse amplitudes from the scintillation counters were used as a measure of the beta energies. The sum of the energies of the two betas was represented by the sum of the amplitudes of coincident pulses which was obtained in the pulse adder. The pulse representing this sum was then displayed on an 18 channel kicksorter of Chalk River design. A gate controlled by the coincidence mixer allowed the output from the pulse adder to reach the kicksorter only in the case of a coincident event.

C. Calibration of the Kicksorter

In order to calibrate the energy scale on the kicksorter, the beta spectrum of Sb$^{124}$ was observed in each crystal separately. The endpoint of the most energetic beta groups of Sb$^{124}$ at 2.4 Mev. was used as a calibration point. Using the same point, the overall gains of the scintillation counters were made equal by adjusting their high voltage supplies.

D. The Optimum Source Thickness

The thickness of material between the two crystals seemed to have an effect on the amount of scattering from crystal to crystal and hence on the coincidence rate. Therefore it was found necessary to substitute a dummy foil for the tin source while taking the background rate in order to keep the background intensity equal to that from the tin. Both the tin and dummy foil were made the same thickness, 100 m$\gamma$/cm$^2$. This particular weight was chosen for the following reasons:

(i) a thick source reduces scattering of stray radiation from crystal to crystal, and
<table>
<thead>
<tr>
<th>Backgro</th>
<th>Tin</th>
<th>Difference</th>
</tr>
</thead>
<tbody>
<tr>
<td>12.9</td>
<td>12.6</td>
<td>11.1</td>
</tr>
<tr>
<td>12.7</td>
<td>12.6</td>
<td>13.0</td>
</tr>
<tr>
<td>10.5</td>
<td>19.4</td>
<td>10.8</td>
</tr>
<tr>
<td>11.0</td>
<td>11.0</td>
<td>11.0</td>
</tr>
<tr>
<td>3.8</td>
<td>4.1</td>
<td>4.1</td>
</tr>
<tr>
<td>24.1</td>
<td>24.1</td>
<td>20.4</td>
</tr>
<tr>
<td>24.2</td>
<td>23.1</td>
<td>23.1</td>
</tr>
<tr>
<td>6</td>
<td>0.05</td>
<td>0.02</td>
</tr>
<tr>
<td>7</td>
<td>0.04</td>
<td>0.01</td>
</tr>
<tr>
<td>-0.1</td>
<td>0.03</td>
<td></td>
</tr>
<tr>
<td>22</td>
<td>0.18</td>
<td>0.04</td>
</tr>
<tr>
<td>24</td>
<td>0.14</td>
<td>0.03</td>
</tr>
<tr>
<td>-0.4</td>
<td>0.04</td>
<td></td>
</tr>
<tr>
<td>37</td>
<td>0.36</td>
<td>0.06</td>
</tr>
<tr>
<td>24</td>
<td>0.24</td>
<td>0.04</td>
</tr>
<tr>
<td>-1.2</td>
<td>0.07</td>
<td></td>
</tr>
<tr>
<td>36</td>
<td>0.36</td>
<td>0.06</td>
</tr>
<tr>
<td>37</td>
<td>0.26</td>
<td>0.07</td>
</tr>
<tr>
<td>0.10</td>
<td>0.09</td>
<td></td>
</tr>
<tr>
<td>43</td>
<td>0.39</td>
<td>0.06</td>
</tr>
<tr>
<td>57</td>
<td>0.37</td>
<td>0.05</td>
</tr>
<tr>
<td>-0.2</td>
<td>0.08</td>
<td></td>
</tr>
<tr>
<td>53</td>
<td>0.48</td>
<td>0.06</td>
</tr>
<tr>
<td>79</td>
<td>0.51</td>
<td>0.06</td>
</tr>
<tr>
<td>+0.3</td>
<td>0.09</td>
<td></td>
</tr>
<tr>
<td>58</td>
<td>0.53</td>
<td>0.06</td>
</tr>
<tr>
<td>63</td>
<td>0.41</td>
<td>0.05</td>
</tr>
<tr>
<td>-1.2</td>
<td>0.09</td>
<td></td>
</tr>
<tr>
<td>59</td>
<td>0.54</td>
<td>0.06</td>
</tr>
<tr>
<td>85</td>
<td>0.35</td>
<td>0.06</td>
</tr>
<tr>
<td>+0.1</td>
<td>0.09</td>
<td></td>
</tr>
<tr>
<td>69</td>
<td>0.63</td>
<td>0.07</td>
</tr>
<tr>
<td>99</td>
<td>0.64</td>
<td>0.07</td>
</tr>
<tr>
<td>-0.1</td>
<td>0.10</td>
<td></td>
</tr>
<tr>
<td>83</td>
<td>0.75</td>
<td>0.08</td>
</tr>
<tr>
<td>102</td>
<td>0.69</td>
<td>0.07</td>
</tr>
<tr>
<td>-0.8</td>
<td>0.11</td>
<td></td>
</tr>
<tr>
<td>72</td>
<td>0.65</td>
<td>0.08</td>
</tr>
<tr>
<td>114</td>
<td>0.74</td>
<td>0.07</td>
</tr>
<tr>
<td>+0.9</td>
<td>0.11</td>
<td></td>
</tr>
<tr>
<td>97</td>
<td>0.88</td>
<td>0.09</td>
</tr>
<tr>
<td>120</td>
<td>0.78</td>
<td>0.07</td>
</tr>
<tr>
<td>-1.6</td>
<td>0.12</td>
<td></td>
</tr>
<tr>
<td>119</td>
<td>0.92</td>
<td>0.10</td>
</tr>
<tr>
<td>142</td>
<td>0.92</td>
<td>0.08</td>
</tr>
<tr>
<td>-0.0</td>
<td>0.13</td>
<td></td>
</tr>
<tr>
<td>215</td>
<td>1.95</td>
<td>0.13</td>
</tr>
<tr>
<td>283</td>
<td>1.98</td>
<td>0.11</td>
</tr>
<tr>
<td>+0.3</td>
<td>0.18</td>
<td></td>
</tr>
<tr>
<td>3785</td>
<td>74.4</td>
<td>0.03</td>
</tr>
<tr>
<td>4876</td>
<td>54.0</td>
<td>0.03</td>
</tr>
<tr>
<td>+0.3</td>
<td>0.18</td>
<td></td>
</tr>
</tbody>
</table>

* Background or Tin.
(ii) Multiple scattering in a thick source tends to remove any angular correlation which might occur in double beta decay and so affect the measured coincidence rate. A compromise between the loss of transmission for betas with a thick source and the favourable effects mentioned above was made at a thickness of 100 m\(^2\)/cm\(^2\).

It is to be expected that the source thickness will not have much effect on the width of the kicksorter peak. This is because the rate of energy loss in the region from 0.3 Mev. to 3 Mev. and the total path length of the two betas are both nearly constant. Thus the total energy lost in the foil tends to be constant, and the peak remains sharp for thick foils.

E. Obtaining the Data

Dummy foils made of silver and of natural tin were found to be slightly active and were discarded in favour of aluminum. The tin and dummy were interchanged periodically during the run in order to compensate for any instrumental drift. They were mounted on a slide with an external control. In this way the source and dummy could be interchanged without disturbing the equipment. The counters were shielded by 16 cm. of lead from above, and by 8 cm. of lead in other directions. The total number of coincidences were recorded on a scaler during each run.

The data shown in the portion of Table III called "kicksorter readings" were obtained during a total counting period of 264 hours. The right side of the table shows the average counts per hour as calculated for each channel. The difference between the tin intensity and the background is shown on the extreme right.
Fig. 22. Coincidence Spectra Obtained on Kicksorter.
The spectra from tin and from background so obtained are plotted in Fig. 22. Standard deviations are shown for the background points. The standard deviations for the tin spectrum are not shown, but are slightly less. The large number of counts in channel 18 (energies of 3 Mev. and larger) are due to cosmic radiation. The counting rate of channel 18 was large enough to provide an experimental check on the stability of the kicksorter and of the pulse amplifiers. The rate in channel 18 is shown at the bottom of Table IIi together with the total coincidence rate obtained on the external scaler. It is seen that the arrangement was satisfactorily stable.

F. The Shape of the Background Spectrum

The first two channels received no counts because the coincidence mixer discriminators were set to accept only 0.3 Mev. events. This was done since the acceptance of lower energy particles increased the background in all channels. The effect of discriminator setting on the background per channel can be seen from these measurements taken in the 2 Mev. channel: the backgrounds at discriminator settings of 0.1, 0.2 and 0.3 Mev. were approximately 2, 1 and ½ counts per hour. A compromise between having a low background and losing part of the double beta spectrum was taken by setting the discriminators at 0.3 Mev.

Another factor to which the background rate proved sensitive was the separation of the anthracene crystals. The total background at 3 mm. crystal separation was 155 ± 10 counts per hour, and at 1 cm. was 44 ± 1 counts per hour. So the 1 cm. separation was preferable notwithstanding the slight loss in solid angle.
In an attempt to lower the background rate still further an anticoincidence device was devised. It consisted of a scintillation counter using a solution of terphenyl in toluene as a scintillator. This solution was contained in a large silvered vessel placed above and around the two anthracene crystals. This had the effect of removing one fourth of the counts from the 18th channel and leaving the important center channels untouched. For this reason it was discarded as an unnecessary complication. The final arrangement had a crystal separation of 1 cm., and no anticoincidence device.

The poor efficiency of the anticoincidence device was rather surprising. However, at the discarded 3 mm. separation it successfully cancelled 30% of the total background. So the explanation of the poor cancellation must lie in a poor geometry for triple coincidences. Probably the anticoincidence device is very sensitive to penetrating showers and the geometry for showers is best when the crystals are close.

The low rate of 0.5 per hour per channel made experimenting with the geometry tedious. At least two days were necessary to test the background with each new arrangement.

G. Comparison with Previous Work

A fair comparison of background in this and previous work can be made if we consider backgrounds at the peak position. It has been shown (part D) that absorption in the foil would not widen the peak. So the peak width may be caused by the energy resolution of the scintillation counters only. If this resolution is 10%, then the peak would occupy one, or at the most two, channels. Since the background is 0.5 counts per hour per channel, the
"effective background" is no more than 1.0 per hour. This compared favourably with that of Fireman\textsuperscript{36} (14 per hour) and that of Libby\textsuperscript{37} (80 per hour).

The second advantage of this experiment was that it was capable of unambiguous interpretation. It is difficult to imagine a radioactive contamination which would have given rise to a peak. On the other hand, contamination effects in conventional counting experiments are indistinguishable from true effects. The conflicting results of Fireman and Libby represent a case in point.
Fig. 23. Difference Between Coincidence Spectra Shown in Fig. 22.

SUM OF THE BETA ENERGIES - Mev.
RESULTS AND CONCLUSIONS

Fig. 23 shows the coincidence spectrum derived from Fig. 22 by subtracting the background. The standard deviations are shown. No trace of a peak was found.

In order to place a lower limit on the half-life of double beta decay from this negative result, the smallest effect detectable in Fig. 23 must be estimated. To do this a knowledge of the peak width is important. The width was estimated in Section IIIG to be one, or at the most two, channels. Thus a peak of 0.2 counts per hour would just be detectable in Fig. 23.

The fractional solid angle subtended by each crystal was slightly less than 0.5. The geometric efficiency for coincidence counting is therefore close to 0.2, a consideration not noted by Fireman\textsuperscript{36}. The correct half-life observed by Fireman is $0.2 \times 10^{16}$ years rather than $0.4 \times 10^{16}$ years.

The losses from absorption in the source may be taken into account by a transmission factor, $F$, which is estimated to be 0.4 at 1.0 Mev. and 0.7 at 3.0 Mev.\textsuperscript{43} This transmission factor includes an estimate of the counting losses incurred by biasing the coincidence mixers at 0.3 Mev.

Thus a detectable disintegration rate is

\[
\frac{dN}{dt} = \frac{0.2}{0.2F} \text{ counts per hour ,}
\]

\[
= \frac{1}{F} \times 8,760 \text{ counts per year .}
\]

The number of Sn$^{124}$ atoms present in the source may be calculated by

\[
N = \frac{Lmf}{N_f} ,
\]
where \( m \) (the mass used) = 200 mg,

\[ L \text{ (Loschmidt's number)} = 6 \times 10^{23}, \]

\[ f \text{ (the fraction of enrichment in Sn}^{124}) = 0.95, \]

and \( M_T \text{ (the molar weight of tin)} = 124 \text{ gms}. \]

So

\[ N = \frac{6 \times 10^{23} \times 0.2 \times 0.95}{124}, \]

\[ = 0.92 \times 10^{21} \text{ atoms}. \]

Then the lower limit of the half-life is given by

\[ T = \frac{\log 2}{\frac{dN}{dt}} N, \]

\[ = \frac{0.693 \times 10^{21} \times 0.92}{\frac{1}{F} 8,760}, \]

\[ = 0.3 \times 10^{17} \text{ to } 0.7 \times 10^{17} \text{ years.} \]

The negative results cannot be construed as showing the Majorana theory wrong, for the transition studied might be forbidden by the change of spin or parity involved.

The work is in agreement with the work of W.F. Libby\(^{37}\) and J.S. Lawson\(^{38}\).\(^*\) The results are in disagreement with the work of E.L. Fireman.\(^{36}\)

---

*\(^{*}\)A similar experiment to the one reported here with essentially the same results has just been reported by J.A. McCarthy, Bull. Am. Phys. Soc. 27, No. 3 18 (1952).
REFERENCES

2. G.T. Seaborg, I. Perlman, Rev. Mod. Phys. 20, 585 (1948).
8. R.C.A. Tube Handbook, Tube Division, Harrison, N.J.
   A, 143: 350 (1934)
References (continued)

31. E. Rutherford, F. Soddy, Phil. Mag. 4, 370, 569 (1902); 5, 441, 576 (1903).
42. Benjamin G. Hogg, Henry E. Duckworth, private communication.