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COINCIDENCE MEASUREMENTS IN
NUCLEAR DECAY SCHEME STUDIES

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DECAY SCHEME STUDIES

John P. Unik

(Thesis)

March 1960

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COINCIDENCE MEASUREMENTS IN NUCLEAR DECAY-SCHEME STUDIES

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March 1960

ABSTRACT

The decay schemes of the isomers of Tc^{95} and Tc^{97} have been studied by using high-resolution conversion-electron spectrographs, gamma-ray scintillation detectors, and coincidence techniques.

The half-life of the 74.6-keV excited state in Np^{239} has been measured by the delayed coincidence technique as 1.2 ± 0.1 μsec .

The directional correlations of two gamma-ray cascades following the beta decay of U^{237} to Np^{237} have been studied and the results are consistent with previous assignments of the total angular momenta of the excited states. An upper limit on the half-life of the 332-keV excited state in Np^{237} was determined to be 1.0 μsec .

Electron-electron coincidence measurements have been performed on the beta decay of Pa^{233} . This work removes some of the ambiguities between previous coincidence work and the previously proposed decay scheme.

The transmission of one half of the original electron-electron coincidence spectrometer has been increased by a factor of six without sacrificing resolution. This increased transmission was achieved by converting the original thin-lens magnetic field to a thick-lens triangular magnetic field.

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I. INTRODUCTION

During the past decade rapid progress has been made toward the understanding of nuclear structure by detailed theoretical studies and the acquisition of precise experimental data. As our knowledge increases, more refined experimental equipment and techniques must be developed to overcome and fully resolve the complexities present in nuclear structure.

This study is concerned with the development and application of coincidence methods used in the study of excited states of nuclei. These coincidence methods are applied to studies of gamma-gamma-ray angular distributions and gamma-ray transition probabilities and to the elucidation of nuclear energy-level spacings in conjunction with high-resolution beta spectroscopy. The decay schemes of a number of radioactive isotopes have been studied and the results are analyzed in terms of nuclear decay theory and current nuclear models.

II. DECAY-SCHEME STUDIES

A. Experimental Methods

1. General Approach

The method used throughout this work in studying the excited states of nuclei consists of an analysis of the emission spectra (alpha and beta particles, gamma rays, internal conversion electrons) of the transitions populating and depopulating the excited states.

From the properties of the emission spectra one can deduce certain properties of the excited states such as total angular momentum and parity. The excited states of the nuclei are populated by means of a radioactive isotope decaying to the isotope that we are interested in studying. In general the decay process populates a number of excited states, the relative populations depending, quite strongly, on the properties of the initial and final states involved in the decay process.

2. Gamma-Ray Spectroscopy

The gamma-ray spectra were obtained by using thallium-activated sodium iodide scintillation crystals in conjunction with 50-channel¹ and 100-channel² pulse-height analyzers. Most of the gamma-ray spectroscopy was performed by using 1-1/2-inch-diameter by 1-inch-thick cylinders and 3-inch-diameter by 3-inch-thick beveled-edge cylinders of thallium-activated sodium iodide. The efficiency curves given by Kalkstein and Hollander³ were used for the 1 x 1-1/2-inch crystals, and the curves given by Heath⁴ were used for the 3 x 3-inch crystals.

The coincidence relationships between various gamma rays were determined by displaying the spectrum of gamma rays in coincidence with a selected gamma ray on a multichannel analyzer. The particular gamma ray of interest was selected by means of a single-channel pulse-height analyzer. A gamma ray would appear on the multichannel analyzer only if its pulse and the pulse from the single-channel analyzer arrived at the slow-coincidence circuit (whose output activated the multichannel analyzer) within its resolving time (about 2 μ sec).

3. Internal-Conversion-Electron Spectroscopy

The internal-conversion-electron spectra were studied by using high-resolution permanent-magnet spectrographs.⁵ These spectrographs consist essentially of a source and an emulsion mounted in a vacuum tank between two parallel iron plates which are permanently magnetized. The source consists of a suitable activity electrodeposited onto a 0.01-inch-diameter platinum wire. Internal-conversion electrons emitted from the source pass through a collimating slit, are deflected in a circle by the magnetic field, and strike the emulsion; the radius of the circle is proportional to the momentum of the electron. An IBM 650 computer was used to calculate the electron energies from the magnetic field calibration, the dimensions of the system, and the distance of the conversion-electron line from a fixed point on the film. Two spectrographs were used with magnetic fields of approximately 100 and 350 gauss, which corresponded to maximum observable electron energies of roughly 260 and 1600 kev, respectively. These spectrographs were calibrated with the well-known transitions in W^{183} ,⁶ and Pb^{206} .⁷ With these spectrographs electron energies could be determined to roughly 0.1%, depending on the intensity of the internal-conversion-electron line.

The relative conversion-electron intensities were determined by taking a densitometer tracing of the emulsion, relating photographic densities to relative intensities by the method of Slätis⁸ and also by visual comparison of the internal-conversion electron lines to standard exposed emulsions.⁹ The intensities so obtained were corrected for the different geometries and photographic blackening efficiencies.

The permanent-magnet spectrographs are very useful for studying long-lived activities, since the photographic emulsion used as a detector can integrate the line intensities over a very long time.

4. Bombardments

High-resolution studies of long-lived accelerator-produced isotopes have been until recently rather difficult to perform since they involve analysis of internal-conversion electrons, generally of rather weak intensity (compared with the total activity), in magnetic spectrometers

of very low transmissions. The large amounts of these long-lived activities necessary for accurate analysis can generally not be produced during convenient-length bombardments on the available accelerators.

We were quite fortunate to have available to us the Livermore A-48 Linear Accelerator¹⁰ before it was dismantled. The A-48 Linear Accelerator was capable of producing 7.4-Mev deuterons with a beam intensity under usual operating conditions of about 30 milliamperes, which is a few orders of magnitude greater than that obtained on the available cyclotrons.

In the use of this accelerator, one is limited with respect to target material by its melting point, since with such high beam currents cooling of the target is a severe problem. Molybdenum and tungsten both made convenient target materials because of their high melting points. Bombardment of natural molybdenum produced several technetium isotopes. The Coulomb barrier for the bombardment of tungsten with deuterons is 11 Mev, slightly greater than the available energy; however, it was found that large amounts of 71-day Re^{183} and 50-day Re^{184} were produced, apparently by barrier penetration. The work on the radioactive decays of Re^{183} and Re^{184} was performed in conjunction with Dr. Strominger and Dr. Gallagher. Since this work has already been completely reported elsewhere,¹¹ it is not presented here.

B. Decay Scheme of the 60-Day Isomer of Tc^{95}

1. Introduction

The energy levels of Mo^{95} populated by the decay of the 60-day isomer of Tc^{95} have previously been studied by Medicus, Preiswerk, and Scherrer¹² and also by Levi, Papineau, and Saunier.¹³ Medicus et al. observed four gamma rays, using the rather poor resolution methods of Compton electron spectrometry and absorption in lead. Levi et al. reported two equally intense positron groups to the ground state and the first excited state of Mo^{95} , and also set upper limits of the electron capture to these states.

The study reported here was undertaken to obtain accurate transition-energy and -intensity data for a more detailed study of the excited states of Mo^{95} .

2. Chemistry

Chemical processing of the molybdenum target was delayed 3 months to allow the shorter-lived activities to decay. The target was dissolved in a solution of HCl and HNO_3 and evaporated nearly to dryness. The precipitate that formed was then digested in a small volume of HNO_3 so that the bulk of the molybdenum would precipitate as molybdic acid. After the solution was cooled to reduce the solubility of the molybdic acid, the supernatant liquid containing most of the technetium activity was removed. The molybdic acid precipitate was dissolved and reprecipitated three times in order to obtain the remaining technetium occluded in the precipitate. The technetium was then separated from the remaining soluble molybdenum by use of an anion-exchange separation.¹⁴ The technetium obtained in this manner was mass-free. Most of the activity was used to prepare a source for the conversion-electron studies. This source was prepared by cathodic electrodeposition of the technetium activity onto a 0.010-inch platinum wire. The remaining activity was used to prepare sources for the gamma-ray analysis.

3. Gamma-Ray Spectra

Spectrum A of Figure 1 illustrates the singles gamma-ray spectra taken with the 1 x 1-1/2-inch NaI crystal. In addition to the K x-rays, gamma rays of 204, 580, 820, and 1040 kev were observed. The 820-kev gamma ray was the only one that appeared to be complex. All the gamma rays decayed with a half-life of 61 ± 2 days. These gamma rays were therefore assigned to the decay of $\text{Tc}^{95\text{m}}$, which has been previously reported to have a half-life of about 60 days.¹⁵ Medicus *et al.* have reported gamma rays of 201, 510, 810, and 1017 kev, but did not observe that the 810-kev gamma ray was complex. The K x-rays decayed with a half-life longer than 60 days, indicating the presence of the 90-day isomer of Tc^{97} .

Table I summarizes the gamma rays of $\text{Tc}^{95\text{m}}$ that were observed and their corresponding relative intensities. The intensities given represent an average of the intensities obtained by using 1 x 1-1/2-inch and 3 x 3-inch cylindrical NaI (Tl) scintillation crystals.

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Fig. 1. Singles and coincidence gamma-ray spectra of $\text{Tc}^{95\text{m}}$.
Curve A - Singles spectrum.
Curve B - Gamma-ray spectrum in coincidence with
204-kev gamma ray.
Curve C - Complex photopeak not in coincidence with
the 204-kev gamma ray.

Table I. Gamma rays of $\text{Tc}^{95\text{m}}$

Energy (kev)	Relative intensity
K x-rays	1.6 ± 0.4
204 ± 2	1.8 ± 0.2
580 ± 6	1.0
820 ± 10	1.45 ± 0.15
1040 ± 10	0.10 ± 0.02

The relative intensity of K x-rays due to Tc^{95} was distinguished from the K x-rays of $\text{Tc}^{97\text{m}}$ by resolving the decay curve of the K x-rays taken over a period of one year.

The results of the gamma-gamma coincidence measurements can be summarized as follows:

(a) The 204-kev gamma ray is in coincidence with the high-energy side of the 820-kev complex photopeak, spectrum B of Fig. 1. It was found by gating on the 204-kev peak and comparing the coincidence spectrum with the singles spectrum that $53 \pm 5\%$ of the 820-kev peak was in coincidence with the 204-kev gamma ray. This gamma ray, then, has an intensity of 0.77 ± 0.08 relative to the 580-kev gamma ray.

(b) The energy of the gamma ray in coincidence with the 204-kev gamma ray is 840 ± 20 kev, and appears to be a single gamma ray when the peak width is compared with the 662-kev gamma ray of Cs^{137} and the 1064-kev gamma ray of Bi^{207} . After subtracting out the contribution of the 1040-kev gamma ray from the singles spectrum (using the 1064-kev gamma ray of Bi^{207} as a standard) and then subtracting the coincidence spectrum from the resultant singles spectrum, one obtains a complex peak at 800 ± 20 kev, spectrum C, Fig. 1. This complex peak has a half width about 20 kev greater than that of the 840-kev gamma ray.

(c) The 580-kev gamma ray is completely in coincidence with the 204-kev gamma ray.

(d) The 580-kev and the complex 820-kev gamma rays are not in coincidence.

(e) The 1040-kev gamma ray is not in coincidence with any gamma ray.

4. Internal-Conversion-Electron Spectra

Tables II and III summarize the conversion-electron work. Because of the small differences between different L- and M-subshell binding energies, the subshell of conversion for a particular transition could not be determined uniquely. The final transition energies are determined from the K-conversion electron lines, by use of the binding energies given by Hill, Church, and Mihelich.¹⁶ For the first three electron lines in Table II, the relative intensities were difficult to estimate, since the lines were rather diffuse and in a high background, and also because of the large uncertainties in the blackening efficiencies of the emulsion at these low energies.

5. Positrons

Since the 510-keV annihilation-radiation photopeak was masked by the intense 580-keV photopeak, the relative number of positrons to a particular gamma ray could not be determined directly by comparing the relative gamma-ray intensities. However, the ratio of positrons to the 580-keV gamma ray was obtained in the following manner. The 510-510-keV annihilation-radiation coincidence counting rate (A) in the technetium sample was determined. According to the previous coincidence work, there is no complication due to other gamma-ray coincidences. From energy considerations it can be shown that the 580-keV gamma ray and higher-energy gamma rays cannot be in coincidence with positrons. Levi and Papineau have determined from (p,n) thresholds that the available energy for electron capture for the isomer is 1.73 MeV.¹⁷ Since the 580-keV gamma ray is in coincidence with the 204-keV gamma ray, and it is to be shown (later in this paper) that the first excited state is at 204 keV, the 580-keV gamma ray must depopulate a level at about 784 keV. From the Q_{EC} the highest energy level that can be populated by positrons is 710 keV. Therefore the 580-keV gamma ray and the higher-energy gamma rays observed cannot be in coincidence with positrons. With the same geometry, the 510-510-keV coincidence rate (B) was measured for a Na^{22} sample. Both samples were surrounded by an appropriate thickness of aluminum to stop all positrons. Then the singles counting rates of both

Table II. Conversion-electron lines of $\text{Tc}^{95\text{m}}$ (99-gauss spectrograph)

Electron energy (kev)	Shell of conversion	Transition energy (kev)	Relative electron intensities
17.88 ± 0.1	K^{a}	38.9 ± 0.1	
36.19 ± 0.4	L^{a}	38.9	
38.44 ± 0.4	M^{a}	38.9	
184.2 ± 0.2	K	204.2 ± 0.2	100
201.3 ± 0.2	L	204.2	13 ± 2
203.7 ± 0.2	M	204.2	4 ± 1

a. Internal conversion occurs in technetium; lines not so designated occur in molybdenum.

Table III. High-energy conversion-electron lines of $\text{Tc}^{95\text{m}}$
(350-gauss spectrograph)

Electron energy (kev)	Shell of conversion	Transition energy (kev)	Relative electron intensities
563.9 ± 0.6	K	583.9 ± 0.6	100
580.8 ± 0.6	L	583.9	13 ± 2
743 ± 2	K	763 ± 2	~ 1
747.9 ± 0.8	K	767.9 ± 0.8	5 ± 1
764 ± 2	K	784.0 ± 2	~ 2
768.0 ± 0.8	K	788.0 ± 0.8	14 ± 2
785 ± 2	L	788.0	~ 2
802.5 ± 0.8	K	822.5 ± 0.8	6 ± 1
817.3 ± 0.8	K	837.3 ± 0.8	40 ± 4
834.5 ± 1.6	L	837.3	4 ± 1

samples were determined, which gave the sum of the 510- and 580-keV gamma rays counting rate (C) in the technetium sample and the 510-keV annihilation-radiation counting rate (D) in the Na^{22} sample. The number of positrons relative to the 580-keV gamma ray is then given by

$$\frac{N(B^+)}{N(580)} = 1/2 \frac{AD}{(BC-AD)} \quad (1)$$

The ratio of positrons to the 580-keV gamma ray was found to be 0.010 ± 0.003 . The number of positrons relative to the 204-keV gamma ray is then 0.0056 ± 0.0018 . This ratio differs appreciably from the value of 1.1×10^{-3} relative to the 204-keV gamma ray reported by Levi, Papineau, and Saunier.¹³ Using the value of 1.6 for the K x-ray intensity relative to the 580-keV gamma ray, a fluorescence yield of 0.73,¹⁸ and a L/K capture ratio of 0.11,¹⁹ one obtains an estimate of the positron-to-electron-capture ratio of $0.41 \pm 0.16\%$. This value agrees with the value of 0.4% reported by Medicus *et al.*¹²

6. Energy Levels of Mo^{95}

The 38.9-keV transition is highly converted, and from the energy spacings of the K and L lines it is clear that the internal conversion occurs in technetium and not in molybdenum. These observations strongly suggest that this transition corresponds to an isomeric transition in technetium. The isomeric transition is probably in Tc^{95} , and was reported by Medicus and Preiswerk.²⁰ Medicus and Preiswerk reported the transition energy as 39.0 ± 0.7 keV, in good agreement with the energy obtained in this work, and they determined the half-life of the transition to be 60 ± 10 days.

The first excited state of Mo^{95} has been shown to be at 204 keV by several investigators,^{12,21} therefore the 204.2-keV transition is assigned as depopulating the first excited state. This is consistent with all the coincidence data and the fact that the 204.2-keV transition is the most intense in the decay.

The 204.2-keV transition was found to be in coincidence with the 580-keV gamma ray, the energy of which was more accurately determined as

583.9 keV from the conversion-electron spectroscopy. This result suggests that an energy level exists at $583.9 + 204.2 = 788.1$ keV. This postulate is substantiated by the observation in the conversion-electron spectra of a 788.0-keV transition, the cross-over transition to the ground state.

The 204.2-keV transition is also in coincidence with the high-energy side of the 820-keV complex gamma-ray peak. The energy of this gamma ray in coincidence with the 204.2-keV transition was found to be 840 ± 20 keV, and it appears to be a single gamma ray. This transition is most likely the 837.3-keV transition observed in the conversion-electron work. Since the 837.3-keV transition is in coincidence with the 204.2-keV transition, there is good evidence that an energy level exists at 1042 keV. This postulated energy level is substantiated by the observation of a 1040 ± 10 -keV gamma ray, which is not in coincidence with any gamma ray.

The gamma-ray photopeak not in coincidence with the 204.2-keV transition was complex, with a width at half maximum about 20 keV greater than that of the 840-keV gamma ray. This photopeak is most likely due to the 767.9-, 788.0-, and 822.5-keV transitions.

The coincidence results confirm the postulate that the 788.0-keV transition observed in the conversion-electron work is the cross-over transition to the ground state.

The 767.9-keV transition is very likely the transition that has been observed in the 20-hour electron capture of the ground state of Tc^{95} (762 keV)¹² and the beta decay of Nb^{95} (767 ± 2 keV,²² 768 ± 1.5 keV,²³ and 770 ± 2 keV²⁴). This transition has previously been shown to go directly to the ground state, in agreement with this work.

Since the 822.5-keV transition is not in coincidence with any gamma ray, it is assigned as going directly to the ground state.

Very little can be said conclusively at this time concerning the coincidence relations of the 763- and 784-keV transitions with the 204.2-keV transition, because of their relatively low intensities. If these transitions have comparable or smaller conversion coefficients relative to the other high-energy transitions, it would be unlikely that they are in coincidence with the 204.2-keV transition.

7. Gamma-Transition Multipolarities

The 204.2-keV transition has been shown by recent Coulomb-excitation work²¹ to be an M1-E2 mixture, with a mixing ratio E2/M1 of 0.34 ± 0.17 . The K/L conversion-electron ratio of 7.7 ± 1.2 obtained in this work is consistent with such an M1-E2 admixture. The theoretical K/L ratio for an M1 transition is 8.8; for E2 the ratio is 7.1. The theoretical conversion coefficients from Sliv and Band²⁵ are used throughout this paper.

The multipolarity assignments from conversion coefficients for the other gamma rays are very difficult to make because, with the exception of the 584- and 837-keV transitions, the relative gamma-ray intensities could not be obtained. Also at this particular atomic number, for the energies under consideration, the conversion coefficients for M1 and E2 multipolarities are very similar. At 584 keV the two conversion coefficients differ by about 15%, while at 837 keV they differ by only about 5%. Also the K/L ratios obtained from the emulsions in this work are considered to have an uncertainty too large to allow one to unambiguously distinguish between M1 and E2 multipolarities.

In this work the absolute conversion coefficient for the 584-keV transition was not determined. However, from the conversion-coefficient work of Medicus et al.¹² the 584-keV transition was shown to be either M1 or E2. The experimental ratio of the K-conversion coefficient for the 584-keV transition to that of the 837-keV transition, from this work, is 1.9 ± 0.3 . If the 584-keV transition were of M1 multipolarity and the 837-keV transition M1 or E2, the theoretical ratio would be 2.2. If the 584-keV transition were E2, the ratio would be 2.6. Therefore the 584-keV transition is more likely to be predominantly M1 and the 838-keV transition M1 or E2.

The 768-keV transition has been shown by Drabkin, Orlov, and Rusinov to be of E2 multipolarity.²⁴

The 788-keV transition cannot involve a parity change, since it is the cross-over transition connecting two states of the same parity. Both the 584- and the 204.2-keV transitions have been shown to involve no parity change.

Limits on the multipolarities of the 788- and 822-keV transitions can be obtained from the relative conversion-electron intensities. The gamma-ray and conversion-electron intensities were normalized to both the 584- and the 837-keV transitions by using the theoretical M1 conversion coefficient and the average of the theoretical M1 and E2 conversion coefficients respectively. The gamma-ray intensities for the transitions were then calculated from the relative conversion-electron intensities by assuming various multipolarities and using the theoretical conversion coefficients. The sum of the calculated gamma-ray intensities was then compared to the experimental sum of 0.68 ± 0.17 for the intensity of the gamma rays, relative to the 584-keV transition, which were not in coincidence with the 204.2-keV transition. In both cases, with the 768-keV transition of E2 multipolarity, the data are consistent with multipolarity assignments of M1 or E2 for the 788-keV transition and E1, M1, or E2 for the 822-keV transition. No limits can be set on the multipolarities of the 763- and 784-keV transitions because of their low intensities.

8. Energy-Level Scheme of Mo⁹⁵

Figure 2 represents the energy-level scheme, with spin assignments and approximate transition intensities in terms of percent of total decay included (assuming the percent decay to the ground state and first excited state is negligible). The relative intensities for the 768-, 788-, and 822-keV transitions were obtained from the relative conversion-electron intensities by normalizing the data to the conversion-electron and gamma-ray intensities of the 584-keV transition by using the theoretical M1 conversion coefficient. The intensity of the 822-keV transition was determined by assuming M1 or E2 multipolarity. The justification of this assignment in preference to E1 is given later in this section. The approximate maximum limits on the intensities of the 763- and 784-keV transitions were determined by assuming the maximum photon intensity, that is, the intensity if both the transitions were E1.

From the observed intensities of the transitions populating the first excited state and the intensity of the 204.2-keV transition, an

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Fig. 2. Decay scheme of Tc^{95m} . Transition intensities are given as percent of total decay.

upper limit of the electron capture to the first excited state can be set as 10%, and the data are consistent with no capture to this state. Because of the larger uncertainty of the K x-ray intensity we can only set the less restrictive upper limit of the electron capture to the ground state at 20%. These limits are consistent with those of Levi *et al.*, who have reported less than 10% electron capture to the ground state and less than 10% to the first excited state.¹³

Table IV lists the experimental log ft values for electron capture to the various energy levels. Levi *et al.* have reported observing approximately equally intense positron groups of 680 ± 30 and 460 ± 30 kev populating the ground state and the first excited state of Mo^{95} respectively.¹³ Assuming equally intense positron groups, we calculate the log ft values for positron emission of 9.0 and 8.2 for the ground state and the 204.2-kev level respectively.

Table IV. Experimental log ft values for E.C.

Energy level	Log ft
0	≥ 7.98
204	≥ 8.17
763	≥ 8.5
784	≥ 8.1
788	7.02
822	7.98
1042	6.92

The ground state of Mo^{95} has a measured spin of $5/2$,^{26,27} in agreement with the shell-model prediction of $5/2$ due to a $(d_{5/2})^3_{5/2+}$ neutron configuration.

The 204.2-kev transition is primarily of M1 multipolarity, which indicates that the first excited state could have a spin of $3/2+$, $5/2+$, or $7/2+$. However, from angular distribution and polarization-direction correlation, McGowan and Stelson have shown that the spin of the first excited state must be $3/2+$.²¹

From this work alone little can be said conclusively regarding the spin and parity of the energy level at 768 kev, but from other work on the decay of the ground state of Nb⁹⁵ the spin has been shown to be 9/2+. ²⁴ In this work, this energy level is probably not populated directly from the decay of the p_{1/2} isomeric state of Tc⁹⁵, but rather from the decay of the g_{9/2} ground state of Tc⁹⁵, which is in turn populated by the isomeric transition. Medicus et al. have reported that 90% of the ground state decay of Tc⁹⁵ goes to the 768-kev level. ¹² Using this percentage, taking the intensity of the 768-kev transition from this work -- as calculated by using the theoretical E2 conversion coefficient -- and assuming no population from the isomeric state, we calculate that approximately 4% of the decay of Tc^{95m} proceeds through the isomeric transition. The value reported by Medicus et al. is 3%. Therefore very little, if any, direct population of this state comes from the 1/2- isomeric state of Tc⁹⁵, as would be expected.

The energy levels at 788 and 1042 kev decay primarily to the 3/2+ first excited state, with an appreciable branching to the 5/2+ ground state. It should be noted that the log ft values and the modes of de-excitation are very similar for these two states. For both these states, spin values of 1/2+ or 3/2+ would be consistent with the modes of depopulation, the assignment of M1 multipolarity for the 584-kev transition, M1 or E2 multipolarity for the 788- and 837-kev transitions, and the log ft values for these states.

The 822-kev transition has been shown to be of either E1, M1, or E2 multipolarity. The occurrence of an odd-parity state at an energy such as this and in this region just three neutrons from a closed shell is unlikely. The log ft value of 7.6, calculated by assuming that the 822-kev transition is of E1 multipolarity, is inconsistent with most log ft values for allowed or second-forbidden transitions. However, the log ft value of 7.98 calculated by assuming M1 or E2 multipolarity is consistent with an assignment of $\Delta I = \pm 1$, or 2 yes for the electron capture to this state. On this basis, the energy level at 822 kev is assigned tentatively as 3/2+ or 5/2+.

The 763- and 784-keV transitions have been tentatively assigned as going directly to the ground state. This assignment is not completely certain because of the low intensity of these transitions. Again, on the assumption that these transitions are both of E1 multipolarity, the calculated log ft values of ≈ 8.5 and ≈ 8.2 for the 763- and 784-keV transitions respectively are not consistent with allowed or second-forbidden transitions. On the other hand, the log ft values of ≈ 8.9 and ≈ 8.6 respectively (assuming M1 or E2 multipolarity) are consistent with $\Delta I = \pm 2$ yes. It should be pointed out at this time that we have not ruled out the possibility that these transitions are populated in the decay of the ground state of Tc^{95} . Medicus et al., in their work, probably could not resolve the transitions of 763 and 768 keV.

9. Discussion

The decay of the $p_{1/2}$ isomeric state and the $g_{9/2}$ ground state of Tc^{95} are very interesting, since in each case a different set of energy levels of Mo^{95} is populated owing to the large difference in the spins of the two initial states. In this work it is shown that the decay from the isomeric state is rather complex, but according to the work of Medicus et al., the decay of the ground state is rather simple.¹² Most of the electron capture from the ground state goes to the energy level at 768 keV, and there is small branching to levels at 930 and 1070 keV. In the beta decay of the ground state of Nb^{95} only the level at 768 keV in Mo^{95} has been observed to be populated.

The $3/2+$ first excited state of Mo^{95} at 204.2 keV is rather difficult to interpret on the basis of an extreme single-particle model as a state formed by the excitation of the unpaired neutron of the ground-state configuration. The energy difference of 204.2 keV between the $(d_{5/2})^3_{5/2+}$ ground state and the $3/2+$ first excited state is much smaller than the normal $d_{5/2}-d_{3/2}$ splitting, and therefore the first excited state cannot be due to a neutron configuration such as $[(d_{5/2})^2_0 (d_{3/2})^1]_{3/2+}$. The log ft value of ≥ 8.2 for electron capture of the $p_{1/2}$ isomeric state of Tc^{95} to this level is rather high for a simple first-forbidden $\Delta I = 1$ transition (log ft ≈ 7). Also, this

excited state has a measured half-life of 7.7×10^{-10} sec.^{28,29} From the E2/M1 gamma-ray mixing ratio of 0.34 and the theoretical M1 and E2 conversion coefficients of Sliv, the M1 gamma-ray half-life is calculated to be 1.1×10^{-9} sec. From Moszkowski's single-particle estimates³⁰ (using the statistical factor) the half-life of a M1 single neutron transition is 2.3×10^{-12} sec; that is, the M1 component of the gamma ray is retarded by a factor of ≈ 480 .

The abnormally high log ft value and the long lifetime of this state may be understood if the state is due to three unpaired neutrons coupled in the configuration $[(d_{5/2})^3]_{3/2+}$. This type of coupling of three $d_{5/2}$ particles to yield a resultant spin of 3/2 has been observed previously for the ground states of Na^{23} and Ne^{21} . Kurath³¹ and Talmi³² have shown theoretically, by using interaction potentials intermediate to the "long-range" and delta-type interactions, that the $[(d_{5/2})^3]_{5/2+}$ configuration in this region is of lower energy than the $[(d_{5/2})^3]_{3/2+}$ configuration. Figure 3 (taken from an article by Moszkowski³³) shows the excitation spectra for configurations of three interacting $J = 5/2$ nucleons. Figure 3a shows the spectra for a short-range interaction, i.e., the range of the nucleon-nucleon interactions is much smaller than the nuclear radius. Figure 3b shows the spectra for a long-range Serber interaction. The energy of a state of angular momentum I is denoted by E_I . The term $E_{\nu=0}$ refers to the energy in the absence of any interaction. All energies were calculated on the assumption of j-j coupling. For the medium-weight nuclei the short-range force has been found to be a better approximation. If the $3/2+$ first excited state of Mo^{95} is due to a neutron configuration of the type $(d_{5/2})^3_{3/2+}$, the electron capture to this excited state must occur by the transformation of a $p_{1/2}$ proton to a $d_{5/2}$ neutron. This process would be classified as a $\Delta I = 2$ (yes) transition and would be expected to have a higher log ft value than the first forbidden $\Delta I = 0$ or 1 (yes) transitions. Empirically the $\Delta I = 0$ or 1 (yes) transitions have log ft values of approximately 7 and the $\Delta I = 2$ (yes) transitions have log ft values of 8 or 9. The large log ft value of ≥ 8.2 experimentally determined for the electron capture of

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Fig. 3. Excitation spectra for three interacting $j = 5/2$ nucleons: (a) refers to short range force, (b) to long range force.

the $p_{1/2}$ isomer of Tc^{95} to the $3/2+$ excited state is therefore strong evidence in favor of a $(d_{5/2})^3_{3/2+}$ configuration and indicates that the amount of $d_{3/2}$ mixing in this excited state is small.

Any discussion as to the nature of the higher-lying energy levels at this time would be unwarranted because of the uncertainty of the spin assignments and also because of the inadequacy of present-day nuclear theory regarding excited states of nuclei in this mass region.

10. Recent Results

Several months after this work was submitted for publication in Physical Review, another paper on the decay of Tc^{95m} was published by Levi, Papineau, Redon, and Saunier.³⁴ In this paper they report some gamma-ray studies performed with NaI scintillation crystals. In column 1 of Table V are listed the gamma-ray energies reported by them, in column 2 the relative intensities, in column 3 their gamma-ray intensities normalized to the 570-keV gamma ray, and in column 4 the corresponding relative intensities obtained in this work. The 770-keV gamma ray in their work consists of the gamma rays of the 820-keV complex gamma-ray peak which are not in coincidence with the 204-keV transition. No limits of error on the gamma-ray energies and intensities were reported by Levi et al.

Table V. Comparison of gamma-ray data of Levi et al. with those obtained in this work

Gamma-Ray energy, (Levi <u>et al.</u>) (keV)	Relative gamma-ray intensity (Levi <u>et al.</u>)	Normalized gamma-ray intensity	Relative gamma-ray intensity (this work)
203	0.95	1.9	1.8 ± 0.2
570	0.49	1.0	1.0
770	0.35	0.71	0.68 ± 0.17
810	0.42	0.86	0.77 ± 0.08
1010	0.055	0.11	0.10 ± 0.02

Levi et al. report the total positron-to-electron-capture ratio as 2.6×10^{-3} , in agreement with the $4.0 \pm 1.6 \times 10^{-3}$ determined in this work. This new value from Levi et al. for the total positron-to-electron-capture ratio resolves the difference between the ratio previously reported by them and the ratio determined in this work and in the work of Medicus et al.¹²

Levi et al. determined that the ground state and the first excited state of Mo^{95} are each populated by 5.5% of the electron capture of $\text{Tc}^{95\text{m}}$. The log ft values for electron capture to the ground state and the first excited state based on these percentages are 8.5 and 8.4 respectively. The similarity of these two log ft values is again good evidence that both these states are formed by the transformation of a $p_{1/2}$ proton to a $d_{5/2}$ neutron.

Levi et al. also obtained anisotropies of -0.21 and -0.22 for the angular distributions of the 584-204- and 837-204-keV gamma-ray cascades. They interpret these results as conclusive evidence that the total angular momenta of the 788- and 1042-keV excited states are $1/2$. The expected anisotropy for the spin sequence $1/2(L1)3/2(L2)5/2$, where L1 (the 584- or 837-keV transition) is a pure M1 transition and the 204-keV transition L2 consists of M1-E2 multipolarities with $(\frac{E2}{M1})^{1/2} = -0.58$,²¹ is -0.24. However, they do not discuss the fact that the experimental anisotropies are also consistent with total angular momenta assignments of $3/2$ for the 788- and 1042-keV excited states and $\frac{E2}{M1}$ gamma-ray mixing ratios of 0.4 or 8.5 for the 584- and 837-keV transitions. To this date, there is no accurate determination of the amount of E2 contribution in these two transitions. The 584-keV transition has been shown to be predominantly of M1 multipolarity in this work, eliminating the possibility of the gamma-ray mixing ratio of 8.5 for the 584-keV transition. However, more precise determinations of the amounts of E2 multipolarities in the 584- and 837-keV transitions must be made before unambiguous assignments of the angular momenta of these excited states can be made.

C. Decay of the Isomer of Tc^{97}

Cork, Brice, Schmid, and Helmer³⁵ have reported the decay scheme for the 90-day isomer of Tc^{97} shown in Fig. 4 from a study of the electron capture of Ru^{97} .

Fig. 4. Decay scheme of Tc^{97m} as reported by Cork et al.³⁵

These assignments were based on the observation of two highly converted transitions in a conversion-electron spectrum. The K conversion-electron intensities of these two transitions were reported to be about equal. No half-life determination was made by Cork et al. on these conversion lines.

In this study the K, L, and M conversion-electron lines of a very highly converted transition in technetium of 96.5 ± 0.1 kev were observed. No other conversion-electron lines were observed in this energy region. As a basis for assigning this transition to Tc^{97m} , the half-life of the conversion-electron lines was determined over a period of one year. The half-life observed was 87 ± 3 days, in good agreement with the reported half-life of Tc^{97m} of about 90 days.¹⁵

The experimental K : L : M relation conversion-electron intensities are $1 : 0.48 \pm 0.05 : 0.13 \pm 0.02$. The K/L ratio of 2.1 ± 0.2 agrees very well with the theoretical ratio of 2.0 for an M4 transition.

The gamma-decay transition probability was also calculated by using the theoretical K-conversion coefficient, the theoretical K/L

ratio, the experimental K/M ratio, and the experimental half-life of 87 ± 3 days. This decay constant is compared with the value calculated from the estimates by Moszkowski³⁰ (M) by use of the appropriate statistical factor, and also with the empirical equation of Goldhaber and Sunyar³⁶ (G and S), in Table VI.

Table VI. Comparison of gamma-decay transition probabilities

	$\lambda \text{ sec}^{-1}$
Experimental	2.9×10^{-10}
M	5.1×10^{-9}
G and S	3.4×10^{-10}

III. LIFETIME MEASUREMENT OF THE 74.6-Kev STATE IN Np^{239}

A. Introduction

In addition to the determination of the total angular momenta and parities of excited states of nuclei, the measurement of gamma-ray transition probabilities between excited states has proved very valuable in testing the validity of various models of nuclear structure. Not only does the gamma ray transition probability depend on the angular momentum carried off by the gamma ray and on the parities of the initial and final states, but it also depends quite strongly on the selection rules of other quantum numbers describing the nuclear states. For example, in the approach of Bohr and Mottelson,³⁷ used to describe the spectra of spheroidal nuclei, transitions forbidden in the K selection rule (where K is the projection of the total angular momentum on the nuclear symmetry axis) have gamma-ray transition probabilities that are slower than that expected for simple single-proton transitions by as much as a factor of 10^{15} .

In addition to the K selection rule, gamma-ray transitions may also be forbidden by selection rules in the asymptotic quantum numbers, which characterize intrinsic properties of the wave functions used to describe the nuclear states in the region of spheroidal deformation. These selection rules have been derived by Alaga³⁸ and others³⁹⁻⁴¹ from the wave functions of Nilsson,⁴² who considered the nucleons as moving in an anisotropic harmonic oscillator potential generated by all the particles. In the limit of very large nuclear deformations N, the principal harmonic oscillator shell, n_z , the number of oscillator quanta along the nuclear symmetry axis, and Λ , the projection of the orbital angular momentum of the odd nucleon along the symmetry axis, become constants of the particle motion. However, because of the approximations involved, these quantum numbers are not truly constants of motion, and it has been shown that gamma-ray transitions forbidden by these selection rules generally are slower than the allowed transitions by an order of magnitude.

In connection with the hindered electric dipole gamma-ray transitions in spheroidal nuclei forbidden by asymptotic quantum-number selection rules, it has been shown by Nilsson and Rasmussen⁴³ and also by Asaro, Stephens, Hollander, and Perlman⁴⁴ that in some cases the experimentally determined internal conversion coefficients for these transitions differ greatly from the theoretical coefficients given by Sliv and Band.²⁵ Furthermore, it has been shown by Asaro et al. that the magnitude of this anomaly is proportional to the retardation of the gamma-ray life-time over that calculated from single-proton transition formulae.

One of the cases cited by Asaro et al. is in Np²³⁷. The half-life of the 59.6-keV E1 gamma ray depopulating an excited state of that energy is 3.1×10^5 times the half-life calculated from the formula of Moszkowski,³⁰ and the L_1 and L_{11} conversion coefficients are much larger than the theoretical estimates (see Table VII).

Table VII. Conversion coefficients for E1 transitions in Np²³⁷ and Np²³⁹ (data taken from Asaro et al.⁴⁴)

Nucleus	Photon energy (keV)	Experimental			Theoretical (Sliv)		
		$\alpha(L_1)$	$\alpha(L_{11})$	$\alpha(L_{111})$	$\alpha(L_1)$	$\alpha(L_{11})$	$\alpha(L_{111})$
Np ²³⁷	59.6	0.22±0.02	0.46±0.05	0.12±0.03	0.13	0.12	0.13
Np ²³⁹	74.6	0.08±0.02	0.06±0.02	0.06±0.02	0.084	0.066	0.063

The 74.6-keV E1 transition in the near-by isotope Np²³⁹ depopulates an excited state of that energy and goes to the ground state of Np²³⁹. Both the state at 74.6 keV and the ground state of Np²³⁹ have been identified as being the same odd-proton states as the 59.6-keV excited state and the ground state, respectively, of Np²³⁷. The E1 gamma-ray transitions in both cases are forbidden, since the selection rules require that $\Delta N = \pm 1$, $\Delta n_z = \pm 1$ and $\Delta \Lambda = 0$ or $\Delta n_z = 0$ and $\Delta \Lambda = \pm 1$, while for these transitions $\Delta N = -1$, $\Delta n_z = -2$, and $\Delta \Lambda = 1$.

Figure 5 shows the partial decay schemes of the two nuclei Am²⁴¹ and Am²⁴³ populating the nuclei of interest. Particular attention should be paid to the great similarity of the excited states in both cases.

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Fig. 5. Partial decay schemes of Am^{241} and Am^{243} .

In Table VII we see that while the L_1 - and L_{11} -internal-conversion coefficients of the 59.6-keV transitions in Np^{237} are definitely anomalous, the internal-conversion coefficients of the same transition in Np^{239} are (within experimental error) in agreement with the theoretical values. From the upper limit of 1.6×10^{-9} sec for the half-life of the 74.6-keV state in Np^{239} determined by Strominger,⁴⁵ one can calculate that the gamma-ray half-life is hindered by less than a factor of 7.6×10^3 from the single-proton estimates, compared with a factor of 3.1×10^5 for Np^{237} .

The following material describes the actual measurement of the total half-life of the 74.6-keV state in Np^{239} by the delayed coincidence method and the calculation of the retardation of the 74.6-keV gamma ray in order to provide more quantitative information to be used in the study of the dependence of the anomalous $E1$ internal-conversion coefficients on the gamma-ray hindrance.

B. Experimental Techniques

1. Coincidence Circuit

The lifetime of the 74.6-keV excited state in Np^{239} was measured by studying the time relationship between the emission of the alpha particles in the decay of Am^{243} and the emission of the 74.6-keV gamma rays depopulating the 74.6-keV excited state in Np^{239} . A block diagram of the coincidence circuit used is shown in Fig. 6. The radioactive sample is placed between two scintillation crystals. One crystal is a 1-inch-diameter by 3/16-inch-thick sodium iodide (thallium activated) crystal which is used to detect the 74.6-keV gamma rays. The other crystal, which was used to detect the alpha particles, was a 1.5-cm-diameter by 0.1-cm-thick Livermore plastic scintillator.* The light output of the scintillators produced by the gamma rays or alpha particles was converted to electronic pulses by using RCA 6655A photomultiplier

*This plastic scintillator is composed of styrene with 2.5% terphenyl, 0.03% tetraphenyl butadiene, and 0.01% zinc stearate.

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Fig. 6. Block diagram of millimicrosecond coincidence circuit.

tubes. To minimize the transit-time spreads of the photoelectrons between the photocathode surface and the first dynode, the diameters of the scintillation crystals were chosen so that their light output impinged only on the concave portion of the photocathode surface of the photomultiplier tube. To reduce electron transit-time spreads between the various dynodes of the phototube, the phototubes were used at a rather high voltage -- 1400 volts. At this voltage the phototubes were very stable over long periods of time for moderate counting rates.

Two pulses are taken from each photomultiplier tube. The pulses used for the "slow" (energy) analysis are taken from the eighth dynode. These pulses are amplified by DD-2 linear amplifiers and passed through single-channel pulse-height analyzers, and the outputs of the single-channel pulse-height analyzers are fed into a slow-coincidence circuit with a resolving time of 2 μ sec. The single-channel analyzers were set so that their outputs corresponded only to pulses in which the radiations deposited their entire energies in the crystals.

The pulses used for the "fast" coincidence analysis are taken directly from the anodes of the photomultiplier tubes, passed through Hewlett-Packard 460-A or -B wide-band amplifiers and into a time-to-pulse-height converter. The time-to-pulse-height converter has a pulse output whose voltage is inversely proportional to the time difference between two input pulses arriving within a given adjustable time interval. (The time-to-pulse-height converter is discussed in the following section.) The output of the time-to-pulse-height converter is amplified by a DD-2 linear amplifier and then passed into a 100-channel pulse-height analyzer as the signal pulse. The output of the slow-coincidence circuit serves as a gate pulse which activates the 100-channel analyzer. In order to have a count recorded on a channel of the analyzer, thus giving the time relationship between an alpha particle and a gamma ray, a signal pulse from the time-to-pulse-height converter and a gate pulse from the slow-coincidence circuit must arrive at the analyzer within 5 μ sec of each other. In order for this to occur a triple coincidence is required, i.e., a pulse from the time-to-pulse-height converter and

the two pulse outputs from the single-channel analyzers corresponding to the appropriate energies deposited in the scintillation crystals must occur within a given short time interval, on the order of a few microseconds. The resolving time of the entire system is clearly dependent on the resolving time of the time-to-pulse-height converter, for if two pulses occur separated by a time that is longer than the resolving time of the time-to-pulse-height converter, no counts are obtained on the multichannel analyzer.

The calibration of the pulse-height output of the time-to-pulse-height converter displayed on the 100-channel pulse-height analyzer in terms of time was obtained by determining the time spectra of any two radiations in reasonably "prompt" coincidence as a function of the time by which signals from one of the detectors is artificially delayed relative to the other. This artificial delay was introduced by passing one of the radiations through variable known lengths of RG-63/U 125-ohm cable. From the known length of this cable and its propagation velocity (the ratio of the velocity of a pulse in the coaxial cable to the velocity of light) of 0.84, one can calculate the time of the introduced delay. The delays introduced by the different lengths of coaxial cable were also checked experimentally. Figure 7 shows such a time calibration using the alpha particles of Am^{243} in coincidence with the 74.6-keV gamma rays. The centroids of the curves were used to define the position of each curve.

2. Time-to-Pulse-Height Converter

Figure 8 shows a schematic of the time-to-pulse-height converter that was used in this work. The two negative inputs arriving at I1 and I2 are stretched by the RC circuit, consisting of the input capacitances of V1 and V2 and the resistances R1 and R1', with a time constant of 0.7 μsec , in order to provide a more uniform pulse input for the Amperex E180F limiting pentode tubes V1 and V2. This stretching procedure was found to improve the time resolution when sodium iodide was used as a scintillator. The negative input pulses cut off the E180F tubes, which are normally conducting, producing a positive pulse at the common plates

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Fig. 7. Time calibration of 100-channel pulse-height analyzer with Am^{243} ; 0.494 mμsec/channel.

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Fig. 8. Time-to-pulse-height converter I.

of the tubes. The current in V1 is adjusted by means of the grid G1 so that the positive pulse output of V1 is of the same amplitude as that of V2. The widths of the pulse outputs of V1 and V2 are determined by the length of the common shorted clipping stub S. The positive pulses are then fed to the diode D2, which is biased to reject single pulses from V1 and V2. In order to understand more easily how the circuit works let us consider the two pulses shown in Figures 9a and 9b.

Let us assume that pulse "a" at V1 is due to the 74.6-keV gamma ray in the decay of Am^{243} and pulse "b" at V2 is due to an alpha particle, and let us also assume that the gamma ray is emitted immediately after the alpha particle. The pulse due to the alpha particle has been delayed artificially relative to that of the gamma ray by the time $t_2 - t_1$. The pulse at the common plate connection appears as shown at 9c. The diode D2 is biased at V_0 volts so that only the coincidence pulse shown in 9d will pass through the diode. It is clear that the width of the pulse in 9d is proportional to the overlap of the pulses a and b and therefore related to the time difference between the two radiations. The pulse d is then integrated by means of the RC circuit consisting of the resistance R2 and the capacitance C1. This circuit has a time constant very long relative to the maximum pulse width used, $t_3 - t_1$, and for times of the order of the pulse width the voltage rise at point F for a square pulse input is proportional to the length of the input signal. Therefore, the pulse height at F of Fig. 8 is proportional to the pulse width of 9d, and therefore related to the time difference between the two pulses a and b. Tubes V3 and V4 are simply used as an amplifier and cathode follower.

If the 74.6-keV excited state has a rather long half-life the probability of a gamma ray being emitted at a time T after the emission of an alpha particle decreases exponentially with T. In Fig. 9a this will mean that some gamma rays will appear later than T_1 relative to T_2 and will yield pulses with a larger pulse height than the prompt event. This is true assuming that $T_2 - T_1$ is much longer than the half-life of the state. The resultant spectrum on the 100-channel analyzer in this case is an asymmetric peak with a tail on the side corresponding to larger pulse-height outputs from the time-to-pulse-height converter.

Fig. 9. Pulse shapes in time-to-pulse-height converter I.

In a coincidence circuit such as this, chance events displayed on the 100-channel analyzer simply appear as a constant number of events for all pulse heights, so that the number of chance events can be determined very easily.

3. General Considerations of Delayed Coincidence Curves

If two radiations are "prompt" -- i.e., the time interval between the two is much too short to be measured with the coincidence circuit ($\sim 10^{-11}$ sec in this case) -- one obtains a "prompt" coincidence curve, the response function of the coincidence circuit, which we shall call $P(x)$. The variable x denotes the artificial delay that has been introduced. If, on the other hand, the intermediate state has a measurable half-life and the daughter emission has a probability $\frac{1}{\tau} e^{-\frac{t}{\tau}} dt$ of occurring t seconds after the decay of the parent (where τ is the mean life of the intermediate state, which is equal to $[\ln 2]$ times the half life), then, as Newton has shown,⁴⁶ for $F(x)$ (the delayed coincidence curve) and $P(x)$ normalized to the same area one has

$$\frac{d}{dx} [\ln F(x)] = -\frac{1}{\tau} \left\{ 1 - \left[\frac{P(x)}{F(x)} \right] \right\}. \quad (2)$$

In the limit for inserted delays greater than x_0 , where $\frac{P(x_0)}{F(x_0)} \ll 1$, the reciprocal of the mean life of the intermediate state can be obtained from the slope of the curve of $\ln F(x)$ as a function of x . If the half-life of the intermediate state is comparable to the resolving time of the coincidence circuit and $P(x_0)/F(x_0)$ is not very much less than unity, then an analysis as described by Bay⁴⁹ must be used. Bay has shown that the centroid of $F(x)$ is delayed relative to the centroid of $P(x)$ by the mean life of the intermediate state. So for very short half-lives (for this coincidence circuit these half-lives are less than a few millimicroseconds) the mean life can be taken directly from the difference of the centroids of the prompt- and delayed-coincidence curves.

Unfortunately the time displacement of the pulses at the diode D2 relative to the gating event in the scintillator is a function of the rise times of the initial pulses from the scintillators to a given voltage.

The rise times defined in this way are energy-dependent. Again referring to Fig. 8, let us assume that we are comparing two different prompt-resolution curves and let us assume that both radiations at b have the same energy and the radiations at a have different energies. The lower-energy radiation at a has a longer rise time and hence seems to appear later than the other radiation relative to b, and as a result of this its prompt-coincidence curve appears to be delayed relative to the other prompt-coincidence curve even though both cases are really prompt. In the choice of a sample to be used as the prompt-coincidence event for the comparison, this fact makes it necessary to have the energies of the two radiations as close as possible to those of the delayed event under investigation. This criterion, in addition to having the material reasonably available and requiring one to actually know that the two radiations in the prompt event are prompt ($\leq 10^{-11}$ sec in this case), has imposed serious limitations on the measurements of half-lives by this method.

4. Am²⁴³ Samples

A sample of Am²⁴³ of unknown origin was obtained and a gamma-ray spectrum and an alpha spectrum showed that it contained Am²⁴³ in equilibrium with its daughter Np²³⁹, and also that a small amount of Cm²⁴⁴ was present. The Cm²⁴⁴ does not interfere with the measurements, since essentially none of its gamma rays are accepted by the single-channel analyzer. The plastic scintillator used to detect the alpha particles is not good for discriminating against electrons, and it was found that electrons of roughly 200 kev gave the same pulse height as the 5.3-Mev alpha particles of Am²⁴³. Thus there is a possibility that the beta-emitting daughter of Am²⁴³, Np²³⁹, could contribute to the coincidence counting rate. However, this contribution is very small, since less than 20% of the gamma rays accepted and less than 10% of the total activity on the alpha side selected by the single-channel analyzers could have come from the decay of Np²³⁹. Also, most of the gamma rays accepted from the decay of Np²³⁹ would be from the 106.1-kev gamma ray which depopulates a level at 391.8 kev in Pu²³⁹. This excited state has a

half-life of 1.9×10^{-7} sec, and the coincidence events due to beta particles in coincidence with this gamma ray would give a time spectrum so long relative to the time interval displayed on the 100-channel analyzer that it would appear as an almost constant background and could be easily subtracted.

By determining the coincidence counting rates as a function of inserted absorber thickness that the alpha and beta particles have to traverse, it was found that only $\approx 5\%$ of the coincidence events in the delayed coincidence curve were due to beta-gamma coincidences. To show that this 5% did not affect the measurements, several determinations of the half-life were made using an Am^{243} sample from which the neptunium daughters had been chemically separated. This separation was done in 6 M HCl with a Dowex-50 cation-exchange column, as described by Diamond et al.⁴⁸ This neptunium-free sample gave the same results as the samples in equilibrium with neptunium.

The centroids of the various coincidence curves were independent of the counting rates: samples from five to ten times as active as the samples generally used showed no appreciable shift in centroids.

C. Results

Since the previous work of Strominger has shown that the half-life of the 74.6-kev excited state in Np^{239} is less than 1.6×10^{-9} sec, the half-life must be measured by the method of centroid shifts, since the resolving time of our coincidence circuit is $\approx 4 \times 10^{-9}$ sec for radiations of energies similar to those in the decay of Am^{243} . A search of the available alpha-particle emitters showed that no really good "prompt"-coincidence standard of the appropriate energies and very short half-life of the intermediate state was available. However, it was decided to measure the half-life of this state relative to the transitions in the decay of Th^{230} . The half-life of the intermediate state in this case has been measured by Vartapetian and Foucher⁴⁹ and found to be $0.63 \pm 0.07 \times 10^{-9}$ sec. The energy of the alpha particles of Th^{230} are 4.6 Mev, and the energy of the gamma ray is 67.8 kev, compared with

the values of 5.3 Mev and 74.6 kev respectively in the decay of Am^{243} . To minimize the shifts in the centroids due to the different energies of the radiations, the single-channel analyzers were set to accept a very narrow energy interval so that the pulse height analyzed for both samples would be almost identical. Figure 10 shows a typical spectrum obtained for the Am^{243} and Th^{230} delayed-coincidence curves. An average of eleven such comparisons showed that the half-life of Am^{243} relative to that of Th^{230} was $0.88 \pm 0.10 \times 10^{-9}$ sec. Using the value of $0.63 \pm 0.07 \times 10^{-9}$ sec for the half-life of the state in Ra^{226} , one obtains a half-life for the 74.6-kev excited state in Np^{239} of $1.51 \pm 0.12 \times 10^{-9}$ sec. This value is consistent with the previous upper limit determined by Strominger. However, in this work, by a careful analysis of the slopes of the delayed-coincidence curves it was shown that the half-life could be no longer than 1.5×10^{-9} sec. This would seem to indicate that the value obtained by using the reported half-life of the state in Ra^{226} may be a little high.

As a second comparison Bi^{207} was used as a prompt-coincidence standard. Bi^{207} is a long-lived electron-capture isotope populating excited states in Pb^{207} . Between these excited states there are two intense transitions of 569.7 and 1064 kev which are in coincidence. The Bi^{207} prompt-coincidence curve was determined by measuring coincidences between the high-energy K internal-conversion electrons of the 569.7- and 1064-kev transitions and the 73-kev K x-rays associated with the internal conversion in the K shell. The coincidences between the internal-conversion electrons and the K x-rays is prompt. A hole in the K-electron shell produced by the internal conversion process decays predominantly through K x-ray radiation. The lifetime for this type of process in gold is 10^{-17} sec, and should be even shorter in lead.⁵⁰

The single-channel analyzer, analyzing the pulses from the plastic scintillator, was set on the peak of the alpha particle distribution of Am^{243} , selecting only a narrow energy interval so that the conversion electrons of Pb^{207} and alpha particles of Am^{243} selected when the two samples were interchanged gave essentially the same pulse heights. Using the internal-conversion electrons in place of alpha particles for

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Fig. 10. Delayed coincidence curves for Am^{243} and Th^{230} compared to Bi^{207} .
 ○ Am^{243} □ Th^{230} △ Bi^{207} .

the comparison is possible in this case, since both particles at these energies give roughly the same pulse heights in the plastic scintillator. Also Bi^{207} is a good comparison, since the K-x-rays of lead (73 kev) have very nearly the same energy as the gamma ray in Np^{239} (74.6 kev). In Fig. 10 are shown the coincidence curves due to Am^{243} and Bi^{207} .

Less than 15% of the gamma-ray events accepted by the single-channel analyzer correspond to Compton events from the 570- and 1064-kev gamma rays, some of which may lead to a delayed coincidence with conversion electrons because the intermediate state has a very short but measurable half-life. With this upper limit of 15% of the coincidence events delayed and a half-life of 8×10^{-11} sec^{51,52} for the intermediate state, the centroid of the Bi^{207} curve would be delayed $\approx 2 \times 10^{-11}$ sec from a true prompt event. This uncertainty is negligible compared with the experimental uncertainties in the observed centroid shifts between the prompt and delayed curves. The measured half-life of the 74.6-kev state in Np^{239} relative to Bi^{207} was determined to be $1.2 \pm 0.1 \times 10^{-9}$ sec.

Because of the apparent disagreement between this value and the value obtained by using Vartapetian and Foucher's half-life of the state in Ra^{226} , the half-life of the state in Ra^{226} was compared with Bi^{207} as a check of consistency. The half-life obtained was $0.49 \pm 0.09 \times 10^{-9}$ sec, which is lower than the value reported by Vartapetian and Foucher; however, within the quoted experimental errors the two determinations do overlap. The half-life of the 74.6-kev state in Np^{239} based on this new value for the half-life of the state in Ra^{226} is $1.37 \pm 0.13 \times 10^{-9}$ sec, which is consistent with the value of $1.2 \pm 0.1 \times 10^{-9}$ sec obtained from a direct comparison to Bi^{207} .

A summary of this work is listed in Table VIII. The best value of the half-life of the 74.6-kev excited state in Np^{239} from this work is $1.2 \pm 0.1 \times 10^{-9}$ sec.

Table VIII. Summary of half-life measurements

Comparison	Half-life (μ sec)
1. Am ²⁴³ compared to Th ²³⁰ , using 5 below	1.51 $\pm$ 0.12
2. Am ²⁴³ compared to Th ²³⁰ , using 4 below	1.37 $\pm$ 0.13
3. Am ²⁴³ compared to Bi ²⁰⁷	1.2 $\pm$ 0.1
4. Half-life of the 68-kev excited state in Ra ²²⁶ compared to Bi ²⁰⁷ from this work.	0.49 $\pm$ 0.09
5. Half-life of the 68-kev excited state in Ra ²²⁶ compared to Au ¹⁹⁸ by V and F.	0.63 $\pm$ 0.07
Best value for half-life of 74.6-kev state in Np ²³⁹ from this work.	1.2 $\pm$ 0.1
Remeasured half-life for 68-kev state in Ra ²²⁶	0.49 $\pm$ 0.09

D. Contribution to the Observed Half-Life from the 118-kev Level

About 11.5% of the alpha decay of Am²⁴³ goes to an excited state at 118 kev in Np²³⁹ which is depopulated by a 43.2-kev M1-E2 transition to the 74.6-kev excited state and a 118-kev E1 transition to the ground state.⁴⁴ If this 118-kev excited state had a very long lifetime approximately 11% of the events in the delayed coincidence curve would be delayed owing to the half-life of the 118-kev excited state and also the 74.6-kev excited state. In this case the centroid of the delayed-coincidence curve would be delayed relative to the prompt curve by the mean life of the 74.6-kev excited state plus some additional contribution due to the lifetime of the 118-kev excited state. On the other hand, if the lifetime of the 118-kev state were very short, then all the coincidence events would have the lifetime of just the 74.6-kev state.

The half-life of the 118-kev excited state can be calculated roughly if one assumes that the 43.2-kev M1-E2 transition has the same amount of E2 mixing as the analogous 43.5-kev transition in Np²³⁷. If

we assume that the intrinsic quadrupole moment of Np^{239} is very close to the value of 9.0 barns measured by Newton⁵³ for Np^{237} , and use formula VII.18 of Bohr and Mottelson,³⁷ which relates the reduced electric-quadrupole-transition probability to the intrinsic quadrupole moment, we can calculate the E2 gamma-ray half-life of the 43.2-keV transition as 1.3×10^{-7} sec. The total theoretical E2 conversion coefficient is roughly 840, and using this we calculate the total E2 half-life as 1.54×10^{-10} sec. Assuming a total E2 admixture (photon plus conversion electrons) of 57%, as in Np^{237} ,⁵⁴ we find the total half-life of the 43.2 keV transition is 0.9×10^{-10} sec. From this half-life and the fact that of the 11.5% of the decays going to this state, 11% goes through the 43.2-keV transition and 0.5% through the 118-keV E1,⁴⁴ one can calculate that the total half-life of the 118-keV level is 0.85×10^{-10} sec. This calculation should be good to a factor of roughly two, and is consistent with the rest of the data. From (a) the half-life of the 118-keV gamma ray obtained in this way (b) the branching rules of Alaga et al. for transitions between members of one rotational band and the energy level of another,⁵⁵ (c) the experimental conversion coefficient of the 74.6-keV transition, and (d) the experimental branching ratio of the 74.6-keV excited state, one calculates a total half-life of 1.7×10^{-9} sec for the 74.6-keV excited state, very close to the value of $1.2 \pm 0.1 \times 10^{-9}$ sec determined experimentally. There is some question as to the applicability of the branching rules of Alaga et al., but their use seems justified in this case since the ratio of the reduced transition probability of the 43.1-keV E1 to the 74.6-keV E1 is predicted to be 0.40 and has been measured experimentally as 0.30 ± 0.08 .

Using the value of 1×10^{-10} sec for the half-life of the 118-keV excited state, and replacing the expression given by Bay with a complex decay, one can calculate that with 11% of the coincidence events delayed by 1×10^{-10} sec in addition to the half-life of the 74.6-keV excited state, the centroid of the delayed-coincidence curve will be affected by only $\sim 0.01 \times 10^{-9}$ sec, which is negligible compared with the quoted experimental errors.

E. Discussion

From (a) the measured half-life of the 74.6-keV excited state of $1.2 \pm 0.1 \times 10^{-9}$ sec, (b) the internal-conversion coefficient of 0.27 for the 74.6-keV transition (experimental $\alpha(L)$ of 0.20 ± 0.05 times an empirical factor of 1.35 to account for M, N, ... conversion), (c) the relative gamma-ray intensities of the 43.1-keV E1 and the 74.6-keV E1 (0.04 ± 0.01 and 0.69 ± 0.03 respectively),⁴⁴ and (d) the theoretical total conversion coefficient of 1.2 for the 43.1-keV E1 transition, one calculates the gamma-ray half life of the 74.6-keV transition to be $1.7 \pm 0.3 \times 10^{-9}$ sec. The half-life calculated from the formula of Moszkowski for a single-proton transition (omitting the statistical factor) is 2.9×10^{-13} sec. The retardation of this gamma ray, then, is greater than that due to a single-proton transition by a factor of $6 \pm 1 \times 10^3$.

The electric dipole transition between the same intrinsic states of Np^{237} and Np^{239} ($5/2 \ 5/2 - [523]$ and $5/2 \ 5/2 + [642]$), demonstrate rather convincingly the effect of the gamma-ray retardation on the internal conversion coefficients. In Np^{237} the gamma ray is retarded by a factor of 3.1×10^5 . The $L_1 : L_{11} : L_{111}$ subshell ratios are definitely anomalous, and the absolute L_1 conversion coefficient is 1.7 times and the L_{11} conversion coefficient 3.8 times the theoretical values of Sliv. In Np^{239} , where the retardation is only 6×10^3 , the $L_1 : L_{11} : L_{111}$ subshell ratios and the absolute conversion coefficients agree with the theoretical values within the experimental error of 20%.

The same intrinsic states appear in the energy levels of Am^{243} populated in the beta decay of Pu^{243} , but their order is reversed, the $5/2 \ 5/2 - [523]$ level being the ground state. In this case the 83.9-keV electric dipole transition is hindered by a factor of 1.3×10^4 . The experimental conversion coefficients are $\alpha(L_1) = 0.047 \pm 0.011$, $\alpha(L_{11}) = 0.057 \pm 0.013$, $\alpha(L_{111}) = 0.04 \pm 0.009$, and $\alpha(L) = 0.145 \pm 0.03$,⁵⁶ compared with the theoretical values of 0.068, 0.052, 0.046, and 0.166 respectively. Within the experimental error, the conversion coefficients agree with the theoretical ones. However, Stephens et al. have shown

that the L subshell ratios are anomalous.⁵⁶ Experimentally they found $L_1/L_{11}/L_{111} = 1.15/1.4/1.0$, compared with the theoretical $1.48/1.13/1.00$.

For these three cases in which the electric dipole transitions violate both the radiative selection rules and the internal-conversion selection rules,⁴³ anomalies do exist. The threshold as defined by Nilsson and Rasmussen⁴³ (retardation above which anomalies are greater than 50%) in this case is between retardations of 1.3×10^4 and 3×10^5 .

It is also interesting at this time to examine more closely the differences between the radiative half-lives of these electric dipole transitions. The expression for the rate of dipole gamma-ray emission is

$$\hat{T} (E1) = \frac{16 \pi}{9} \left(\frac{1}{\hbar^4 c^3} \right) E^3 B (E1). \quad (3)$$

Here $T (E1)$ is the transition probability in sec^{-1} ,

$\hbar$ is Planck's constant divided by 2π ,

c is the velocity of light,

E is the energy of the transition,

$B (E1)$ is the reduced transition probability.

The quantity $B (E1)$ contains all of the dependence of the transition probability on the details of the nuclear structure. For Np^{237} and Np^{239} one has the same proton configuration, but Np^{239} has two more neutrons in addition to the neutron configuration of Np^{237} . One would not expect a large change in $B (E1)$ in going from Np^{237} to Np^{239} just due to the addition of two neutrons. However, a very drastic change does occur. In Table IX are listed the electric-dipole reduced transition probabilities relative to Np^{239} .

Table IX. Relative reduced transition probabilities for electric-dipole transitions between the same intrinsic states in Np^{237} , Np^{239} , and Am^{243}

Nucleus	Gamma-Ray energy (kev)	Gamma-Ray half-life (sec)	Relative $B (E1)$
Np^{237}	59.6	1.75×10^{-7}	0.019
Np^{239}	74.6	1.7×10^{-9}	1
Am^{243}	83.9	2.6×10^{-9}	0.46

At present the difference between the reduced transition probabilities of Np^{237} and Np^{239} cannot be quantitatively explained. Strominger has pointed out that the rather erratic fluctuations in E1 transition rates from isotope to isotope can be qualitatively understood by the fact that in the calculation of the transition rates using the wave functions of Nilsson, the lifetimes depend rather sensitively on near cancellation of large terms in the matrix elements.⁴⁵ Therefore a small change in the wave function describing the proton state may drastically change the reduced transition probability.

IV. GAMMA-GAMMA-RAY ANGULAR DISTRIBUTION STUDIES IN THE BETA DECAY OF U^{237}

A. Introduction

The decay scheme of the 6.7-day beta emitter U^{237} taken from the work of Rasmussen, Canavan, and Hollander⁵⁷ is shown in Fig. 11. The work reported here was undertaken primarily for two reasons: (a) To study the dependence of the angular distributions of the gamma-ray cascades DC,CB and CB,BA on the chemical state of the systems, (b) To verify from measurements of these two angular distributions the assignment of the total angular momentum of energy level D as $1/2$ and to determine the E2 admixture in transition CB. As a by-product of these investigations, the half-lives of the excited states B and C were remeasured and an upper limit on the half-life of level D was determined.

The angular distribution of a gamma ray represents the probability per unit solid angle $W(\theta)$ for emission of a gamma ray at a given angle with respect to some fixed direction in the laboratory system. This direction in turn is related to the initial orientation of the total angular momentum of the nucleus. If the nuclei emitting gamma rays have their total angular momenta distributed randomly in space, then there exists no single fixed direction relative to the total angular momenta of the nuclei and the gamma rays are emitted isotropically. To obtain an anisotropic angular distribution one must introduce a means by which only nuclei whose total angular momenta are oriented in a particular direction will be considered.

In studies of gamma-gamma directional correlations, the coincidence counting rate of two gamma rays emitted successively from a particular nucleus (as shown in the hypothetical case of Fig. 12) is measured as a function of the angle between the two gamma rays.

Figure 12. Hypothetical gamma-ray cascade

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Fig. 11. Energy-level scheme of Np^{237} .

The selection of the direction of one gamma ray as in the direction of the first counter defines the orientation of the total angular momentum of the intermediate state relative to this direction (assuming that the intermediate state j has an angular momentum different from zero or one-half). The events recorded in the second gamma-ray counter as a function of the angle θ which are in coincidence with the gamma rays of the first counter represent the angular distribution of gamma rays emitted from nuclei whose orientation has been defined by the first gamma ray.

The directional correlation for a gamma-ray cascade in which one gamma ray consists of the multiplicities L_1 and $L_1 + 1$ and the other gamma ray is of single multiplicity L_2 is given as

$$W(\theta) = 1 + \sum_K A_K P_K(\cos \theta), \quad (4)$$

$$A_K = \frac{1}{1+\delta^2} \left[F_K(L_1 j_1 j) F_K(L_2 j_2 j) + \delta^2 F_K(L_1 + 1 j_1 j) F_K(L_2 j_2 j) \right]$$

$$+ 2\delta (-1)^{j-j_1-1} ([2j+1] [2L_1+1] [2L_1+2])^{1/2} G_K(L_1 j_1 j) F_K(L_2 j_2 j),$$

where $P_K(\cos \theta)$ is the K th Legendre polynomial,

$K = 2, 4, 6, \dots, K_{\max}$, where $K_{\max}$ minimum $(2j, 2(L_1+1), 2L_2)$,

$$\delta^2 = \frac{\text{intensity of } (L_1+1) \text{ multipolarity photon}}{\text{intensity of } (L_1) \text{ multipolarity photon}},$$

$$\delta = \frac{(j_1 \parallel L_1+1 \parallel j)}{(j_1 \parallel L_1 \parallel j)}.$$

The F_K and G_K coefficients and the phase of δ are given by Biedenharn and Rose.⁵⁸ The more general expression for $W(\theta)$ for which both transitions are of mixed multiplicities is given by Rose.⁵⁹ Throughout this work reference will be made to the "gamma ray anisotropy" or more simply "anisotropy" which is defined as $[\frac{W(180^\circ)}{W(90^\circ)} - 1]$.

Equation (4) is valid if there are no interactions between the nuclei and their environment that will change the orientation of the total angular momentum of the nuclei in the intermediate state j before the emission of the second gamma ray. This assumption is particularly

important in this study since the intermediate states have rather long half-lives. The half-life of excited state C is 5.2×10^{-9} sec and the half-life of level B is 63×10^{-9} sec. In these cases the intermediate states may exist for lengths of time long enough for electric and magnetic fields present in the source material to interact with the quadrupole and magnetic moments respectively of the nuclei, and hence change the initial orientation of the intermediate state. These interactions result in a time-dependent angular distribution different from the true or unperturbed angular distribution.

In this work an attempt was made to study the anisotropy as a function of the time interval between the emission of the two gamma rays. If one were to study the events in which the two gamma rays were emitted simultaneously, one would expect to observe nearly the true anisotropy, since there would be no time for internal fields in the source material to interact with the nuclei. On the other hand, if the anisotropy was determined at large time intervals between the emission of the two gamma rays, appreciable lengths of time might have elapsed and allowed the external fields to change the orientation of the intermediate state and hence change the angular distribution from its true value. In addition to obtaining the unperturbed angular distribution by studying the time dependence, one may also be able to obtain information concerning the interactions between the nuclei and their environment.

The time-dependent angular distributions can be written as

$$W(\theta, t) = 1 + \sum_K A_K G_K(t) P_K(\cos \theta) \quad (5)$$

All the time dependence of the angular distribution is contained in $G_K(t)$. The factor $G_K(t)$ may be periodic or decrease exponentially with time, depending on the type of interaction. These $G_K(t)$ coefficients for electric and magnetic interactions in various types of media are derived by Abragam and Pound⁶⁰ and are also given in an excellent review article by Devons and Goldfarb.⁶¹ As an example, the form of $G_K(t)$ for quadrupole interactions in solutions is

$$G_K(t) = e^{-\lambda_K t},$$

(6)

$$\lambda_K = \frac{3}{80} \left\langle \frac{eQ}{h} \right\rangle^2 \left\langle \left(\frac{\partial E}{\partial Z} \right)^2 \right\rangle_{av} \tau_c \frac{K(K+1) [4j(j+1) - K(K+1) - 1]}{j^2(2j-1)^2},$$

where Q is the nuclear quadrupole moment,

$\left\langle \left(\frac{\partial E}{\partial Z} \right)^2 \right\rangle_{av}$ is the average over time and space of the square of the electric field gradient at the nucleus produced by the ions and molecules in the solution,

and τ_c is the correlation time of the liquid.

For this type of interaction the correlation decreases exponentially with time, the decrease depending on the strength of the quadrupole interaction and the correlation time of the solution. Krohn, Novey, and Raboy have studied the time-dependent angular correlations between the alpha particles in the decay of Am^{241} and the 59.6-keV gamma ray BA in Np^{237} for various solutions.⁶² Figure 13, taken from the work of Krohn *et al.*, shows the time dependence of the coefficient $A_2 G_2(t)$ of Eq. (5) for various solutions. For HCl , $HClO_4$, and H_2SO_4 solutions the experimentally determined time dependence is of the form given by Eq. (6). However, for acetic acid solutions the time dependence does not follow Eq. (6). This has been interpreted as due to complexing of the neptunium daughter with acetate ions, producing a large electric field gradient at the neptunium nucleus. In this case it may not be valid to assume that the probability of a transition between nuclear substates is small for times of the order of the correlation time of the liquid fields used to derive Eq. (6).

A comparison of the time dependence of the angular distributions of the gamma rays for various chemical environments of Np^{237} formed by alpha decay of Am^{241} and beta decay of U^{237} would be interesting. If different oxidation states of the neptunium atom are formed in the two modes of decay, different interactions or strengths of interactions may exist between the neptunium nucleus and its environment. These differences may be observed as different time dependences of the angular distributions of the gamma rays. This type of information may be very useful in

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Fig. 13. $A_2G_2(t)$ of Eq. (5) vs. delay time (corrected) for Am^{241} alpha decay, taken from Krohn et al.⁶²

— HCl, HClO₄ solutions - - - H₂SO₄ solution
 ○ Acetic acid solution.

deducing the oxidation states formed shortly after the radioactive decay processes, in studying on a millimicrosecond time scale the rates of chemical complexing of daughter atoms "born" in solution, etc.

Rasmussen *et al.* have reported that the 208-keV transition is of mixed M1-E2 multipolarity and from conversion-electron intensities, estimated the E2/M1 photon-mixing ratio as 0.005 ± 0.005 .⁵⁷ A re-estimation of the relative L-subshell internal-conversion electron intensities by this investigator, by studying the permanent-magnet emulsions obtained in the work of Rasmussen *et al.*, showed that the E2/M1 photon mixing ratio may be larger, 0.015 ± 0.01 . It is very difficult to accurately determine small E2 admixtures from relative internal-conversion electron intensity on photographic emulsions, because of the large uncertainties involved in relating relative photographic densities to relative electron intensities. A much better estimate of the $L_1 : L_{11} : L_{111}$ conversion-electron ratios and hence the E2 photon admixture could be made by using a direct recording spectrometer, such as a double-focusing spectrometer. Unfortunately, a double-focusing spectrometer with the necessary 0.1% momentum resolution (needed to resolve the different L-subshell internal-conversion electron lines) is not readily available to this investigator at present.

The 59.6-keV transition BA is thought to be almost pure E1 in spite of its large hindrance factor and the anomaly of the internal-conversion coefficients. Asaro *et al.* have estimated from the experimental value of $\alpha(L_{111})$ that the maximum possible contribution of M2 photons is 0.015% and E3 photons 0.0015%.⁴⁴ These amounts of M2 and E3 contributions are too small to affect the experimental angular distributions. From the work of Rasmussen *et al.*,⁵⁷ the 64.8-keV transition also appears to be pure E1. The ratios of the L-subshell internal-conversion electron intensities and also the absolute L-subshell conversion coefficients agree within experimental errors with the theoretical predictions. Therefore, for the remainder of this work, the 208-keV transition CB will be the only one considered to consist of mixed multipolarities.

Figure 14 shows the dependence of the anisotropy on the E2 photon mixing ratio of the 208-keV transition for the two cascades, assuming spins $1/2+$ (12b) and $3/2+$ (12c) for level D in the cascade DC,CB.

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Fig. 14. Theoretical anisotropies as a function of δ_{208}^2 .

- (a) Gamma-ray cascade CB,BA
 $3/2 (M1 + \delta^2 E2) 5/2 (E1) 5/2$
- (b) Gamma-ray cascade DC,CB
 $1/2 (E1) 3/2 (M1 + \delta^2 E2) 5/2$
- (c) Gamma-ray cascade DC,CB
 $3/2 (E1) 3/2 (M1 + \delta^2 E2) 5/2$.

The two values of the anisotropy at a given mixing ratio are due to the choice of either a positive or negative phase for δ_{208} . The value of δ_{208}^2 (the ratio of the intensities of the two multipolarities) is, of course, the same in both cascades, DC,CB and CB,BA. However, Ofer has recently pointed out that the sign of δ in the formalism of Biedenharn and Rose actually depends on whether the mixed transition occurs first or second in the gamma-ray cascade.⁶³ Biedenharn and Rose define δ as

$$\frac{(j_1 \parallel L_{1+1} \parallel j)}{(j_1 \parallel L_1 \parallel j)}$$
, where j is the intermediate state of the cascade.
For the cascade DC,CB,

$$\delta_{208}(\text{DC,CB}) = \frac{(5/2 \parallel 2 \parallel 3/2)}{(5/2 \parallel 1 \parallel 3/2)} \quad \text{and for the cascade CB,BA,}$$

$$\delta_{208}(\text{CB,BA}) = \frac{(3/2 \parallel 2 \parallel 5/2)}{(3/2 \parallel 1 \parallel 5/2)}.$$

The inversion of the reduced matrix element is given by Biedenharn and Rose as $(2j+1)^{1/2} (j_1 \parallel L \parallel j) = (-1)^{j_1+L-j} (2j_1+1)^{1/2} (j \parallel L \parallel j_1)$ so that

$$\delta_{208}(\text{DC,CB}) = \frac{(5/2 \parallel 2 \parallel 3/2)}{(5/2 \parallel 1 \parallel 3/2)} = - \frac{(3/2 \parallel 2 \parallel 5/2)}{(3/2 \parallel 1 \parallel 5/2)} = -\delta_{208}(\text{CB,BA}).$$

For the sake of consistency, the sign of δ_{208} for the cascade DC,CB shown in Fig. 14 has been changed so that the reduced matrix elements of $\delta_{208}(\text{DC,CB})$ are defined the same as $\delta_{208}(\text{CB,BA})$.

B. Experimental Methods

1. Chemistry and Sources

The U^{237} was produced by neutron capture of U^{236} in the Materials Testing Reactor at Arco, Idaho. The radioactive material was dissolved with heating in a solution of 6 M HCl containing a small amount of H_2O_2 . After complete dissolution the solution was heated to destroy the excess H_2O_2 and evaporated nearly to dryness. The activity was then dissolved in concentrated HCl and placed on a Dowex A-1 anion-exchange column which had previously been equilibrated with concentrated HCl. The fission products and other impurity activities were separated from the uranium by elution from the column with concentrated HCl, uranium having a much

higher distribution coefficient. After elution of the impurities, the uranium was stripped off the column in 1 M HCl. A gamma-ray spectrum showed that there were no foreign activities present in the separated uranium fraction.

UO₂Cl₂ Source. The radioactively pure solution of UO₂⁺⁺ ions in 1 M HCl from the anion-exchange column was concentrated by evaporation almost to dryness. The activity was then dissolved in 1 M HCl.

U⁺⁴ Source. Two methods for reduction of UO₂⁺⁺ to U⁺⁴ were used. The first reduction was performed by using a Walden Silver Reductor at ~90°C.⁶⁴ The resultant solution in this case was U⁺⁴ in 6 M HCl. A second reduction was performed by using finely divided metallic zinc in a 1 M HClO₄ solution. The resultant solution in this case was U⁺⁴ in 1 M HClO₄.

Source Containers. The liquids were contained in sealed glass capillaries 3 inches long and 1 mm in diameter. The glass walls of the capillaries were less than 0.1 mm thick. The glass capillaries, which were sealed in the center, were warmed slightly and the open end of the capillary was placed into the radioactive solution. As the capillary cooled the liquid was drawn up into the capillary. The column of liquid, which was generally 2 to 3 mm high, was centrifuged down to the sealed portion of the capillary. The glass capillary was then sealed off quickly at roughly 1 cm above the liquid to prevent evaporation of the solution or slow oxidation by air. Theoretical calculations show that less than 4% of the 60-kev radiations undergo a Compton scattering event in the source.

2. Coincidence Circuit

The coincidence circuit used in this work was the same as that shown in Fig. 6, with the exception of a slightly different time-to-pulse-height converter. The photomultipliers and detectors were mounted on a circular graduated aluminum ring. One counter remained fixed while the other one could be rotated at various angles with respect to the fixed counter. The radioactive source was centered to within 0.1 cm of the circle described by the movable counter. The detectors, which were

normally placed 4.8 cm from the source, were 1-inch-diameter by 1-inch-thick cylindrical NaI(Tl) scintillation crystals mounted on RCA 6655A phototubes. The energy resolution of the NaI detectors was insufficient to resolve the 59.6-keV and 64.8-keV gamma rays. Therefore, one single-channel discriminator was set to accept all the 59.6- and 64.8-keV gamma-ray photopeaks, and the other discriminator was set to accept the 208-keV gamma-ray photopeak.

The fast-pulse outputs from the anodes of the photomultipliers were passed through Hewlett-Packard 460-A and -B amplifiers and into a diode coincidence circuit, which was the same as the circuit shown in Fig. 8 to the point E. The pulses on the plates of the E180F tubes (Fig. 8) were clipped to a duration of 250 μ sec. The 208-keV gamma-ray negative pulse input to the diode coincidence circuit was split; one part of the pulse went to the diode coincidence circuit, the other part of the pulse was passed through a delay cable and into the "stop" input of a time-to-pulse-height converter. This time-to-pulse-height converter was a modification of one described by Weber *et al.*⁶⁵ The output of the diode coincidence circuit at the point E of Fig. 8 was amplified and served as the "start" pulse of the time-to-pulse-height converter. A block diagram of this part of the circuit replacing the time-to-pulse-height converter in Fig. 6 is shown in Fig. 15.

Fig. 15. Millimicrosecond time sorter II

The working of this type of circuit can be more easily understood by referring to Fig. 16, which shows the time relationships between the various pulses for a hypothetical prompt coincidence. Let us assume at first that either the 59.6-kev or the 64.8-kev gamma ray is emitted simultaneously with the 208-kev gamma ray.

Fig. 16. Time relationships of various pulses in time sorter II for a hypothetical prompt coincidence.

Pulse "a" of Fig. 16 corresponds to the output of the E180F limiting tube due to a 60- or 65-kev gamma ray which has been delayed artificially relative to pulse "b" of the 208-kev gamma ray. The output of the diode coincidence circuit at point E of Fig. 8 corresponds

to pulse c. Note that the start time of this output pulse c is related to that of the 60- or 65-kev gamma ray and not that of the 208-kev gamma ray. Pulse c is stretched and clipped to 300 μ sec in the time-to-pulse-height converter and put on a grid of a 6BN6 tube in which both control grids are normally biased so that the tube is nonconducting. This positive input on the grid starts the tube conducting and the voltage on the plate of the 6BN6 tube decreases linearly with time. The negative pulse d, which is the second part of the 208-kev input to the diode coincidence circuit and which has been delayed, is stretched in the time-to-pulse-height converter and passed to the second control grid of the 6BN6 tube. This negative input then stops the tube from conducting and the voltage at the plate of the 6BN6 tube rises again to the B^+ voltage with a time constant $RC = 45 \mu$ sec. The resultant pulse at the plate of the 6BN6 tube for this coincidence event is a negative pulse whose voltage is proportional to the time difference between pulses c and d. Now consider the case in which the 59.6-kev gamma ray is delayed with a half-life of 63×10^{-9} sec with respect to the 208-kev gamma ray corresponding to the cascade CB,BA. The 59.6-kev gamma ray has a probability of $\lambda e^{-\lambda t}$ of being emitted at time t after the 208-kev gamma ray. Some pulses in 16a and therefore in 16c appear later than those in a prompt event and the resultant delay curve on the multichannel analyzer for this cascade has a slope corresponding to a half-life of 63×10^{-9} sec on the lower-pulse-height or shorter-delay side of the prompt event. When the 208-kev gamma ray is delayed relative to the 64.8-kev gamma ray with a half-life of 5.2×10^{-9} sec in the cascade DC,CB, some pulses of 16d appear later than in a prompt event relative to 16a and in this case the slope corresponding to a half-life of 5.2×10^{-9} sec appears on the larger-pulse-height or longer-delay side of the prompt event.

The time-to-pulse-height converter shown in Fig. 8 would give the same time spectrum, i.e., the resolution of the two cascades, as the present circuit. However, the chance coincidence counting rate in the time-to-pulse-height converter of Fig. 8 is twice as large as with the present system. This is because in time sorter I there are two positions in time of pulses a and b relative to each other in a chance event that

yields the same pulse height. In the present coincidence system if pulse b arrives at the diode coincidence circuit before pulse a in a chance event, then the chance coincidence spectrum appears as a constant background on the multichannel analyzer. On the other hand, if the 208-keV gamma-ray pulse b arrives after pulse a in a chance event then the output of the time-to-pulse-height converter is a constant simply related to the time delay introduced in the split 208-keV pulse. This appears as a sharp peak on the 100-channel pulse-height analyzer and should correspond to a constant delay. This chance peak was very valuable in showing that no shifts occurred in the position of the time spectrum during the determinations at different angles. Also the reduction of the chance coincidence counting rate by a factor of two by using this new time-to-pulse-height converter greatly aided in reducing the statistical uncertainties. A typical time spectrum is shown in Fig. 17.

3. Treatment of Data

The delayed coincidence curves were taken in time intervals of from 100 to 600 minutes at 90, 180, and 270 degrees with respect to the first counter. After a series of such determinations, a chance-coincidence spectrum was taken by using two different U^{237} sources. The chance-coincidence spectrum, after suitable normalizations, was subtracted from the delayed-coincidence spectra and the resultant spectra were corrected for the radioactive decay of the source and slight changes in the singles counting rates.

Figure 17 shows an uncorrected spectrum, the spectrum corrected for the chance coincidence spectrum, and the resolution of the two cascades from the resultant time spectrum. The two cascades were resolved by (a) extrapolating the back slope which corresponded to a half-life of 5.2×10^{-9} sec, (b) requiring that the ratio of the coincidence counting rates of the cascade CB,BA to DC,CB be 16, as determined from the data given by Rasmussen *et al.*⁵⁶ and (c) requiring that the centroid of cascade DC,CB be displaced by the mean life of level C from the centroid of the prompt-coincidence curve determined with Na^{22} . The resolution of the two cascades is not perfect, but one can see that for delays less

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Fig. 17. Time spectra for 64.8-208-kev and 208-59.6-kev gamma-ray coincidences in Np^{237} .

than 238 μsec and for delays greater than 244 μsec the cascades CB,BA and DC,CB, respectively, are resolved from each other.

C. Results

A summary of the anisotropies obtained for various chemical states is given in Table X. The data were analyzed in a number of ways. To determine the existence of a rather weakly time-dependent anisotropy, the different corrected time spectra were first integrated from -3 to -100 μsec for the cascade CB,BA and from +3 to +10 μsec for the cascade DC,CB. The anisotropies for the two cascades were then calculated by using these integrated spectra (columns 2 and 4). The time zero was defined by the centroid of the prompt coincidence curve. The anisotropy of the cascade CB,BA was also calculated from the spectra integrated from -3 to -10 μsec to determine the existence of a more strongly time-dependent anisotropy (column 3). A 9% correction has been applied to the anisotropies to correct for the finite angle subtended by the detectors. This correction was calculated from the equation of Rose⁶⁶ by using an IBM 704 computer.

Table X. Summary of measured anisotropies for different sources

Source	Limits of integration (μsec)		
	-3 to -100	-3 to -10	+3 to +10
UO_2^{++} 1M HCl	-0.010 ± 0.005	-0.01 ± 0.03	-0.06 ± 0.02
U^{+4} 6M HCl	-0.001 ± 0.005	-0.005 ± 0.015	-0.01 ± 0.04
U^{+4} 1M HClO_4	-0.005 ± 0.008	-0.022 ± 0.02	-0.028 ± 0.036
UO_2^{++} solid	-0.022 ± 0.007	-0.039 ± 0.02	-0.10 ± 0.04

Krohn and Fried have also studied the anisotropy of the cascade CB,BA for various chemical states.⁶⁷ Unfortunately, they did not report the resolving time of their coincidence circuit (the time over which the anisotropies were averaged). However, assuming that they used the same coincidence circuit as in their previously reported work, one could say

that the resolving time would be between 20 and 70 μsec . Their results, which are in good agreement with the results of this work, are given in Table XI.

Table XI. Anisotropy of cascade CB,BA as determined by Krohn and Fried⁶⁷

Source	Anisotropy
0.5N UO_2Cl_2	0.008 ± 0.013
6N $\text{UO}_2(\text{NO}_3)_2$	-0.016 ± 0.009
Dry $\text{UO}_2(\text{NO}_3)_2$	-0.028 ± 0.006
4N UO_2SO_4	-0.011 ± 0.007
6N UO_2Cl_2	-0.006 ± 0.009
6N UCl_4	-0.001 ± 0.009
Dry UCl_4	-0.011 ± 0.005

In order to investigate the anisotropies in more detail, the anisotropies were calculated as a function of the delay between the emission of the two gamma rays for the various sources. Figure 18 shows three of these cases near the prompt event. It is very difficult to obtain good statistics in this type of analysis, since the fraction of the total coincidence contained in these time intervals is small. The decrease in the anisotropy from +2 to 0 μsec is probably due to the overlap of the two gamma-ray cascades in the time spectrum.

D. Discussion

The data in Table X show that the anisotropies obtained for the cascade CB,BA are much smaller than the -0.14 or -0.29 expected for the previously reported $\delta^2_{208} = 0.005$. Seven out of eight of the time-integrated anisotropies (for integrated times greater than 10 μsec) in this work and the work of Krohn and Fried for the cascade CB,BA in liquid

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Fig. 18. Time-dependent anisotropy analysis.

a. UO_2^{++} 1 M HCl b. UO_2^{++} solid c. U^{+4} 6 M HCl.

sources were negative and less than -0.02. From the integration from -3 to -10 msec for liquid sources, an upper limit of -0.04 can be set for the anisotropy for cascade CB,BA. From the study of anisotropy vs delay in Fig. 18 an upper limit of -0.09 can be set for the anisotropy at zero delay. Because of the very small anisotropies observed experimentally and the uncertainty in the mixing ratio of the 208-kev gamma ray, the form of $G_2(t)$ and hence the nature of the interactions between the nuclei in their intermediate states and their environment could not be determined. In the following discussion $G_2(t)$ is assumed to be of the form $e^{-\lambda_2 t}$, as observed in the work by Krohn et al. on the alpha decay of Am^{241} .⁶² With the assumption that no extremely strong time dependence of the anisotropy exists, the limits on the anisotropies obtained for cascade CB,BA are consistent with $\delta_{208}^2 > 0.014$ and $\delta_{208} > 0$. An extremely strong time dependence with $\lambda_2 > 1.4 \times 10^9 \text{ sec}^{-1}$ ($G_2(t)$ decreasing by a factor of two in less than 0.5 msec) would decrease the minimum expected anisotropy of -0.30 for $\delta_{208} < 0$ to less than the limit of -0.09 set for the anisotropy of cascade CB,BA at zero delay. If the interaction were this large, the data for cascade CB,BA would then be consistent with either phase of δ_{208} . The effect of the finite resolving time of the coincidence circuit on the anisotropy at a given delay was taken into account by expressing the product $A_2 G_2$ at delay D by

$$A_2 G_2(D) = \int_0^\infty A_2 e^{-(\lambda_2 + \frac{1}{\tau})t} P(D-t) dt \bigg/ \int_0^\infty e^{-\frac{t}{\tau}} P(D-t) dt, \quad (7)$$

where τ is the mean life of the intermediate state,

$P(x)$ is the counting rate of the prompt coincidence curve at delay x ,

$P(x)$ for the coincidence circuit and the appropriate energy of gamma rays was determined with a Na^{22} source.

The integrated anisotropies for cascade DC,CB in Table X and the dependence of the anisotropy on delay (Fig. 18) show that a negative anisotropy exists for this cascade. Also, from the time dependence of the anisotropies for cascade DC,CB in Fig. 18a and 18b there is good evidence that the time dependence of the anisotropy and hence approximately

of $G_2(t)$ is not extremely strong, i.e., $\lambda_2 < 1.4 \times 10^9 \text{ sec}^{-1}$. The dashed lines of Fig. 18 a, b, and c show the time dependence of the anisotropy for $\lambda_2 = 1.4 \times 10^9 \text{ sec}^{-1}$, assuming a spin of 3/2 for level D, $\delta_{208} < 0$, and the maximum unperturbed anisotropy of -0.17. Clearly this time dependence is too strong and the case of $\delta_{208} < 0$ can be ruled out. With the requirements $\delta_{208}^2 > 0.014$ and $\delta_{208} > 0$, the negative anisotropy for cascade DC,CB is consistent with a spin assignment of 1/2 for level D, in agreement with the assignment by Rasmussen et al.⁵⁷ A positive anisotropy would be expected under the same requirements if the spin of level D were 3/2.

If the estimate of the E2 photon mixing ratio for the 208-keV transition of 0.015 ± 0.01 is correct, some time dependence of the anisotropies must exist. From Fig. 14 the minimum anisotropy for the cascade CB,BA consistent with this E2 photon mixing ratio is -0.04, whereas the average integrated anisotropy from -3 to -100 μsec observed for the solution sources in this work is -0.005 ± 0.004 . Also, from the work of Krohn and Fried, the average of the anisotropies for solution sources is -0.005 ± 0.004 . In order for the integrated anisotropy from -3 to -100 μsec of -0.005 ± 0.004 to be consistent with a δ_{208}^2 of 0.015 ± 0.01 , λ_2 must be greater than $4.3 \times 10^7 \text{ sec}^{-1}$ (the value of $G_2(t)$ must decrease by a factor of two in less than 16 μsec). This value of λ_2 was calculated to satisfy the equation

$$W(\theta) = \int_{t_1}^{t_2} [1 + A_2 G_2(t) P_2(\cos \theta)] e^{-\frac{t}{\tau}} dt \bigg/ \int_{t_1}^{t_2} e^{-\frac{t}{\tau}} dt \quad (8)$$

The symbol τ is the mean life of level B. This lower limit of λ_2 is larger than the values of $1.35 \times 10^7 \text{ sec}^{-1}$ and $2.1 \times 10^7 \text{ sec}^{-1}$ obtained by Krohn et al. for the alpha decay of Am^{241} in HCl and in H_2SO_4 solutions respectively, indicating that a much stronger interaction may be present in the intermediate state formed in beta decay. The average of the observed integrated anisotropies for solution sources from +3 to +10 μsec for the cascade DC,CB is -0.033 ± 0.02 . With $\lambda_2 > 4.3 \times 10^7 \text{ sec}^{-1}$ the unperturbed anisotropy would be greater than roughly -0.04,

which is consistent with $\delta_{208}^2 = 0.015 \pm 0.01$, $\delta_{208} > 0$, and a spin of $1/2$ for level D.

Because of (a) the very small observed anisotropy of the gamma-ray cascade CB,BA, most likely due to appreciable E2 admixture in the 208-kev gamma ray, (b) the small fraction of the total coincidences due to the cascade DC,CB, and (c) the inability to achieve perfect resolution of the two gamma-ray cascades, further studies of the dependence of the anisotropy on the chemical state and a more precise determination of the E2 admixture in the 208-kev transition cannot be performed at present.

The stronger time dependences of the gamma-ray angular distributions consistent with this work for Np^{237} formed by beta decay over that found by Krohn et al.⁶² for formation of alpha decay may be qualitatively understood on the basis that different oxidation states are formed. In the alpha decay of Am^{+3} , Np^{+1} is initially formed. The degree of additional ionization due to the recoiling neptunium ion going through solution and electronic excitation of the Np^{+1} ion is questionable, but no appreciable ionization is expected. Krohn et al. speculate that the most likely valence state after a few millimicroseconds is Np^{+3} . This ion exists as the simple hydrated ion in water, stable to oxidation over periods of time very long relative to the time intervals investigated. In the beta decay of U^{237} as UO_2^{++} , Np^{+7} is initially formed, and in U^{+4} decay, Np^{+5} is formed. These ions do not undergo additional ionization due to recoil effects because of the very low recoil energy of 0.6 ev. Np^{+7} is not stable and probably reacts with water rapidly to form NpO_2^{++} . The Np^{+5} ion probably reacts with water rapidly to form NpO_2^+ ion. The formation of both of these ions probably takes place in less than 1 msec. If in the Am^{241} alpha decay the lower oxidation states of neptunium actually existed during the time of Krohn et al.'s measurements and the neptunyl ions were formed in the beta decay of U^{237} , much stronger quadrupole interactions and hence a stronger time dependence of the anisotropy would be expected in the latter case. The electric field gradients existing at the site of the neptunium nucleus in the linear NpO_2^+ and NpO_2^{++} ions are very much larger than the time-averaged electric field gradient produced

by water molecules and noncomplexing ions in the hydrated lower-oxidation-state ions. Strong quadrupole interactions have been shown to exist in NpO_2^{++} .⁶⁸

The quadrupole interaction in the neptunyl ions and during their formation may be expected to be of the order of the interaction in the complexing of the supposed Np^{+3} ions with acetate ions observed in the work of Krohn *et al.*⁶² From their work (Fig. 13) the interaction in acetate complexing is very strong; the anisotropy at 5 μsec is one-half the value of the maximum observed anisotropy in noncomplexing solutions.

In addition to the quadrupole interactions, magnetic interactions may also be important, since the ground states of the neptunium ions are paramagnetic. The interactions between the magnetic moment of the neptunium nucleus and the magnetic field produced by the 5f electrons may be very strong, especially for NpO_2^+ and NpO_2^{++} .

E. Half-Lives of Np^{237} Excited States

1. 59.6-keV Excited State

From the slope of the delayed-coincidence curve for the 208-keV gamma rays in coincidence with the 59.6-keV gamma rays (Fig. 17), a half-life of $63 \pm 5 \mu\text{sec}$ was obtained for the 59.6-keV excited state. This value is in good agreement with the value of $63 \pm 5 \mu\text{sec}$ reported by Beling *et al.*,⁶⁹ who studied the alpha-gamma coincidences in the decay of Am^{241} .

2. 268-keV Excited State

Figure 19 shows a delayed-coincidence curve obtained for beta particles in coincidence with the 208-keV gamma rays. An anthracene crystal was used to detect the beta particles. The prompt-coincidence curve also shown in Fig. 19 was obtained with beta particles in coincidence with 280-keV gamma rays in the decay of Hg^{203} ; the half-life of the intermediate state in this case is 0.29 μsec . From the slope of the delayed-coincidence curve a half-life of $5.2 \pm 0.2 \mu\text{sec}$ was obtained for the 268-keV excited state. An analysis of the beta particles in coincidence with the 268-keV gamma rays gave a half life of $5.0 \pm 0.5 \mu\text{sec}$. The best value obtained in this work, $5.2 \pm 0.2 \mu\text{sec}$, is in good agreement with the value of $5.4 \pm 0.5 \mu\text{sec}$ reported by Bunker *et al.*⁷⁰

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Fig. 19. Delayed coincidence curve for beta particles of U^{237} in coincidence with 208-kev gamma-rays of Np^{237} .

3. 332-keV Excited State

Figure 20 shows the delayed-coincidence curve for beta particles in coincidence with the 332-keV gamma rays. The slope of the curve corresponds to a maximum half-life of 1.0 μsec . Since this slope is almost the same as that of a prompt event with similar energy radiations, the value of 1.0 μsec must be considered only as an upper limit of the half-life of the 332-keV excited state.

From (a) the upper limit of 1.0×10^{-9} sec for the half-life of the 332-keV excited state, (b) the gamma-ray intensities of 2.2 and 1.4 per 100 beta particles for the 64.8-keV E1 and the 332-keV E2 transitions,⁵⁷ and (c) the theoretical total conversion coefficients of 0.43 and 0.16 for the two transitions, upper limits for the half-lives of the 64.8- and 332-keV gamma ray transitions are calculated to be 2.1×10^{-9} sec and 3.5×10^{-9} sec respectively. The theoretical K- and L-shell internal-conversion coefficients of Sliv and Band²⁵ and the theoretical M-shell conversion coefficients of Rose⁷¹ were used for calculating the total conversion coefficients. The corresponding theoretical E1 and E2 gamma-ray transition half-lives for single-proton transitions calculated from the formulae of Moszkowski³⁰ (assuming the statistical factor to be unity) are 4.4×10^{-13} sec and 7.3×10^{-10} sec. The 64.8-keV E1 gamma ray is therefore hindered by a factor of $\leq 4.8 \times 10^3$ and the 332-keV E2 ≤ 4.8 from the single-proton estimates.

Rasmussen et al. have assigned the 332-keV excited state as a proton state designated by the quantum numbers $1/2 \ 1/2^+ \ [400]$ and the 268-keV excited state as $3/2 \ 3/2^- \ [521]$, (using the notation $IK\pi[Nn_z\Lambda]$).⁵⁷ If, as assumed, these assignments are correct, the 64.8-keV E1 gamma ray between these two excited states is twice forbidden in the Δn_z selection rules. Stephens et al. prefer the assignment of $3/2 \ 1/2^- \ [530]$ for the 268-keV excited state.⁷² According to this assignment the 64.8-keV E1 is also twice forbidden in the Δn_z selection rules. Since this E1 gamma ray has been found to be retarded by less than a factor of 4.8×10^3 the role of the selection rules of Δn_z does not seem very important in this case. That (a) the L-subshell internal-conversion coefficients of the 64.8-keV E1 transition agree with the theoretical values of Sliv within

Fig. 20. Delayed coincidence curve for beta particles of U^{237} in coincidence with 332-kev gamma rays of Np^{237} .

the experimental error of ~30%, and (b) the gamma ray of this transition is hindered by less than 4.8×10^3 is consistent with the previous information on the dependence of the anomaly of conversion coefficients on gamma-ray retardation, i.e., for gamma-ray hindrances less than 10^4 the experimental E1 conversion coefficients agree with the theoretical values within the experimental error of ~30%.^{43,44}

From the assignments by Rasmussen et al. for the 332-keV excited state, the 332-keV E2 gamma ray between this state and the $5/2^-$ $5/2^+$ [642] ground state should be four times forbidden in the Δn_z quantum-number selection rule. However, the hindrance factor of ≤ 4.8 for this gamma ray seems to be inconsistent with the hindrance factors for other known "forbidden" E2 gamma rays. In Table XII are listed some known "forbidden" E2 gamma rays and their hindrance factors. The hindrance factor H is defined as the ratio of the experimental E2 gamma-ray half-life to that calculated from the formula of Moszkowski³⁰ assuming the statistical factor to be unity. The orbital assignments are taken from the review paper by Mottelson and Nilsson.³⁹

Table XII. Hindrance factors for E2 gamma rays forbidden by the asymptotic quantum-number selection rules

Nucleus	Orbital assignment		E (keV)	Experimental E2 gamma-ray half life	H
	Initial	Final			
Ta ¹⁸¹	1/2 1/2 + [411]	5/2 5/2 + [402]	133	4.2×10^{-5}	400
Ta ¹⁸¹	5/2 5/2 + [402]	9/2 7/2 + [404]	346	7.9×10^{-8}	93
Ta ¹⁸¹	5/2 5/2 + [402]	7/2 7/2 + [404]	482	1.3×10^{-8}	81
Np ²³⁷	3/2 3/2 - [521]	7/2 5/2 - [523]	165	1.7×10^{-7}	7
Np ²³⁷	3/2 3/2 - [521]	5/2 5/2 - [523]	208	1.5×10^{-6a}	200
Pu ²³⁹	5/2 5/2 + [622]	1/2 1/2 + [631]	286	1.5×10^{-7}	98

^aUsing $\delta_{208}^2 = 0.015$.

The small hindrance factors for the 64.8-keV E1 and the 332-keV E2 gamma rays may be an indication that these transitions are not forbidden and that the assignment of $1/2^+ 1/2^- + [400]$ for the 332-keV excited state is not correct. The only other near-by $I = 1/2^+$ state, according to the Nilsson diagram,³⁹ is the $1/2^+ [660]$ orbital, but for this case the E1 and E2 gamma rays would also be forbidden.

An alternative interpretation of this excited state may be that it is the first gamma vibrational state ($n_\gamma=1$, $K=-2$) of the $\frac{5}{2}^+ [642]$ ground state, or may contain an appreciable admixture of this state. Unfortunately very little is known at this time concerning vibrational states in odd-A nuclei, and the nature of the 332-keV excited state cannot be determined uniquely from just an upper limit of the half-life. The actual measurement of the E2 reduced transition probability for the 332-keV gamma ray by the method of delayed coincidences or by Coulomb excitation may distinguish between a (unexplained) low hindrance factor for a "forbidden" transition or an E2 transition between a vibrational state and its ground state. The transition probability for an E2 transition between vibrational states should be larger than single-particle transition probabilities but smaller than rotational-transition probabilities.³⁷

V. ELECTRON-ELECTRON COINCIDENCE MEASUREMENTS

A. Introduction

A nuclear decay scheme for a particular isotope can be constructed from sum relationships of the energies and intensities of the observed transitions between different nuclear states. However, ambiguities generally exist in the establishment of a unique decay scheme by this method, the number of ambiguities strongly dependent on the complexity of the decay scheme and the accuracy of the determined transition energies. By determining the coincidence relationships between various transitions, these ambiguities can generally be removed. Coincidence spectrometers using thallium-activated NaI scintillation crystals to detect the gamma rays have been widely used for this purpose. However, the use of NaI crystals as scintillation detectors is quite limited in the study of complex decay schemes because of its poor resolution ($\sim 9\%$ at 500 keV and $\sim 14\%$ at 100 keV) and also because of the interference of the continuous background of coincidence with the Compton distribution from higher-energy gamma rays.

These bad features can be removed by using magnetic spectrometers to resolve the internal-conversion electrons of the various transitions in place of NaI crystals resolving the various gamma rays. The energy resolution of the electron spectrometers can be very much better than that of NaI, and in electron-capture decay there is no coincident continuous distribution under the electron lines. Unfortunately, with the improved features of electron-electron coincidence techniques there are a number of associated bad features. The equipment and experimental methods are very much more complex, and the coincidence counting rates are less by several orders of magnitude than for gamma-gamma ray coincidence measurements.

B. Original Electron-Electron Coincidence Spectrometer

A spectrometer designed for electron-electron coincidence work was constructed by Passell and others and finally brought into operation by Juliano at this laboratory. Since this instrument is described in detail elsewhere,⁷³ only a brief description is presented here. The instrument consisted of two collinear thin-lens beta spectrometers back to back (Fig. 21). Electrons emitted from the source S in the center of the evacuated spectrometer were focused on detectors D1 and D2 at the ends of the spectrometer by using the focusing properties of the thin-lens magnets M1 and M2. The trajectories of the electrons and hence the momentum resolution and transmission were defined by a suitably constructed baffle system. Lead plugs L1 and L2 shielded the detectors from direct radiations from the source. The entire spectrometer was constructed free of iron so that the magnet current would be directly proportional to the momentum or magnetic rigidity ($H\rho$) of the electrons. By varying the currents in magnets M1 and M2, electrons of different energies could be focused on detectors D1 and D2. The detectors were either anthracene crystals or Livermore plastic scintillators. In order to make each half of the spectrometer magnetically independent of the other half, the compensating magnets C1 and C2 were used. Each of these compensating magnets was connected in series but with opposite magnetic polarity to the main magnet on the same side of the spectrometer. The numbers of turns of wire in the compensating magnets were chosen so that their magnetic fields would essentially cancel out the tailing magnetic fields in the other half of the spectrometer due to the main magnets. With the magnets used in this way, the two halves of the spectrometer were essentially independent of each other.

The light outputs of the scintillators D1 and D2 were converted to electronic pulses by using RCA 6655 photomultiplier tubes. These pulses were then passed into a time-to-pulse-height coincidence circuit similar to the one shown in Fig. 6 except that (a) only a double coincidence was required between the output of the time-to-pulse-height converter and a pulse from one detector, (b) the time-to-pulse-height converter was similar to one described by Weber,⁶⁵ (c) a 50-channel

Fig. 21. Original electron-electron coincidence spectrometer.

analyzer was used in place of the 100-channel Penco analyzer. Displaying the coincidence relationships between two electrons of different energies on a time scale is a definite improvement over the standard fast-slow coincidence analysis. In addition to obtaining the coincidence relationship between two electrons, the time relationship and hence information on the lifetime of the intermediate state is simultaneously recorded. Also one obtains simultaneously the chance-coincidence counting rate, which appears as a constant background on the time scale and hence is very easy to determine. Figure 22 shows a coincidence spectrum for a 56-keV conversion electron in coincidence with beta particles of 49 keV superimposed on a chance spectrum.

The spectrometer had a fixed resolution of 0.023 (resolution defined as fractional momentum width at half maximum for a monoenergetic electron peak.) The transmissions of both halves of the spectrometer were measured by this investigator with a calibrated Bi^{207} source as 0.0020 and 0.0023. The absolute number of 570- and 1064-keV gamma rays in the Pb^{207} daughter were determined with a 3x3-inch NaI crystal under known geometry and using the photopeak efficiencies given by Heath.⁴ The K conversion coefficients of 0.0165 and 0.103 were used for the 570- and 1064-keV transitions respectively. These represent the average of the conversion coefficients determined by McGowan and Campbell⁷⁴ and by Wapstra.⁷⁵ The transmissions were also checked by using Pa^{233} and Cs^{137} sources. The very low transmission of 0.002 and an associated resolution of 0.023 represents a low figure of merit for present-day beta-ray spectrometers. The low transmission is a serious limitation for coincidence work.

Let us consider the hypothetical decay scheme shown in Fig. 23. The coincidence counting rate between K internal-conversion electrons of transitions E1 and E2, assuming 100% efficient detectors, is given by

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Fig. 22. Typical coincidence and chance spectra displayed on a time scale.

$$N(E1-E2) = N_0 T_1 T_2 X_1 X_2. \quad (9)$$

Fig. 23 Hypothetical decay scheme

N_0 represents the population per unit time of excited state A, T_1 and T_2 are the transmissions of each half of the spectrometer, and X_1 and X_2 are the K-electron internal-conversion probabilities for E1 and E2. X is simply related to the conversion coefficients by $X = \frac{\alpha_K}{1+\alpha_T}$, where α_K is the K-conversion coefficient and α_T is the total conversion coefficient. For an example assume $Z = 80$, $E_1 = E_2 = 350$ kev, and both transitions of E2 multipolarity. For this case $\alpha_K = 0.042$ and $\alpha_T = 0.068$. To obtain a "reasonable" coincidence counting rate of 10 coincidences per minute, N_0 must be $2.5 \times 10^9 \text{ min.}^{-1}$. In general this represents a lower limit on source activity, since more active sources are required for an equivalent coincidence counting rate if excited state A is populated in only a small fraction of the total disintegrations of the parent and there are competing modes of de-excitation for excited states A and B.

Since the coincidence counting rates depend on the product of the transmissions of both halves of the spectrometer and the product of the conversion coefficients (in the limit $\alpha_T \ll 1$), the very low transmissions placed severe limitations on the radioactive isotopes that were to be studied. Only coincidences between rather intense and highly converted transitions could be successfully investigated. The beta decay of Pa^{233} was studied first, since in addition to a number of highly converted transitions it also had a conveniently long half-life of 27 days.

C. Beta Decay of Pa²³³

1. Previous Work

The energy levels of U²³³ populated by the beta decay of 27-day Pa²³³ have been studied by Keller and Cork,⁷⁶ Elliott and Underhill,⁷⁷ Brodie,⁷⁸ Ong and Sizoo,⁷⁹ Ong and Kramer,⁸⁰ and Albouy and Valadaras.⁸¹ All these investigations consisted of analysis of the energies and intensities of the internal-conversion electrons and beta-ray spectra. The results reported in Refs. 78, 79, 80 agree with the decay scheme (obtained from sum relationships of the transition energies and intensities) shown in Fig. 24a. The energies of the transitions are taken from Ref. 80. The various observed beta groups are listed in Table XIII.

Table XIII. Beta Groups of Pa²³³

Reference	Energy (kev)	Intensity
76	~200	
77	570	
	~260	
78	568 ± 5	5
	264 ± 4	45
	~140	50
80	568 ± 5	5
	257 ± 5	58
	145 ± 10	37

The 568-kev beta group is believed to go to the ground state and the ~260-kev beta group to the excited state at 312.9 kev, and the 140-kev beta group is believed to consist of several beta groups populating the higher excited states.

Very little coincidence work has been performed, and some of the work performed is not consistent with the decay scheme of Fig. 24a. Ong and Kramer studied electron-electron coincidences by absorption methods.

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Fig. 24. Energy levels of U^{233} populated by the beta decay of Pa^{233} .
 (a) from References 78, 79, and 80.
 (b) from Reference 77.

They found that the 86-kev transition is correlated with electrons of 210 kev, presumably the K electrons of the 313-kev transition as in the proposed decay scheme. However, they also found that the 312.6-kev transition is in coincidence with electrons of 180 kev. From the decay schemes of Fig. 24a and b the 313-kev transition should be in coincidence with the 255-kev beta group. Elliot and Underhill have also performed electron-electron coincidence measurements between some of the transitions. Their work is quoted in the Brodie paper.⁷⁸

On the basis of their coincidence and conversion-electron work, Elliot and Underhill deduced the decay scheme shown in Fig. 24b (taken from C). One important difference between the decay scheme proposed by Elliot and Underhill and the one proposed by C, D and E is the placement of the 75-kev transition. Elliott and Underhill have the transition depopulating an excited state at 474 kev, while the other investigators have the transition depopulating an excited state at 416 kev, 12.2 kev, 103.7 kev, 86.2 kev, and 74.8 kev.

2. Coincidence-Measurement Results

A mass-free and radioactively pure sample of Pa^{233} was obtained from Royal Albridge (Lawrence Radiation Laboratory). This Pa^{233} was produced by neutron capture of Th^{232} and the subsequent beta decay of Th^{233} . Since some of the electrons in electron-electron coincidence measurements must pass through the source backing material, the backing material must be made very thin relative to the range of the electrons. In this case the source backing material consisted of a laminated VYNS film⁸² with a very small amount of gold vaporized on the plastic to prevent electrostatic charging of the source. The total thickness of the source backing material was $20 \mu\text{g}/\text{cm}^2$.

The conversion electrons most prominent in the electron spectra are due to the 74.8-, and 86.2-, 103.7-, and 312.6-kev transitions. The first three of these transitions have intensities of approximately 10% of the total disintegrations of Pa^{233} , and the 312.6-kev transition has

an intensity of 80%. All four of these transitions are thought to be of M1 multipolarity, with theoretical L-conversion coefficients for the 74.8-, 86.2-, and 103.7-keV transitions of 8.8, 5.6, and 3.4 respectively and a theoretical K-conversion coefficient of 0.7 for the 312.6-keV transition. The L lines of the 74.8- and 86.2-keV transition could easily be resolved, whereas the M-conversion electrons of the 86.2-keV transition and the L electrons of the 103.7-keV transition, which are about equal intensity, could not be resolved from each other.

Figure 25 shows the Fermi plot obtained for the beta spectrum in coincidence with the K electrons of the 313-keV transition. The end point of the maximum-energy beta group is 250 ± 5 keV. The beta spectrum is definitely complex, with other beta groups of ~ 174 and ~ 150 keV. Relative intensities are difficult to obtain because of the small number of experimental points and also because the two lowest-energy points may be high owing to contributions from the 86- and 104-keV transitions. However, it was found that the 250-keV beta group and the lower-energy complex beta spectrum are of nearly the same intensity. These data are in agreement with the decay scheme of Fig. 24a, and show that the coincidence result of Ong and Kramer is in error.

Figure 26a shows the Fermi plot for the beta spectrum in coincidence with the L electrons of the 74.8-keV transition. This Fermi plot indicates that there is just one beta group with an end point of 155 ± 7 keV in coincidence with the 74.8-keV transition. This observation is in accord with the decay scheme of Fig. 24a, for if the beta group to the ground state is 568 ± 5 keV, the 74.8-keV transition then depopulates an excited state at 413 ± 9 keV or less. This rules out the placement of the 74.8-keV transition in the decay scheme of Elliot and Underhill as depopulating an excited state at 474 keV, and is in agreement with the 74.8-keV transition depopulating an excited state at 416 keV.

Figure 26b shows the Fermi plot of the beta spectrum in coincidence with the 86.2-keV transition. The maximum beta end point of 175 ± 8 keV is consistent with the 86.2-keV transition depopulating an excited state at 393 ± 9 keV, again in agreement with the decay scheme shown in Fig. 24a. This Fermi plot also shows the existence of a

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Fig. 25. Fermi plot of beta spectra of Pa^{233} in coincidence with 313-kev transition, showing resolution into components.

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Fig. 26. Fermi plots of beta spectra of Pa^{233} in coincidence with the 74.8- and 86.2-keV transitions.
△ coincidence with the 74.8-keV transition
○ coincidence with the 86.2-keV transition.

lower-energy beta group, indicating that this excited state is also populated by transitions from higher excited states.

In this work the 74.8-, 86.2-, and 103.7-keV transitions were also shown to be in coincidence with the 313-keV transition, in agreement with the work of Elliot and Underhill.

An investigation of the more prominent highly converted transitions in the beta decay of Pa^{233} was possible despite the low transmission of the spectrometer. However, the work was tedious and a complete quantitative investigation of the decay scheme would have been very difficult. With a source of approximately 10^8 disintegrations per minute, the average coincidence counting rates for the 86.2- and 74.6-keV transitions in coincidence with beta particles was 30 per hour. For the highest-energy points taken on the beta spectra, the coincidence counting rate was about 10 per hour. Hence, for even poor statistics very long counting times would be necessary to study the weaker transitions. The coincidence counting rates could be increased slightly by increasing the source activity, since the chance-coincidence rates were $\leq 20\%$ of the true coincidence counting rates, but the original coincidence circuit was not capable of handling higher singles counting rates.

At approximately 10^4 "start" pulses per minute, the coincidence curve began to be displaced, primarily because of the limitations of the time-to-pulse-height converter of Weber et al. At higher "start" counting rates, the time spectrum became distorted, again because of the limitations of the time-to-pulse-height converter and also because of the inability of the amplifier associated with the 50-channel pulse-height analyzer to amplify small input pulses linearly and without distortion in the presence of a large number of pulses whose voltage is sufficient to saturate the amplifier. At 10^5 "start" pulses per minute, the time spectrum was completely useless for analysis.

At this point the decision was made to modify the spectrometer in an attempt to achieve a higher transmission, and also to construct a new coincidence circuit capable of handling higher counting rates.

D. Modification of Electron-Electron Coincidence Spectrometer

1. Introduction

Several other laboratories have constructed similar thin-lens electron-electron coincidence spectrometers, and they have all generally found them of limited utility. This type of spectrometer has much too small a transmission to become a generally useful research tool. One laboratory has tried to circumvent the difficulty of low transmissions in both halves of the spectrometer by converting the spectrometer into an electron-gamma ray coincidence spectrometer, i.e., one half of the spectrometer has been replaced by a NaI scintillation crystal. Also attempts have been made to use the existing magnets to construct a single high transmission beta ray spectrometer of the Slätis-Siegbahn type.⁸³ Sergienko has modified a thin-lens electron-electron coincidence spectrometer by adding toroidal correcting magnets to each half of the spectrometer.⁸⁴ The original spectrometer had transmissions of 0.006 and 0.013, with associated resolutions of 0.025 and 0.04 respectively, for a 2-mm-diameter source. With the toroidal correcting magnets, a transmission of 0.02 was obtained at a resolution of 0.035. Since the coincidence counting rate depends on the square of the transmission, Sergienko's modification increased the coincidence counting rates by a factor of roughly four.

The most successful electron-electron coincidence spectrometer to date has been constructed to Gerholm.⁸⁵ Each half of this spectrometer is a thick lens iron spectrometer with a triangular-shaped magnetic field, zero at the source and increasing roughly linearly along the axis. Theoretical calculations by Lindgren show that the effect of source width on the resolution is much smaller with a triangular magnetic field than with a uniform magnetic field.⁸⁶

The Gerholm spectrometer has a quoted resolution of 0.013 at a transmission of 0.03 for a 2-mm-diameter source and a resolution of 0.031 at a transmission of 0.03 for a 5-mm-diameter source.⁸⁵ However, most of the work with the spectrometer has been performed at a resolution of 0.025 and transmissions of 0.025 to 0.030 for a 2-mm source.

Since the Gerholm spectrometer has such good characteristics, it seemed worth-while to examine the possibility of obtaining the same magnetic field shape with the existing spectrometer. If the same magnetic field shape could be obtained, a resolution and transmission similar to that of the Gerholm spectrometer might also be obtained.

2. Magnetic Field

The design and construction of the original electron-electron coincidence spectrometer left only a small number of variables by which the magnetic field shape could be changed. The positions of the main magnets M1 and M2 were permanently fixed, while the compensating magnets C1 and C2 could be moved 6 inches along the axis of the spectrometer. The main magnets were divided into three sections, each section containing a total of 950 turns of copper wire. Taps were provided in these sections so that either the full 950 turns or the inner 790 turns could be used. The compensating magnets were divided into two sections each consisting of 550 turns of copper wire. Taps were placed in the coils so that 25, 50, 125, 275, or the full 550 turns could be used.

In order to obtain a "thick-lens" magnetic field the entire number of turns in the compensating magnet C1 was connected in series to the main magnet M1 with the same magnetic polarity. In order to shield the source position from magnetic fields and to make each half of the spectrometer magnetically independent, two large mild steel disks (I1, Fig. 27) 3/4 inch thick by 26 inches in diameter, with 1-1/2-inch-thick iron steps (I3), were placed on either side of the original center section. A 1/4-inch-thick iron band (I2) was also placed around the original center section.

A number of magnetic field determinations were performed with different numbers of turns in the main magnet and various positions of the compensating magnet. With 950 turns in two sections of the main magnet, a magnetic field shape (Fig. 28a) very similar to that of the Gerholm spectrometer (Fig. 28c) was obtained. Both the ordinate and the abscissa of the Gerholm magnetic field plot vs axial distance were

normalized to that of Fig. 28a. The two magnetic field shapes are very similar for axial distances of less than 46 cm from the source. The magnetic field in the Gerholm spectrometer falls rapidly to zero at 46 cm in Fig. 28c owing to the presence of an iron wall. No such magnetic shielding exists in the curve of Fig. 28a, and the magnetic field for distances greater than 46 cm falls off very slowly.

An iron cylinder with a wall thickness of 1/4 inch (I4 Fig. 27) was used to cut off this tailing magnetic field so that a photomultiplier tube (P) could be located near the detector (D). An iron wall similar to that in the Gerholm spectrometer could not be used in this case, since the vacuum pumps used to evacuate the spectrometer were at the extremities of the vacuum tank. Also, the 0.1-inch-thick vacuum tank and original supporting structure could not support the weight of an iron wall. The introduction of this iron cylinder changed the magnetic field a of Fig. 28 rather drastically. Curve b of Fig. 28 shows the best approximation to the magnetic field shape of the Gerholm spectrometer that could be obtained with the inserted iron cylinder. Only 790 turns in two sections of the main magnet were used, instead of the previous 950, for a in Fig. 28.

Another point of interest is the magnetic field at the source position and in the other half of the spectrometer. The magnetic fields shown in a and b of Fig. 28 were determined without the iron steps and without the iron band around the center section. With these added shields, the magnetic field at the source was reduced by a factor of two over those shown in a and b in Fig. 28, so that the magnetic fields at the source position and in the other half of the spectrometer are smaller than in the Gerholm spectrometer. Therefore, the magnetic independence of the two halves of this spectrometer should be much better than that of the Gerholm spectrometer.

The magnetic field at the site of the photomultiplier tube was not completely eliminated by the iron shielding. To reduce the magnetic field to less than 0.1 gauss, two concentric cylindrical mu-metal shields (Sh of Fig. 27) were placed around the phototube P, and over this entire assembly was placed a solenoid magnet So. The solenoid

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Fig. 27. One half of modified electron-electron coincidence spectrometer.

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Fig. 28. Magnetic field along the axis of the spectrometer.
(a) 950 turns in two sections of main magnet - no iron cylinder.
(b) 750 turns in two sections of main magnet - with iron cylinder.
(c) magnetic field in Gerholm spectrometer (normalized).

magnet was constructed so that its magnetic field shape approximated the magnetic field inside the iron cylinder and was of opposite magnetic polarity. By use of a 6-inch light guide and the mu-metal shields, the phototube was completely isolated from magnetic flux at all field strengths below that corresponding to focusing a 650-kev electron. At higher magnet currents the solenoid was used to cancel out most of the magnetic field within the iron cylinder.

3. Construction

The trajectories and the position of ring focus of the electrons were determined by exposing Kodak No-Screen X-ray Film placed in the spectrometer to a Cs^{137} source. The films were placed at various positions in the spectrometer, both parallel and perpendicular to the axis of the spectrometer. The entire baffle system was constructed on a circular brass and aluminum rack so that it could be pushed in and out of the spectrometer for adjustments. After the film exposures the trajectories of the electrons were defined by the conical aluminum outer envelope (F1, Fig. 27) and the inner defining ring (F2). The conical outer envelope was machined in steps to minimize scattering of the electrons from the aluminum.

Once the minimum width of a monoenergetic electron bundle was defined by F1 and F2 (for a solid angle of acceptance from the source of 3% of 4π), finer adjustments of the resolution and transmission were performed in two ways: (a) by screwing the inner defining ring F2 in and out, which required breaking the vacuum, or (b) moving the outer envelope F1 along the axis simply by turning rod R1 from outside the spectrometer. Rod R2 merely prevents the envelope F1 and supporting structure from twisting.

Two types of detectors were used. For electrons more energetic than roughly 100 kev, a Livermore plastic scintillator D of the shape shown in Fig. 27 was used. Because of the rapid divergence of the electron bundle after the ring focus, rather large detectors were required. At the maximum transmission of 0.030 the plastic scintillator had a maximum diameter of 4.5 cm and was 4.5 cm long. At lower

transmissions a smaller detector can be used. For lower-energy electrons and at a transmission of 0.015, an anthracene crystal 3.8 cm in diameter by 0.6 cm thick was used.

4. Performance of Modified Spectrometer

This modification of the spectrometer has decreased the source-to-detector distance from 100 cm to 40 cm and increased the maximum solid angle accepted from the source from 0.4% to 4% of 4π . As a comparison, the source-to-detector distance in the Gerholm spectrometer is 25 cm and the maximum solid angle is 5%. To this date, only the dependence of the resolution and transmission on the position of the inner defining ring F2 have been studied, primarily because of mechanical difficulties in accurately positioning the outer defining envelope. With a 4-mm-diameter electroplated Bi²⁰⁷ source, resolutions of 0.031 and 0.021 were achieved at transmissions of 0.031 and 0.013 respectively. Figure 29 shows the K-conversion-electron line of the 570-keV transition at a resolution of 0.021. These figures represent an increase in the transmission of slightly greater than a factor of six at the same resolution as the original spectrometer. This represents a substantial improvement over the original spectrometer, but the modified spectrometer is still inferior to the Gerholm spectrometer. The difference in the characteristics of the two spectrometers may be due to the slightly different magnetic field shapes but the most probable cause of the poorer characteristics may be deviations of the magnetic field from axial symmetry. The field was found not to be axially symmetric during the initial experiments and attempts were made to correct this by re-aligning the magnets. It is difficult to produce an axially symmetric magnetic field over the entire volume of this spectrometer, since (a) the actual length of the magnets is roughly only one-third of the length of the spectrometer, and (b) the coils are wound randomly with a relatively small number of turns of thick wire, and hence the magnet coils themselves are probably not axially symmetric. On the other hand, an axially symmetric field is much more easily achieved in a spectrometer such as

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Fig. 29. K internal-conversion electron line of the 570-kev transition in Pb^{207} .

the Gerholm spectrometer, since the coils producing the magnetic field are wound over the entire length of the spectrometer and the entire coil system is surrounded by iron.

The current required to focus the electrons in the modified spectrometer was found to be directly proportional to their momenta or magnetic rigidity H_p :

$$H_p = 456.0 I,$$

where H_p is in gauss-cm and I is in amperes.

The modified spectrometer requires currents 1.75 times those in the original spectrometer to focus an electron of given energy.

The source positioning was found to be rather critical. With a 3-mm-diameter source, no appreciable change in resolution or transmission was observed if the source was displaced ± 1.5 mm from the true position. For displacements of ± 4 mm perpendicular to the axis of the spectrometer, the resolution increased from 0.02 to 0.04 with the peak counting rate, and hence the transmission decreasing by a factor of two.

5. Discussion

Since the coincidence counting rates are proportional to the product of the transmissions of both halves of the spectrometer, this modification has increased the coincidence counting rates by a factor of roughly 40 over that of the original spectrometer at the same resolutions. Also a very simple adjustment of the inner defining rings will enable the present spectrometer to operate at a slightly poorer resolution of 0.03 in one half or in both halves of the spectrometer, with an increase in the coincidence counting rates to 85 or 225 times the original counting rates respectively.

The coincidence circuit shown in Fig. 6 with the time-to-pulse-height converter shown in Fig. 15 will be used with the modified spectrometer. This new circuit is much more stable over long periods of time than the original coincidence circuit, and is capable of handling much higher counting rates without distortion of the time spectra.

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