FISSION YIELDS OF CESIUM, RUBIDIUM, AND STRONTIUM ISOTOPES AND THEIR RELATION TO FINE STRUCTURE IN FISSION

by

Donald Roy Wiles, B.Sc., B.Ed.

A Thesis

Submitted in Partial Fulfilment of the Requirements

for the Degree of Master of Science

McMaster University

September 1950

The author of this thesis holds the following degrees:

Bachelor of Science, Honour Chemistry, 1946 (Mount Allison)

Bachelor of Education, 1947 (Mount Allison)

This thesis was prepared under the supervision of:

Professor H. G. Thode, Department of Chemistry

A B S T R A C T

The various concepts of the fission process, mass and charge distribution, and fine structure in fission yields are reviewed. Details and results of mass spectrometric analyses of fission-product samples of cesium, rubidium, and strontium are reported. Absolute yield values have been assigned to the mass 87, 135, and 137 chains. Relative yields of the mass 88 and 90 chains have been determined. A mechanism is postulated to account for anomalies in fission fine structure. The Glendenin mechanism of instantaneous neutron emission is found to require modification.

Acknowledgements

The author wishes to acknowledge the encouragement of Dr. H. G. Thode, under whose direction this work has been done. He is especially indebted to Mr. R. J. Horsley and Mr. B. W. Smith, who have spent many hours analysing samples. Without their cooperation this work could not have been done.

TABLE OF CONTENTS

Table of Contents (Continued) Page

INTRODUCTION

The method of mass spectrometer analysis has been applied by Thode and his co-workers[1,2] to the determination of relative and absolute fission yields with high precision. Their investigations of the isotopes of xenon and krypton originating in the fission of uranium-236 have revealed fine structure in the mass-yield curve in the neighbourhood of the eighty-two neutron shell. Recently, Inghram, Hayden and Reynolds[3] reported mass spectrometer abundance data for the fission product isotopes of cesium. Their abundance data for these isotopes indicate an abnormally high yield for cesium-135, which cannot be explained on the basis of present theories. Since the mass chains ending in isotopes of cesium fall in the neighbourhood of the eighty-two neutron shell, and are predicted by the Glendenin mechanism of chain branching[4] to have low yields, it seemed important to redetermine the abundances of the cesium isotopes and to extend the work to the isotopes of other elements which fall in either the eighty-two or the fifty neutron shell region. This has now been done, and the results are reported in this thesis.

The Fission Process and Mass Distribution

When a nucleus is subjected to irradiation with high energy particles or electromagnetic radiation, various events are likely to occur, depending on the nature and energy of the radiation, and on the nature of the target nucleus. At low energies of incident charged particles, elastic scattering occurs. Here the most serious effect is the random removal of a few of the electrons from atomic orbitals.

Neutrons at different energies, and charged particles at high energies

are likely to cause one or more of the following processes:

1. Absorption of the incident particle or photon. In this case an excited nucleus is formed which emits some form of radiation in its return to the ground state. Examples of such reactions are the familiar (n,γ), (γ,n), (α,p), (α,n), (d,p), and many others.

2. Spallation. This occurs more frequently in larger nuclei, and only with high-energy particles. It is the splitting off of a light fragment from the original nucleus, leaving a resultant nucleus which is lighter by sometimes as much as fifty mass units.

3. Fission. This important reaction results when the target nucleus, necessarily a heavy nucleus, splits into two (or rarely, three)[5] parts of roughly equal size. It is this reaction and some of its ramifications which are the main topic of this thesis.

Fission takes place in several ways, and under the excitation of nearly all forms of energetic radiation. The first fission reaction discovered was that of uranium-235 bombarded with slow neutrons[6]. It is the most important of all modes of fission, because it can easily be adapted for use in self-sustaining nuclear reactors. The reaction may be represented by the following equation:

$$_{92}U^{235} + {}_{0}n^{1} \longrightarrow (_{92}U^{236}) + {}_{Z_1}X^{A1} + {}_{Z_2}Y^{A2} + \nu\,{}_{0}n^{1} + \gamma + Q$$

where X and Y represent the two fission products, ν the number of neutrons released in the fission process (about 2.3 in this particular reaction), the release of gamma ratiation, and Q the kinetic energy of the fission fragments and neutrons. Other forms of fission are listed in Tables I and II.

Because the ratio of neutrons to protons in the uranium-235 nucleus is higher than in lighter nuclei, the nuclear fragments formed are unstable. They usually undergo a series of successive beta decays. As result of the reaction

$$0n^1 \longrightarrow 1p^1 + \beta^-$$

the original nucleus is transformed eventually into a nucleus with the same mass, but a lower, more stable neutron-proton ratio. This groups the fission products into chains, of which some sixty-four are known.

An important aspect of the study of fission is the investigation of the distribution of mass among the possible fission products. Isotopes of xenon and krypton are the stable end products of several chains. The relative abundances of these isotopes can be measured with very high precision by a mass spectrometer. This gives accurate values for the relative yields of these mass chains in fission. Another method of determining yields involves the measurement of the radioactivity of separated fission products. The mass yield curves for uranium-235, uranium-233, and plutonium-239 are given in Figures 1 and 2. It is seen that there is a high probability for the splitting of a large nucleus into two fragments of masses about 96 and 139. The probability of formation of fragments of equal size is low. Spontaneous fission and the slow neutron fission of other elements is expected to follow essentially the same pattern.

Not all the fission processes take place in this asymmetric manner, and for fission induced by high-energy projectiles, the fragments are most often of equal mass. For example, the mass-yield curve for fission of thorium-232 with 37.5 mev alpha particles, (Figure 3), still shows asymmetric fission, but the very shallow minimum indicates a higher proportion

TABLE I

Some Fission Modes of Uranium and Thorium Isotopes

Nucleus	Activation	Cross Section (barns)
U^{233}	Thermal Neutrons[7]	
U^{235}	Thermal Neutrons[6]	
U^{238}	Thermal Neutrons	
U^{238}	Fast Neutrons	
U^{238}	Protons	
U^{238}	Deuterons[8,9]	5×10^{-3}, 2.2×10^{-2}
U^{238}	Alpha Particles (9 mev)[10]	5×10^{-3}
U^{238}	Gamma Radiation[11]	3.3×10^{-3}
$U^{?}$	Spontaneous[12,13]	$T_{\frac{1}{2}} = 2.1 \times 10^{16}$ yr.
Th^{230}	Thermal Neutrons[14,15]	$2.5 \times$ Th^{232}
Th^{232}	Thermal Neutrons	
Th^{232}	Protons[16]	
Th^{232}	Deuterons[17,18,19]	3.5×10^{-3}
Th^{232}	Alpha Particles[18]	
Th^{232}	Gamma Radiation[11]	1.7×10^{-3}
Th^{232}	Spontaneous[19]	$T_{\frac{1}{2}}$ 10^{19} yr.

TABLE II

Some Fission Modes of Heavy Elements with Energetic Particles. (After Perlman et al.[20])

Element	Projectile	Energy (Mev)	Element	Projectile	Energy (Mev)
Bi		400	Pb	n	100
Bi	d	50	Tl		400
Bi	n	100	Tl	d	200
Pb		400	Pt		400
Pb	d	100	Ta		400

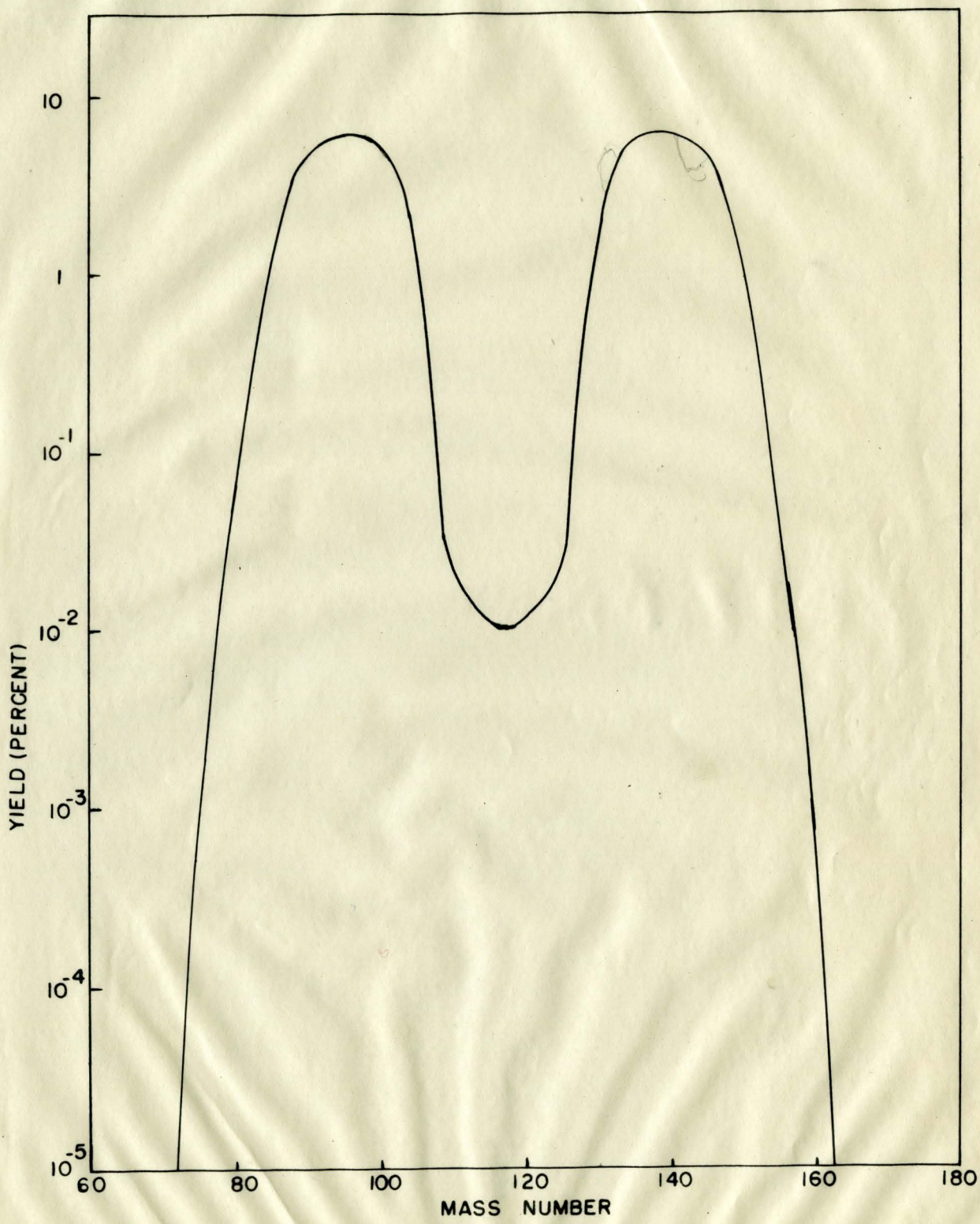

FIG. 1. FISSION YIELDS IN URANIUM-235

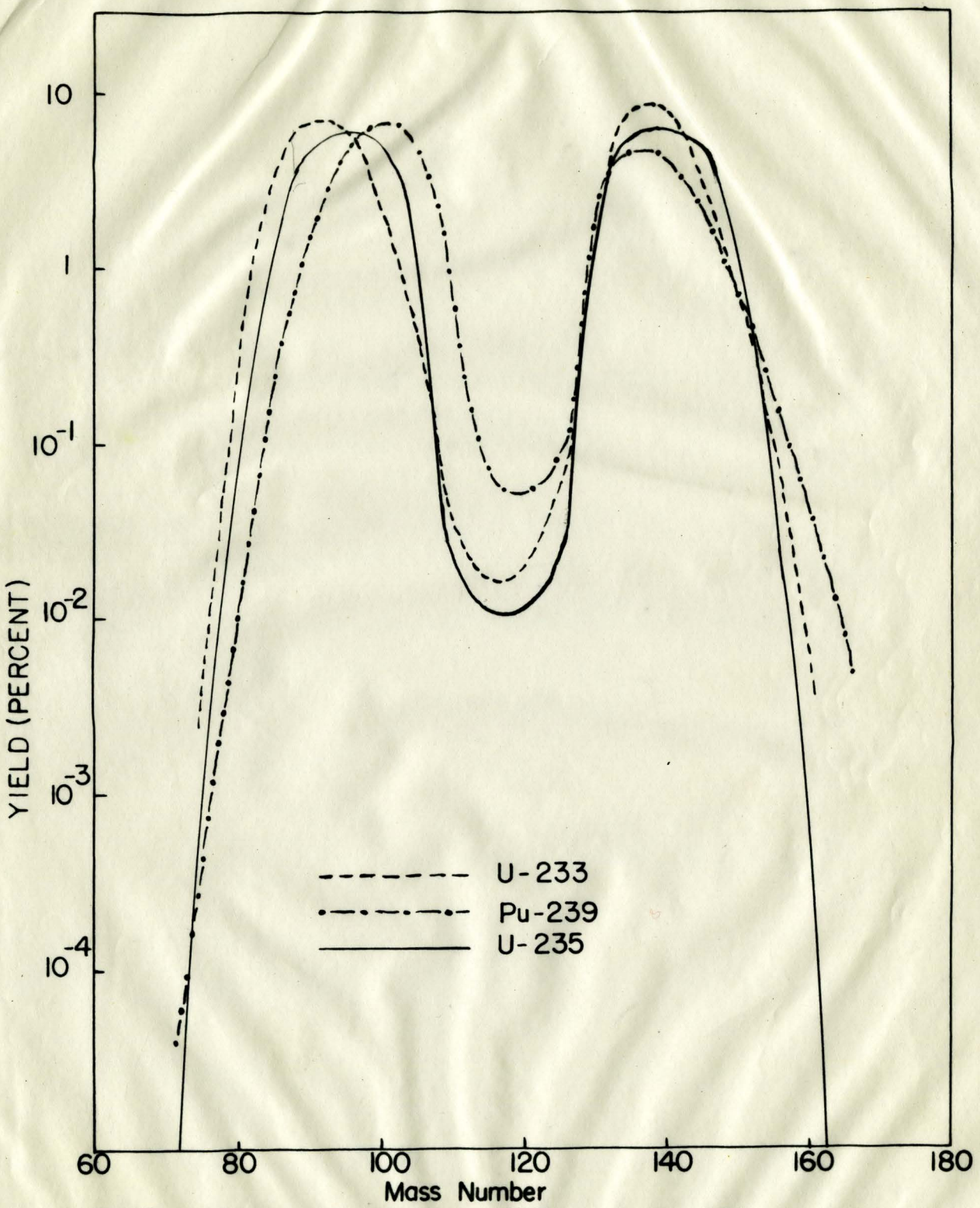

FIG. 2. FISSION YIELDS IN U-233 AND Pu-239. Showing shift from U-235 fission yields.

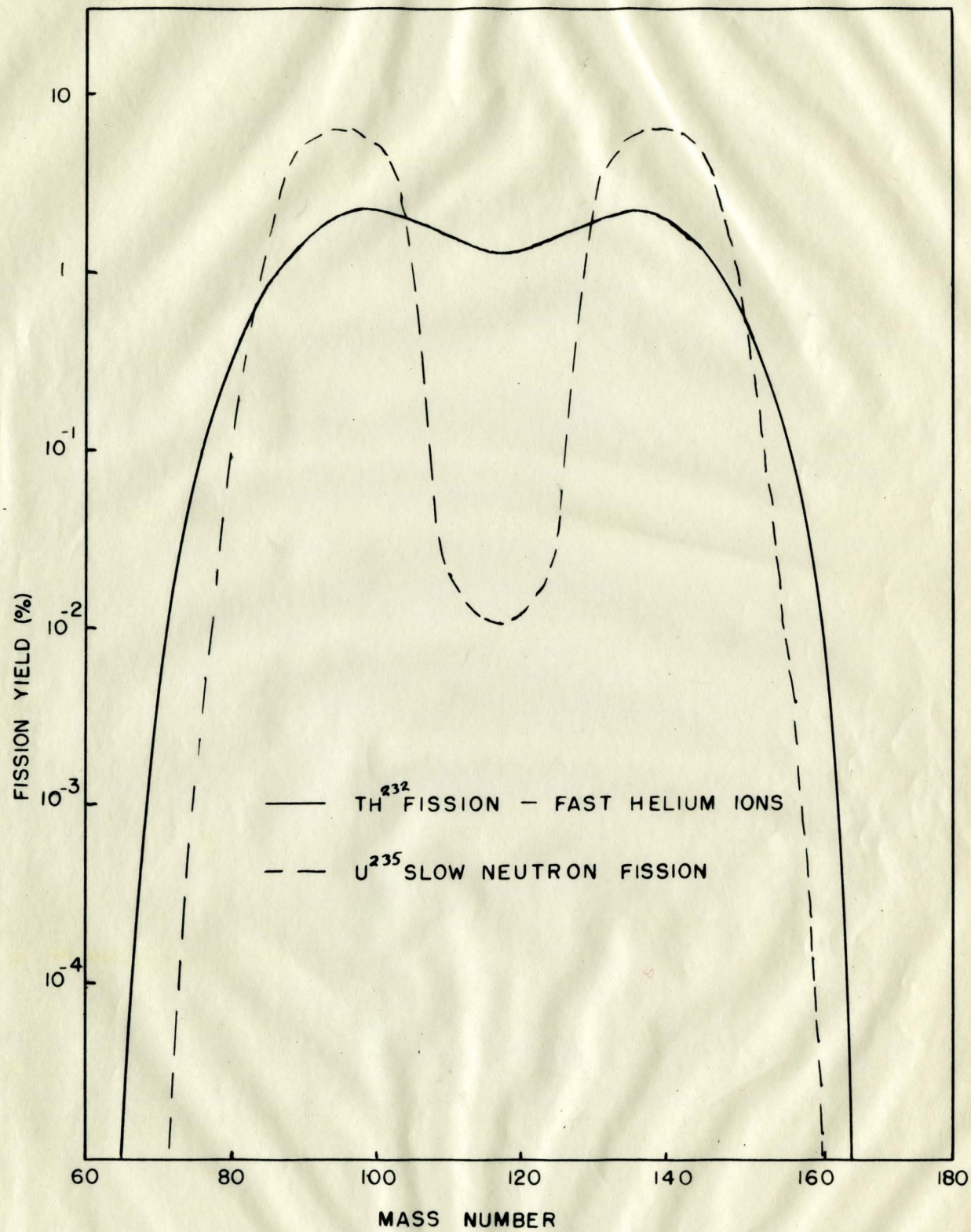

FIG. 3. MASS-YIELD CURVE FOR THORIUM FISSION WITH 38 MEV HELIUM IONS.

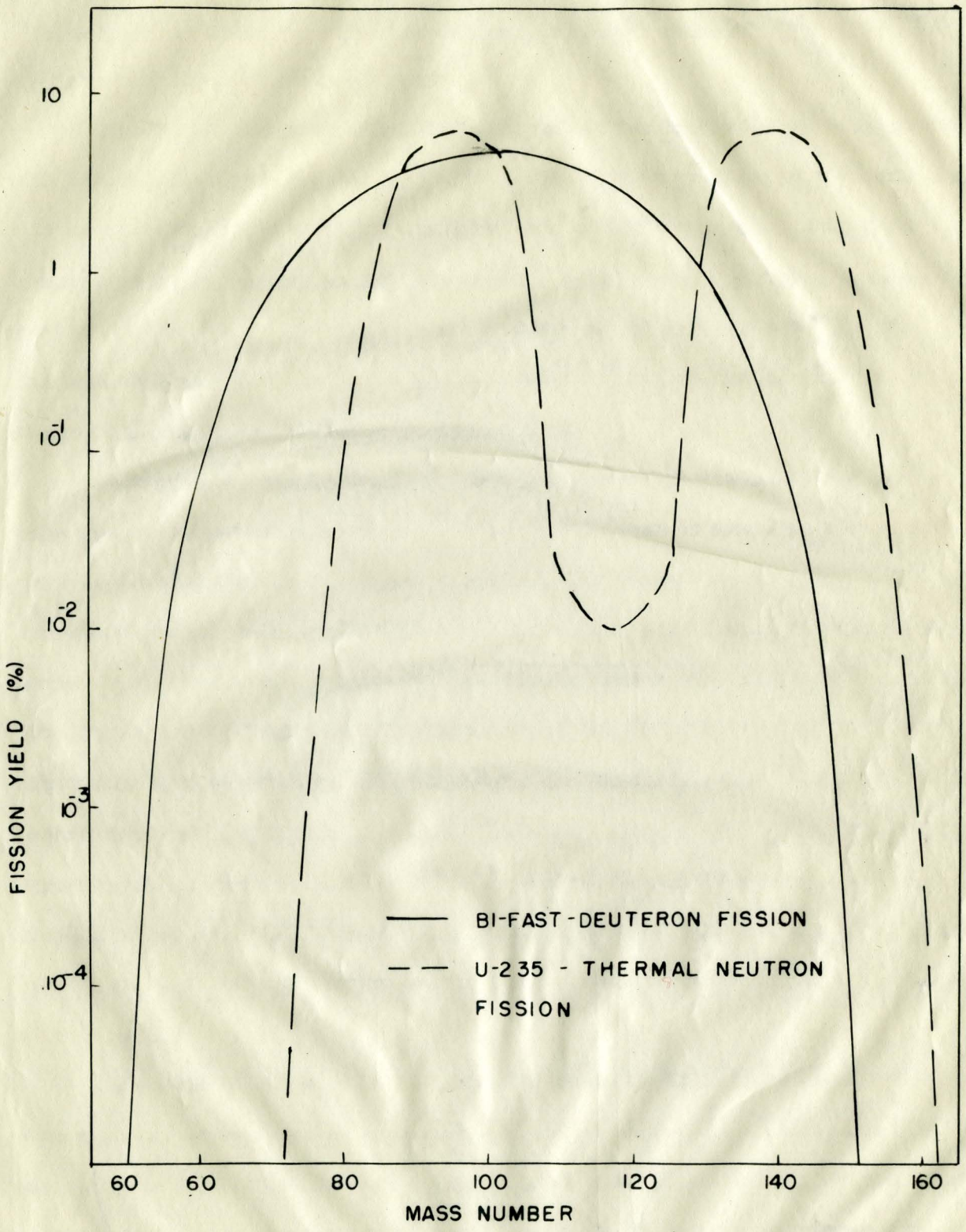

FIG. 4. MASS-YIELD CURVE FOR BISMUTH FISSION WITH 190 MEV DEUTERONS.

of symmetric fission. The fission of Bismuth with 190 Mev deuterons[21,22] (Figure 4) is completely symmetrical.

Mayer[23] has noted that, in cases of asymmetric fission, the location of the maxima indicate that the most probable primary fission products might be those with fifty or eighty-two neutrons*. It seems, then that even during the fission process the new nuclei tend to approach stable configurations. Meitner[24] has noted that in the case of deuteron fission of Bismuth, where the fissioning nucleus, Po^{199}, has too few neutrons to allow such a distribution, fission is symmetrical.

Goeckermann and Perlman[22] have proposed that the speed with which the fission process takes place determines whether or not a rearrangement into a stable configuration is possible. For elements near uranium-235 in the periodic table, the energy of excitation for fission is approximately 6 mev, that is, roughly equal to the binding energy of a slow neutron. Thus, for slow-neutron bombardment of such a nucleus, the probability of fission occurring is rather high. If, however, the threshold energy for fission is about twice as high as in the previous case, neutron emission is favoured over fission. Several neutrons may be released, simultaneously, and the resultant nucleus will have a much lower fission threshold than the original nucleus. This new fissioning nucleus is highly excited, and will distinegrate very rapidly.

The theory is, then, that it would be energetically uneconomical for one primary product to be β^+ unstable while the other is β^- unstable. Therefore, a complete redistribution of nucleons is in order, and actually takes place in the case of slow-neutron fission. Here the fission takes place before the neutrons are emitted, and the redistribution is possible. On the other

---

*See Appendix for a note on nuclear shell structure.

hand, if the neutrons are emitted prior to fission, thus causing very rapid fission, then we might expect the distribution of neutrons and protons to be random, and the fission products to be of equal mass. Such is the case with the fission of deuteron-bombarded bismuth, where the release of ten or twelve neutrons gives rise to the excited nucleus Po^{199}, which then undergoes very rapid symmetrical fission.

Charge Distribution in Fission

The problem of the distribution of charge in fission is perhaps more difficult to study than the distribution of mass. While the mass of a fission fragment remains, with the exceptions which will be noted later, the same throughout all its decay transformations, the charge changes very rapidly. Mass distribution determinations may be made on stable or long-lived isotopes, in which case they are usually determinations of the cumulative mass yield. On the other hand, charge distribution must take into account the very short-lived isotopes, many of which are very difficult to separate in a short time.

Normally a fragment is found to undergo an average of about three beta disintegrations before finally arriving at a stable state. The charge of the primary fission product, then, is considerably displaced from the charge of the nucleus which, according to Bohr and Wheeler[25], is the most stable configuration for that mass. If Z_A is the calculated most stable nuclear charge (not necessarily integral) for a given mass, and the most probable primary fission fragment has a nuclear charge Z_p, the 'displacement' of the primary nucleus is $Z_A - Z_p$. The problem, then is to find an expression for Z_p or for the displacement.

Several propositions have been made to explain the known facts:

1. The simplest theory is that of unchanged charge distribution. The ratio of neutrons/protons is the same in each of the two fragments from a single fission. For uranium-235 fission, this ratio would be (236 - 2.3)/92. This postulate seems to apply to fission induced by high-energy particles, but not to slow neutron fission[22].

2. The postulate of minimum potential energy is that the sum of the energies of radioacitve decay of the two fission fragments and the energy of electrostatic repulsion at the moment of fission is a minimum.

3. The postulate of equal charge displacement, derived empirically by Glendenin, Coryell, and Edwards[26], states that the most probable distribution of charge from fission is such that the two fragments from a single fission are equally displaced from the most stable charge. That is, $(Z_A - Z_P)_1 = (Z_A - Z_P)_2$. The most probable primary charge, then, may be calculated from the equation.

$$Z_P = Z_A - 0.5\ (Z_A + Z_{(233.7 - A)} - 92)$$

4. Present's theory of non-uniform charge distribution[27] has been developed theoretically from a spherical nuclear model, in which the distribution of protons is radially non-uniform. For fission where the mass ratio of the light and heavy fragments is about 2:3, this theory leads to approximately equal values for the charge displacement of primary fission products, and thus it is similar to the theory of equal charge displacement. For very unsymmetrical fission, however, where the mass ratios are about 1:2, the lower mass chains are predicted to have smaller displacement values than the heavier chains.

Glendenin[4], on examining these four theories, found that the first two were untenable for slow-neutron fission. The available data seem to check equally well with both of the other two, the equal charge displacement theory, and the non-uniform charge distribution theory, and at present it is not possible to choose between them. Glendenin utilized the known independent fission yields of the shielded nuclei Iodine-134, iodine-134, xenon-135, cesium-136, bromine-82, and rubidium-86. These yields he plotted as a function of the displacement from the most probable charge, as calculated from each of the two theories. These data give the curves shown in Figure 5. The yields which Glendenin determined lie quite well on both curves. Although iodine-133 seems to be an exception, it is suspected that the experimental data on this isotope may be in error. In all this work, he assumed that one distribution curve, represented by

$$P(Z) = a\, e^{-b(Z - Z_p)}$$

will fit all mass chains.

Fine Structure Effects in Fission

The mass yield curves published by the Plutonium Project[28] and by Grummitt and Wilkinson[29] were obtained from early radiochemical data. The experimental values at that time indicated a smooth curve, and all values lay close to the curve. The one point which lay very far from the curve, tin-123, is explained by the assumption that it is associated with a stable or long-lived isomer which has so far escaped radiochemical detection. It was suggested by Kingdon[30] that nuclear stability considerations might predict a certain favouring of fragments with even numbers of protons. However, this has not yet received experimental support.

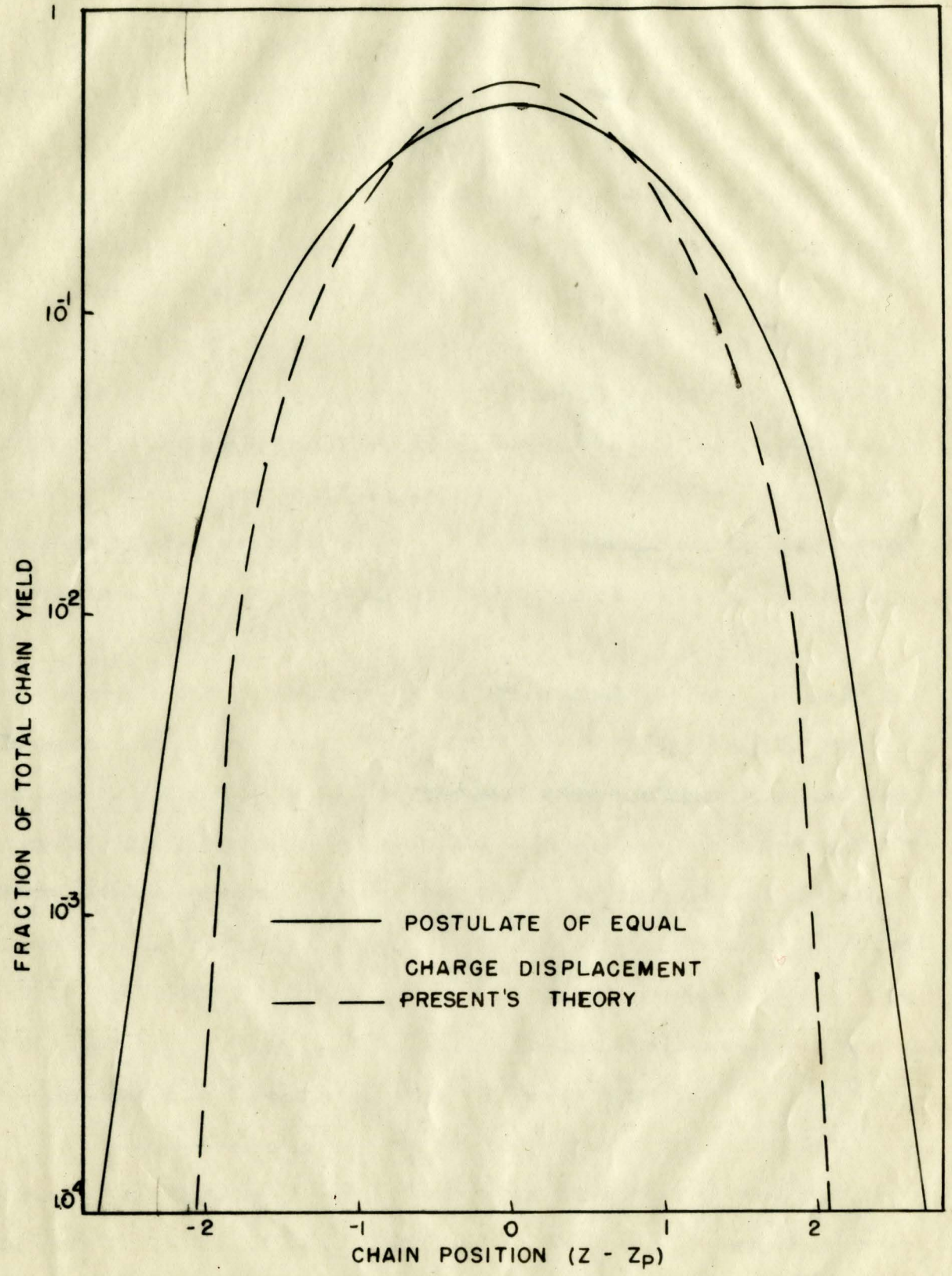

FIG 5. CHARGE DISTRIBUTION IN FISSION

In addition to this one case, tin-123, which can easily be explained, others have been reported which are very difficult to account for, Thode and Graham[1] have reported anomalous yields of Krypton and xenon isotopes from fission. The values for krypton-84, xenon-133[2] and xenon-134 are all considerably above the smooth curve, although the other isotopes of these elements lie right of the curve. The results of the xenon measurements are shown in Figure 6. Stanley and Katcoff[31] have found that the cumulative yield of iodine-136 is very low, although the yield of xenon-136 appears to be on the curve. Inghram, Hayden and Reynolds[3] have measured the relative abundances of the cesium isotopes from fission mass spectrometrically, and their results did not conform to expectations. The results of all these determinations are given in Table III.

A qualitative explanation for this fine structure has been given by Glendenin. It is well known[23] that nuclei with fifty or eighty-two neutrons show a greater stability than most other configurations, and that in nuclei with fifty-one or eighty-three neutrons, the binding energy of the 'extra' neutron is somewhat lower than normal, by approximately 2-3 mev. It has been postulated by Glendenin[4] that in cases where an eighty-three or fifty-one neutron nucleus is formed as a primary fission fragment, this extra neutron is emitted instantaneously, instead of the usual beta particle. This proposed chain branching is shown in Table IV and V.

An example will help to point out the effect of this mechanism. Following the Glendenin theory of charge distribution, the total primary yield of tin-133 is 0.3 percent of fission. This will be completely transformed into tin-132 by instantaneous neutron emission. Thus, the yield of

the mass 132 chain is predicted to be 0.3 percent of total fission higher than the smooth curve value, or 4.5 percent of fission. The total primary yield of antimony-134 - 1.3 percent, is postulated to transform into antimony-133. The total effect on the mass 133 chain is a loss of 0.3 percent, of total fission a gain of 1.3 percent, of a net gain of 1.0 percent. Thus, the predicted value for the yield of the mass 133 chain is 6.0 percent.

Glendenin has calculated that if one assumes Present's theory of charge distribution, then it is necessary to postulate only sixty percent neutron emission, instead of one hundred percent. The values calculated from both theories are given in Tables VI and VII.

Up to the present, only a few chains have been measured accurately. Moreover, all the fine structure exhibited by these chains has shown an increased yield. It would, therefore, be of considerable interest to investigate some of the chains for which low values are predicted. It was with this in view that the present investigation was undertaken.

The relative abundances of the fission product isotopes of cesium have been redetermined. At the same time, the relative abundance of the rubidium isotopes of mass 85 and 87 have been determined. This will make possible a close estimate of the absolute yield of the mass 87 chain.

Two fission product chains end in stable or long-lived isotopes of Strontium, those with masses 88 and 90. In this case, it will only be possible to determine relative yields since little absolute yield data is available in this mass range. The relative yields of these two isotopes have been determined.

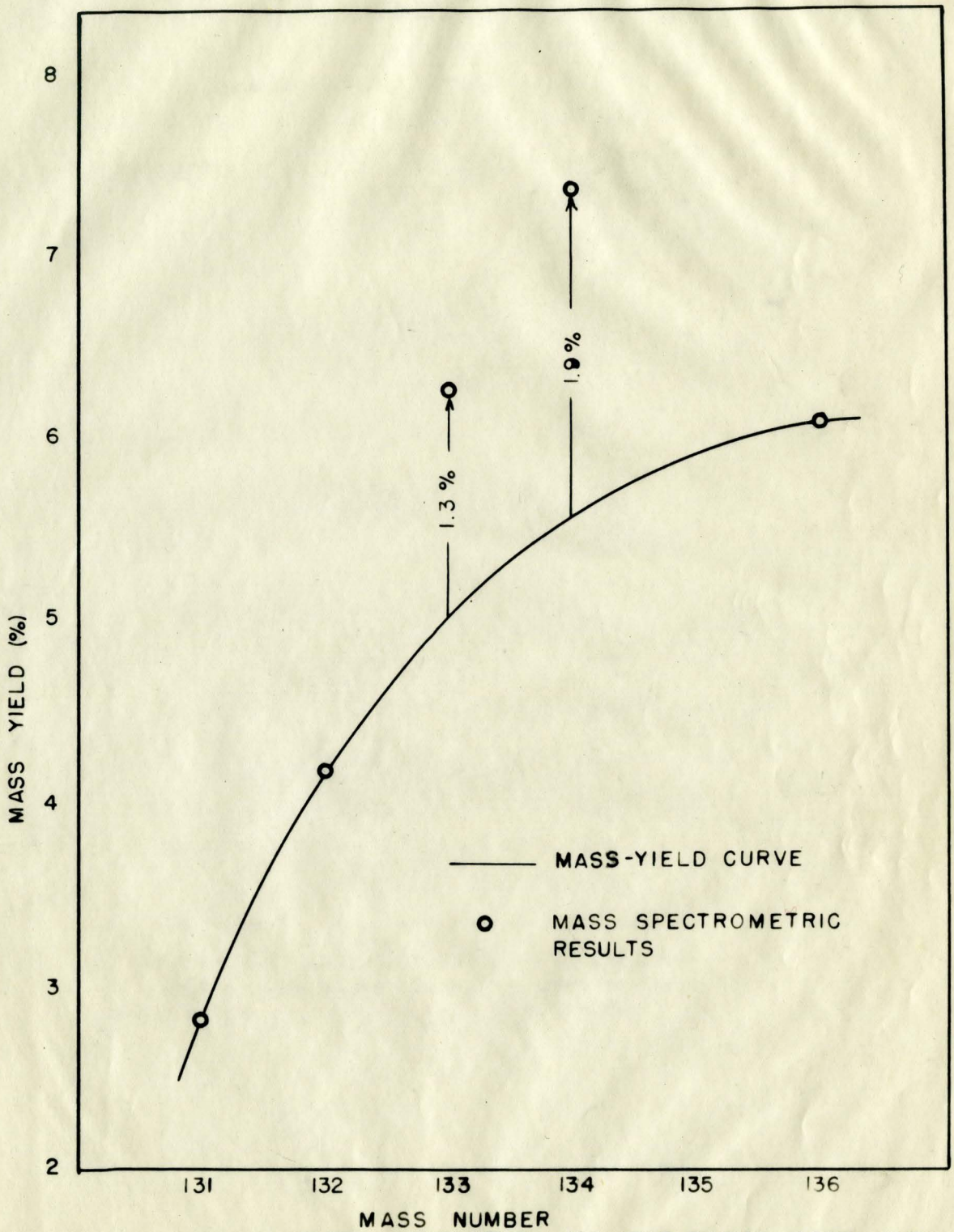

FIG. 6. FINE-STRUCTURE IN FISSION YIELDS

TABLE III

Anomalous Fission Mass-Yields

Mass Chain	Yield	
	Smooth Curve	Experimental
83	0.5	0.39
84	0.8	1.1
85	1.3	- -
86	2.0	2.1
87	2.9	- -
131	2.8	2.8
132	4.2	4.2
133	5.0	6.3
134	5.5	7.4
135	5.9	7.8[a]
136	6.1	6.1
137	6.2	6.1[a]
I^{136}	5.2	3.1

[a] These are the values as determined by Inghram, Hayden, and Reynolds. They have been redetermined as part of the work of this thesis.

TABLE IV

Chain Branching in the Higher Mass Range.

Nuclear Charge						
50	51	52	53	54	55	56
Sn^{131}	Sb^{131}	Te^{131}	I^{131}	$\underline{Xe}^{131}$		
Sn^{132}	Sb^{132}	Te^{132}	I^{132}	$\underline{Xe}^{132}$		
n↑						
Sn^{133}	Sb^{133}	Te^{133}	I^{133}	Xe^{133}	$\underline{Cs}^{133}$	
	n↑					
	Sb^{134}	Te^{134}	I^{134}	$\underline{Xe}^{134}$		
		n↑				
	Sb^{135}	Te^{135}	I^{135}	Xe^{135}	$\underline{Cs}^{135}$	
			n↑			
	Sb^{136}	Te^{136}	I^{136}	$\underline{Xe}^{136}$		
				n↑		
		Te^{137}	I^{137}	Xe^{137}	$\underline{Cs}^{137}$	$\underline{Ba}^{137}$
					n↑	
		Te^{138}	I^{138}	Xe^{138}	Cs^{138}	$\underline{Ba}^{138}$

Arrows indicate the branching of a chain by emission of a neutron from a primary fission product having eighty-three neutrons. Nuclides under lined are the effective ends of the respective chains.

TABLE V

Chain Branching in the Lower Mass Range

Nuclear Charge						
34	35	36	37	38	39	40
Se^{84}	Br^{84}	Kr^{84}	Rb^{84}	$\underline{Sr}^{84}$		
n↑						
Se^{85}	Br^{85}	Kr^{85}	Rb^{85}			
	n↑					
Se^{86}	Br^{86}	Kr^{86}	Rb^{86}	$\underline{Sr}^{86}$		
		n↑				
	Br^{87}	Kr^{87}	$\underline{Rb}^{87}$			
			n↑			
	Br^{88}	Kr^{88}	Rb^{88}	$\underline{Sr}^{88}$		
				n↑		
		Kr^{89}	Rb^{89}	Sr^{89}	$\underline{Y}^{89}$	
					n↑	
		Kr^{90}	Rb^{90}	Sr^{90}	Y^{90}	$\underline{Zr}^{90}$
						n↑
		Kr^{91}	Rb^{91}	Sr^{91}	Y^{91}	Zr^{91}

Arrows indicate the branching of a chain by emission of a neutron from a primary fission product having fifty-one neutrons. Nuclides underlined are the effective ends of the respective chains.

TABLE VI

Fission Yields in the Region of the Fifty-Neutron Closed Shell

Mass Number of Chain	Fission Yields			
	Smooth Curve (Experimental)	Mass Spectrometry (Experimental)	Glendenin (Calculated)	Present (Calculated)
83	0.5	0.39	0.7	0.6
84	0.8	1.1	0.9	1.1
85	1.3		1.3	1.5
86	2.0	2.1	1.8	1.8
87	2.9		2.6	2.5

TABLE VII

Fission Yields in the Region of the Eighty-Two Neutron Closed Shell

Mass Number of Chain	Fission Yields			
	Smooth Curve	Mass Spectrometry	Glendenin	Present
131	2.8	2.8	2.9	2.8
132	4.2	4.2	4.5	4.2
133	5.0	6.3	6.0	5.4
134	5.5	7.4	6.5	6.7
135	5.9		5.7	6.4
136	6.1	6.2	5.5	5.9
137	6.2		4.9	4.5
138	6.2		5.7	6.0

EXPERIMENTAL

Chemical Procedures

a. Cesium and Ribidium

The procedure adopted for the simultaneous isolation of carrier-free cesium and rubidium from irradiated uranium is as follows. The uranium metal was dissolved in nitric acid, and the solution evaporated to crystals. The crystals were dissolved in diethyl ether, and the fission products separated from this solution by extraction with several very small portions of dilute nitric acid. It was sometimes necessary to repeat this extraction procedure on the aqueous extract to remove excessive quantities of uranium. Two or three milligrams of ferric ion was added, and subsequently precipitated as hydroxide with ammonia gas. This removed essentially all of the hydroxide-forming elements, and left in solution nearly all of the cesium, rubidium, barium, and strontium. The solution was evaporated, converted to the chloride, fumed, and redissolved in 6N hydrochloric acid. Two milligrams of ammonium ion was now added, and precipitated as ammonium chloroplatinate. This precipitate carried down cesium and rubidium only. The supernatant solution at this point was used for subsequant isolation and purification of strontium. The chloroplatinate crystals were dissolved in hot water, and the platinum was removed by reduction with formic acid. The solution was again evaporated, and fumed. The carrier-free cesium and rubidium chlorides were converted to nitrates by repeated evaporation with nitric acid. The resultant nitrates were in a form satisfactory for mass spectrometric analysis.

In order to determine the amount of contamination by natural cesium and

rubidium, a blank was prepared in the following way. Two uranium discs of equal weight, one disc irradiated, and the other not, were dissolved separately, in nitric acid. They were treated in the manner just described. Care was taken to ensure that exactly the same quantities of reagents were added to each sample. After the redissolution of the chloroplatinate crystals, half of each sample was removed, and the two halves combined. In this manner, the synthetic sample is made up to have exactly twice as much natural cesium and rubidium as the original active sample. A comparison of the abundances of isotopes in the two active samples will make possible a determination of contamination-free isotopic abundances. In subsequent runs, the combining of samples was done immediately following the dissolution of the uranium metal. In this way, the efficiency of extraction and precipitation procedures has no effect on the accuracy of the blank.

b. Strontium

The isolation of carrier-free strontium was completed by the following process. The strontium solution, after the chloroplatinate precipitation just described, was evaporated to dryness and fumed. Contaminants at this point were silica, platinic chloride, some organic matter, and small amount of iron and uranium. The residue was dissolved in dilute hydrochloric acid, and two milligrams of ferric ion was added. This was precipitated as ferric hydroxide at pH 9 with ammonium carbonate. This caused the quantitative sorption of strontium carbonate and/or strontium hydroxide. In this way, the desired component was separated from all substances soluble in basic solutions.

This step was followed by redissolution in an excess of hydrochloric

acid, and repricipitation of the hydroxide with carbonate-free ammonia gas, at pH 6. This caused the coprecipitation of all hydroxide components, but the strontium was not adsorbed[32,33]. The supernatant solution was evaporated and fumed. The residue was essentially pure barium and strontium chlorides, which were converted to nitrates by evaporation with nitric acid. The analysis of strontium isotopes was done on this nitrate residue.

Mass Spectrometer Analysis

(a) Dempster-Type, Double Focussing Mass Spectrometer.

This type of instrument, first described by Dempster[34,35], was originally used as a mass spectrograph, for absolute mass determinations. The instrument used in this work[36] was modified so that the ions could be collected, and ion currents measured, instead of recorded photographically. This change was made to permit high-precision abundance measurements. The ions were produced in a modified Shaw-type crucible source[37]. All crucibles used in this work were made of columbium, 0.06" O.D. x 0.03" I.D. x 3/8".

Analyses were made by measuring singly-charged ions emitted from the oxide of the element under study. The oxides were used because they were found to give more steady emission than any other compounds investigated. The crucible temperature of cesium and rubidium analyses was estimated at between 350 and 400° C. Strontium analyses required much higher temperatures - in the range between 1700 and 1800° C. Before each analysis a blank crucible was run in the mass spectrometer to ensure that no residual ion currents were being caused by ions being baked out of other parts of the source assembly.

(b) 90° Sector-type Mass Spectrometer

The 90° mass spectrometer originally described by Graham, Harkness, and Thode[38] was modified to permit the use of a heated filament ion source. Ions of cesium and rubidium were produced from the oxides at temperatures estimated at between 600 and 650° C.

(c) Sample Sizes

It was found that satisfactory ion currents could be obtained with very small samples of material. For example, the cesium results were obtained with less than 0.1 microgram of cesium oxide. In the case of rubidium, the samples were considerably smaller, because of the lower fission yields of rubidium isotopes, possibly of the order of 0.02 micrograms. The lower limit has not been investigated, but it is felt that the limit of detection for cesium and rubidium is less than 0.001 microgram. 0.5 to 1 microgram of strontium was required in order to obtain satisfactory ion currents with the heated crucible ion source.

(d) Sample Fractionation

It is claimed by Inghram, Hayder, and Reynolds[3] that a fractionation of cesium isotopes of as much as five percent occurs at the filament of their mass spectrometer. We have found no indication that this fractionation takes place in our instruments[36,39]. In our work, two different mass spectrometers were used which had two widely different types of ion source. In every case, the results obtained from the two instruments were in excellent agreement. No differences in the isotopic ratios has been noted between the first and last ten percent of each sample. We have therefore assumed that no fractionation of the isotopes occurred with our samples.

Preparation of Samples

(a) Cesium and Rubidium

The preparation of samples of cesium and rubidium was relatively simple. The method involved merely evaporating a small portion of the total sample of cesium and rubidium on the surface of the tungsten filament, or in the columbium crucible.

(b) Strontium

The preparation of samples of strontium was very much more difficult than for cesium and rubidium because the ionizing efficiency of the mass spectrometers for alkaline earth oxides is considerably lower than for alkali metal oxides. It was not possible to investigate fission product strontium with the 90° filament source mass spectrometer, as the samples of strontium oxide available were too small to give satisfactory ion currents. Even with the heated crucible source it was just possible to get results with the one microgram extracted from our irradiated discs.

Since evaporation of larger amounts than one or two microliters in such a small crucible presented prohibitive difficulties, the carbonate was precipitated on a small amount of ferric hydroxide as carrier, and centrifuged into the crucible by means of a specially adapted centrifuge tube.

Results

(a) Cesium

The mass spectrometer abundance data must be corrected for the radioactive decay of cesium-137 in order to obtain the true mass yields. This

TABLE VIII

Yields of Cesium Isotope From Thermal Neutron Fission of Uranium-235

Mass	Absolute Yield
133	6.29[a]
135	6.18[b]
137	5.87[c]

[a] Value for Xe^{133} based on 2.8% yield of I^{131}

[b] Corrected for neutron absorption

[c] Corrected for decay using 33-year half-life.

correction was made by extrapolation to zero time, on the basis of a 33-year half-life[40]. The value obtained for the yield of cesium-135 will also be low, because of the neutron absorption reaction: Xe^{135} (n,γ) Xe^{136}. The final results after correcting for decay are given in Table VIII. The results obtained are plotted in Figure 7.

TABLE VIII

Yields of Cesium Isotope From Thermal Neutron Fission of Uranium-235

Mass	M.S. Results[a]	Corrected for Cs^{137} Decay[b]	Absolute Yield[c]
133	1.118	1.072	6.3
135			[d]
137	1.000	1.000	5.9

[a] Average of four values, which showed no detectable contamination by natural cesium. See chemical procedures for details of blank.
[b] Extrapolated to zero time according to 33 yr. half-life.
[c] Obtained by normalizing to the known value for Xenon-133
[d] This value is somewhat low because of the high neutron capture cross-section of Xenon-135.

(b) Rubidium

The rubidium samples from fission were found to contain normal rubidium as an impurity. Correction for the normal isotopic impurity was made in the following manner. If the ratio of the fission yields of Rb^{85} and Rb^{87} is 1/a, the amount of rubidium-87 present as contamination is xm and the amount of rubidium-85 contamination is 2.6x, then

$$\frac{1 + 2.6x}{a + x} = \text{ratio found in normal sample} \qquad (1)$$

$$\frac{1 + 5.2x}{a + 2x} = \text{ratio found in blank sample} \qquad (2)$$

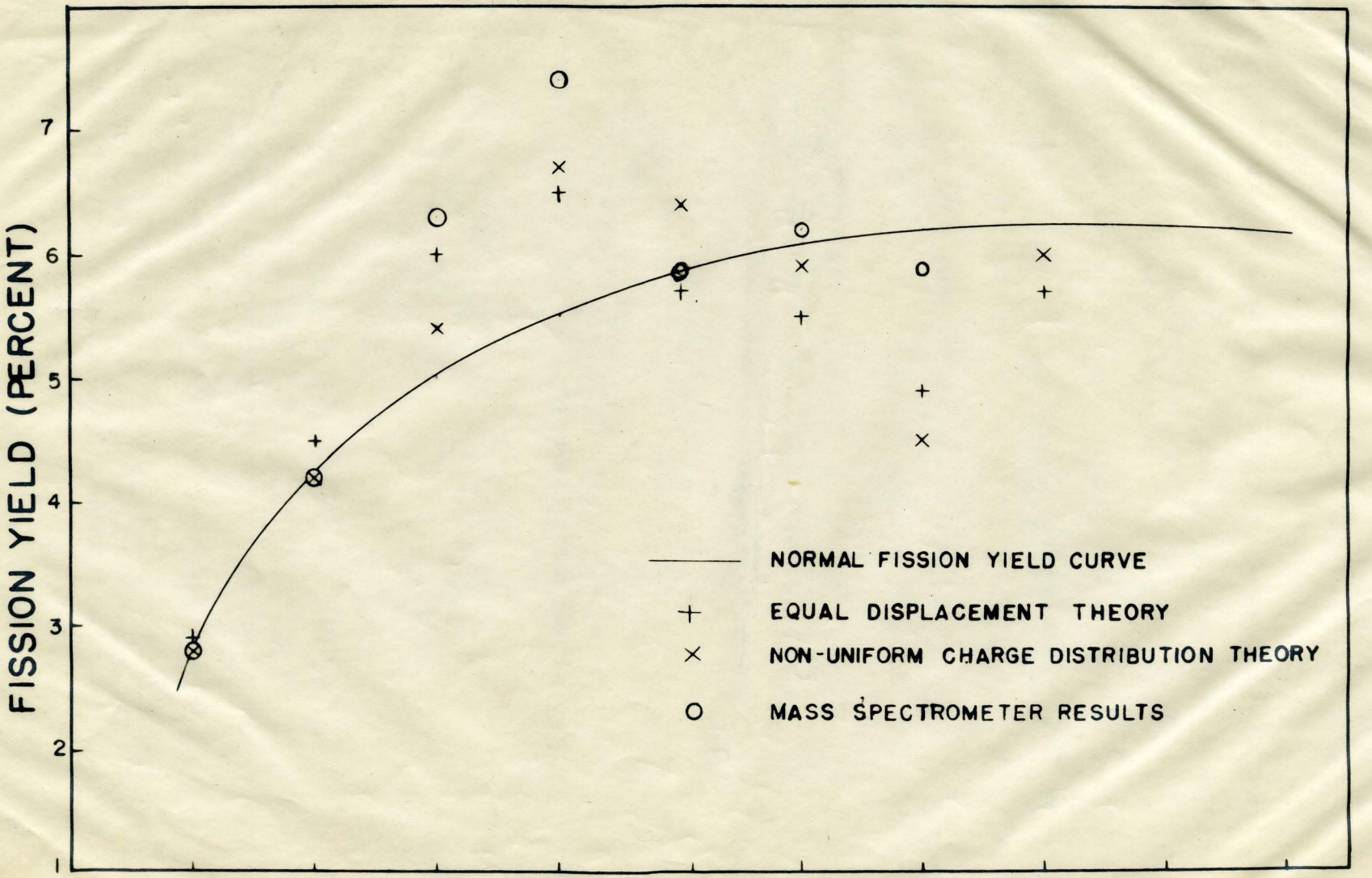

FIG. 7.

FISSION YIELDS IN THE HEAVY MASS RANGE

Elimination of x between (1) and (2) gives the net ratio of fission isotopes.

Thode[41] has reported that the 9.4 year isomer of krypton-85 accounts for 25% of the total yield of the mass 85 chain predicted from the smooth mass-yield curve. Thus, it is calculated that in the samples used in this work, 22% of the cumulative yield of rubidium-85 is still blocked by the krypton isotope. The mass spectrometer results were corrected for this loss according to the 9.4 year half-life of krypton-85. The samples used in this work were 2.1 years old.

A consideration of the probable neutron capture cross sections of rubidium-86 indicates that it is very unlikely that the reaction Rb^{86} (n,Y) Rb^{87} would contribute any appreciable error.

The final results for rubidium isotopes in fission are reported in Table IX.

TABLE IX

Yields of Rubidium Isotopes From Thermal Neutron Fission of Uranium-235

Mass	Sample UB-1b[a]	Sample UB-1a[a]	Net Value[b]	Corrected Value[c]	Absolute Yield[d]
85	0.506	0.442	0.374	0.475	1.3
87	1.000	1.000	1.000	1.000	2.74

[a]Sample UB-1b contained the blank, whereas Sample UB-1a did not. (See procedures for an explanation of the blank)
[b]See discussion above for the method of calculating net yields.
[c]This correction is for the decay of 9.4 year krypton-85
[d]Absolute yield values are assigned on the assumption that the value for mass 85 lies on the smooth curve.

(c) Strontium

It was not necessary to run blanks to determine the extent of normal strontium in the fission product material, since of the natural isotopes of strontium, only one, Sr^{88}, is produced in fission. Thus, an accurate measure of the contamination by natural strontium-88 could be calculated from the amount of strontium-86 present in the sample. The value obtained for 19.9 year strontium-90(42) had to be extrapolated to zero time to correct for decay. The final values are presented in Table X.

TABLE X

Relative Yields of Strontium Isotopes From Thermal Neutron Fission of Uranium-235

Mass	Yield				
	M.S. Values		Corrected For Sr^{90} Decay		Average
88	0.80[a]	0.65[b]	0.71	0.61	0.66
90	1.00	1.00	1.00	1.00	1.00

[a] This sample was 4.66 years old.

[b] This sample was 2.12 years old.

DISCUSSION

General

The experimental results provide further evidence of fine structure in fission yields. The mass 133 chain is found to have a high relative yield, in agreement with results reported previously for the isotopes of xenon. The yields of the 135 and 137 mass chains are somewhat lower, and when normalized at 6.3 percent for cesium-133, give values in fair agreement with the predictions of the smooth curve. With the exception of the 137 mass chain, there is qualitative agreement with the yield values predicted by the Glendenin mechanism. However, the Glendenin mechanism requires that yields greater than the smooth curve values must be counterbalanced exactly, in their totals, by yields in other chains which are below the smooth curve. It is obvious from Figure 7 that this is not the case for the mass range under study. The total of the yields determined experimentally is greater than the total given by the smooth curve as drawn in the Plutonium Project report.

Since the smooth mass yield curve was obtained by plotting all the known experimental data, some of which was subject to large errors, one might suggest a revision of the smooth curve upward in the 133-137 mass range to bring the yields predicted by Glendenin in closer agreement with experimental results. However, if this is done, one obtains a hump on the side of the curve. This suggests an effect other than that proposed by Glendenin to account for departure from the smooth yield curve.

TABLE XI

Comparison of Experimental Results with Various Predicted Yields

Mass	Yield Curve	From Glendenin's theory	From Present's theory	Experimental
131	2.8	2.9	2.8	2.8
132	4.2	4.5	4.2	4.2
133	5.0	6.0	5.4	6.3
134	5.5	6.5	6.7	7.4
135	5.9	5.7	6.4	5.9
136	6.1	5.5	5.9	6.1
137	6.2	4.9	4.5	5.9
138	6.2	5.7	6.0	(6.0)[a]
Total	41.9	41.7	41.9	44.6

[a]Average of the three predictions. The true value is not expected to be sufficiently different to change the total significantly.

Possible Explanation of Anomalous Results

The results obtained to date suggest that the discrepancy between the predicted and experimental total yields in the region from mass 131 to mass 138 is probably caused by some characteristic of the fission process itself, rather than by a post-fission effect. The results could be accounted for qualitatively, for example, if we were to assume that nuclei with eighty-two neutrons have a slightly higher primary yield than would be predicted from symmetrical charge distribution curves.

On the basis of this assumption alone, then, one would expect that the yield of tin-132, which has both eighty-two neutrons and fifty protons,

would be very high. This, however, is not the case. A consideration of the relative stabilities of nuclei in this region will explain this apparent anomaly. The neutron-proton ratio of tin-132 is 1.64, as compared to 1.41 predicted by the Bohr-Wheeler theory as the most stable ratio for mass 132. On the other hand, cesium-137, also with eighty-two neutrons, has a neutron-proton ratio of 1.49, as compared with 1.42 for the most stable ratio. Therefore, any particular tendency for tin-132 to form in fission would be opposed by the high degree of instability of such a high neutron-proton ratio. It seems, then, that the increased yield of eighty-two-neutron primary fission fragments would be a function of the stability of the nculeus. The effect would thus be appreciable only in masses 133, 134, 135 and 136. Figure 8 is a smooth curve obtained empirically which gives the variation of this effect with mass. The yield values obtained on the basis of this variations are shown in Table XII.

It will be seen from Table XII that considerable improvement is made by combining the two effects. This improvement is especially noticeable in the close agreement obtained between predicted and experimental total yields.

TABLE XII

Fission Yields Assuming an Eighty-Two Neutron Effect in Primary Fission

Mass	Smooth Curve	Increase		Corrected Yield	Corrected Yield Plus Glendenin Effect[a]
		% of Primary Yield of Nuclide	% of Total Fission		
131	2.8	0	0.0	2.8	2.9
132	4.2	2	0.0	4.2	4.5
133	5.0	14	0.3	5.3	6.3
134	5.5	32	0.9	6.4	7.4
135	5.9	50	0.8	6.7	6.5
136	6.1	60	0.5	6.6	6.0
137	6.2	65	0.1	6.3	5.0
138	6.2	-	0.0	6.2	5.7
Total	41.9			44.5	44.3

[a]In this and following calculations, the eighty-three neutron effect is based on Glendenin's theory of charge distribution. Analogous predictions could be made on the basis of Present's theory of charge distribution.

Large discrepancies still exist in certain chains, notably the mass 137 chain. In this chain, almost the whole predicted fine structure effect is due to the Glendenin mechanism, and it is seen that the experimental results do not show nearly as large an effect as is predicted. At the same time, large corrections due to the Glendenin effect are required to account for the very high yields obtained for masses 133 and 134. Mass yields in better agreement with experimental values can be obtained if it is assumed that the Glendenin effect, as well as the eighty-two neutron

effect, is a function of the neutron-proton ratio.

This variation of the Glendenin effect with neutron/proton ratio seems reasonable from a consideration of nuclear stability. For example, the most stable neutron/proton ratio for the mass 133 chain is 1.41. Thus, tin-133, with a ratio of 1.66, will be very unstable, and would be quite likely to emit a neutron if it were highly excited. On the other hand, cesium-138, whose neutron/proton ratio of 1.51, as compared to the most stable ratio 1.42 for mass 138, would be considerably more stable, and only under extreme excitation would neutron emission be favoured over beta disintegration. It would therefore be expected that the probability of instantaneous neutron emission would diminish as the mass of the nuclide with eighty-three neutrons increases from 130 to 140. An empirical curve showing this variation is given in Figure 9.

The yield values, calculated assuming both the modified Glendenin effect and the closed shell effect in primary fission, given in Table XIII, are the best of several sets calculated from various possible curves of neutron emission vs. mass number, and of eighty-two neutron effect vs. mass number. The curves used in calculating the tabulated values are those of Figures 8 and 9.

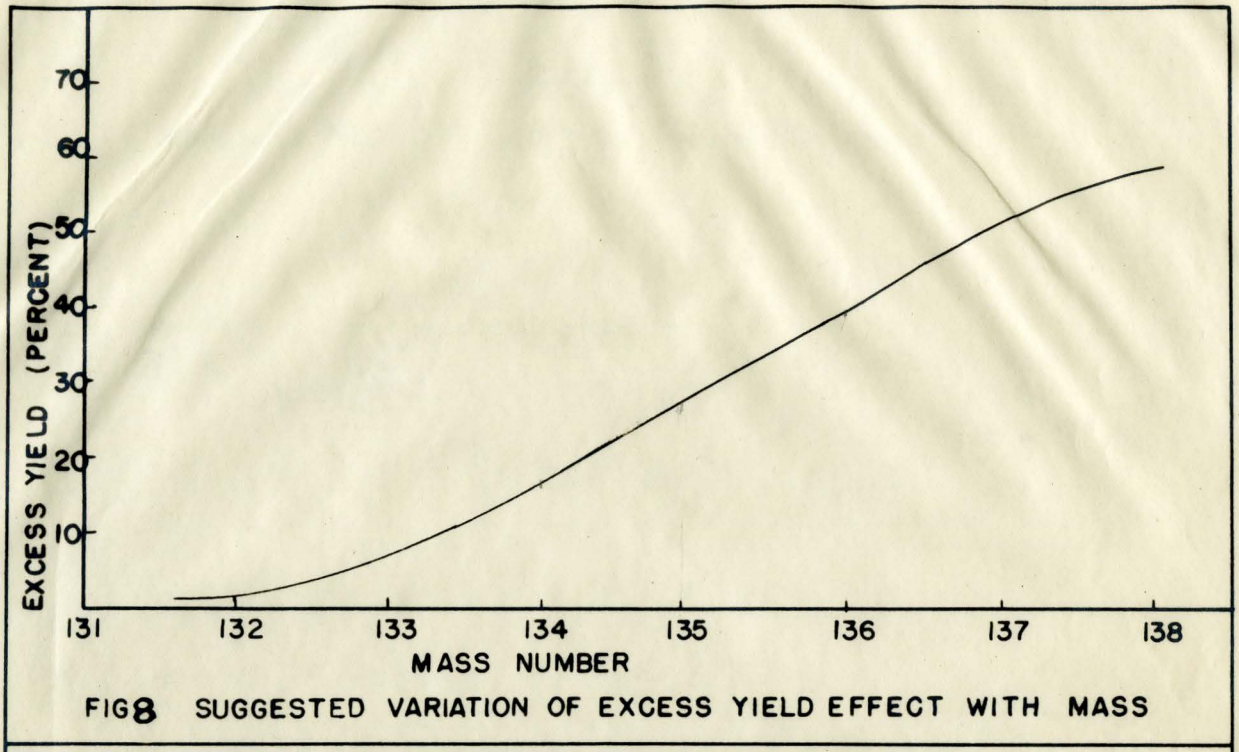

FIG 8 SUGGESTED VARIATION OF EXCESS YIELD EFFECT WITH MASS

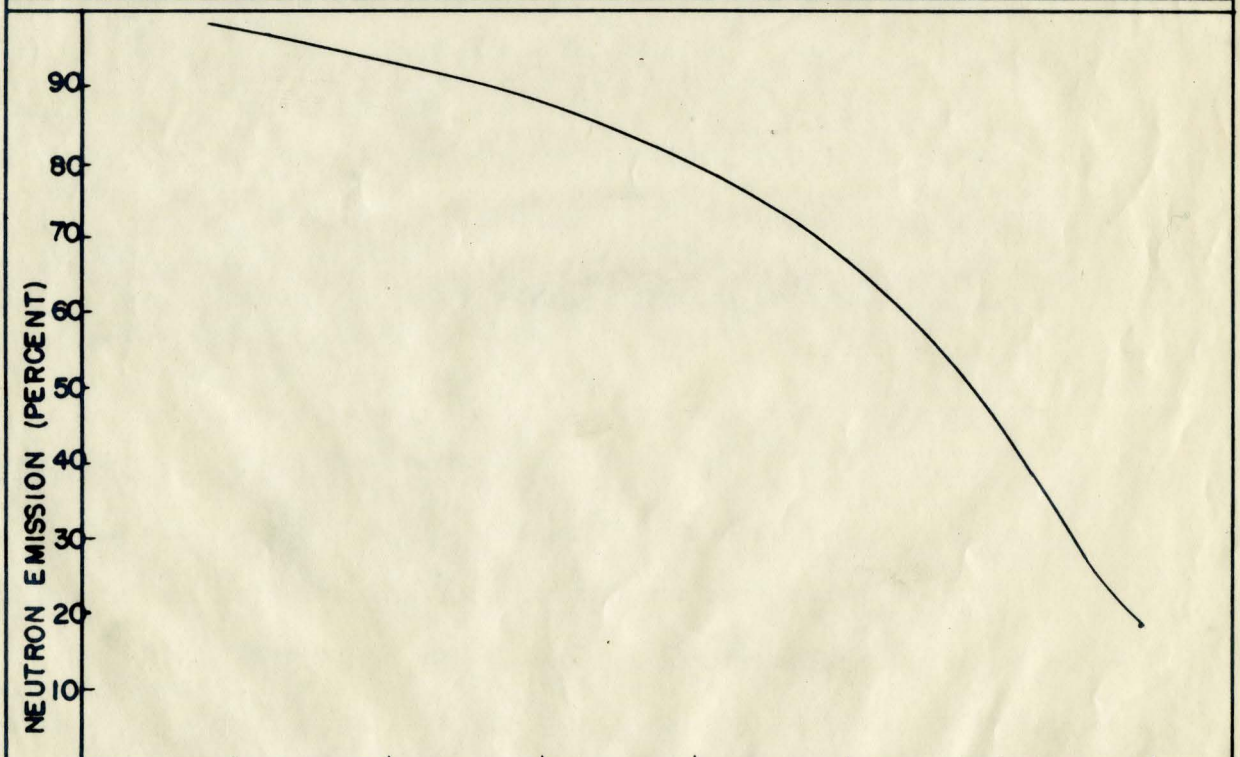

FIG 9 SUGGESTED VARIATION OF THE GLENDENNIN EFFECT WITH MASS

TABLE XIII

Predicted Fission Yields in the Heavy Mass Range

Assuming Closed Shell Effects

Mass Number	Yield Curve	Predicted by Glendenin		Postulated in this Thesis[a]	Predicted from Experimental
		From Equal Displacement Theory	From Present's Theory		
131	2.8	2.9	2.8	2.9	2.8
132	4.2	4.5	4.2	4.5	4.2
133	5.0	6.0	5.4	6.3	6.3
134	5.5	6.5	6.7	7.1	7.4
135	5.9	5.7	6.4	6.0	5.9
136	6.1	5.5	5.9	5.8	6.1
137	6.2	4.9	4.5	5.7	5.9
138	6.2	5.7	6.0	6.2	- -

[a]In these calculations the equal charge displacement was used in calculation of the Glendenin effect. Analogous results could be obtained using Present's non-uniform distribution theory.

Summary of Proposed Mechanism

In an attempt to explain anomalous results in the fission fine-structure effect, the following assumptions were made:

1. The distribution of both charge and mass fundamentally follow simple probability laws, and any fine structure is due to other effects superimposed upon the elementary process. These other factors may be either during or immediately following the fission process.

2. The Glendenin mechanism of neutron emission is assumed to be correct. However, the probability of neutron emission from a nucleus with eighty-three neutrons is assumed to be a function of the displacement of that nucleus from the Bohr-Wheeler stability line.

3. The probability of formation as primary fission products is greater for nuclei with eighty-two neutrons than for other nuclei. The magnitude of this effect will vary as some function of the displacement of those nuclei from the Bohr-Wheeler stability line.

In addition to the foregoing, two tacit assumptions have been made for the sake of simplicity. They are 1. the excess yields of eighty-two neutron fission fragments has no effect whatsoever on the probability of formation of other fission fragments, and that these others follow the smooth, symmetrical charge and mass distribution curves; 2. there is no significant effect due to the even or odd numbers of protons in the primary nucleus. Considerably more data must be accumulated before these assumptions can be tested.

One of the most important weaknesses of the proposed modification of the Glendenin mechanism is that the known neutron emission of xenon-137 seems to be a contradiction. It might be, however, that in this case neutron emission is favoured because in that way the product nucleus has even numbers of both protons and neutrons, whereas the nucleus resulting from a beta disintegration would have an odd number of protons, and a resultant high nuclear angular momentum.

The proposals made in this thesis might be tested by analysing the fission products from fission induced by neutrons of higher energies. At

higher energies, one would expect the preferential formation of eighty-two neutron fragments to decrease, and the fission process to become less selective. One would not expect the Glendenin mechanism, which occurs after the fission process, to be affected.

APPENDIX

Nuclear Shell Structure

The concept of shells and energy levels in the nucleus roughly analogous to those of electrons in atomic orbitals was first proposed by Bartlett[43] in 1932, and was soon taken up in the theoretical work of Elsasser[44]. Only recently, however, has it been given much serious consideration in the light of experimental evidence. Although there is considerable disagreement, even in those regions of the periodic table where evidence is abundant, and especially among heavier nuclei, for which it is difficult to make satisfactory calculations, much preliminary work has been done. The recent work of Mayer[23,45,46,47], Nordheim[48] and Feenberg[49,50,51] is of great importance in opening up this field.

Empirical evidence has rapidly been accumulating leading to observations that certain numbers of protons and/or neutrons in a nucleus seem to be more stable than the others. These numbers, the so called "magic numbers', are 2, 8, 20, 28, 50, 82 and 126. A certain stability is also noted in nuclei with 40 nuclides of one kind.

The evidence in support of these observations is summarized by Mayer[23]. Data is drawn from many types of observations, including the following:

1. Isotopic abundances. Of all the isotopes of atomic number greater than thirty-three, only three have abundances greater than sixty percent of that element. Of these, strontium-89 has 50 neutrons, and barium-138 and cerium-140 each have eighty-two neutrons.

2. The number of isotones*. Although the average number of

---

*Isotones are nuclides with the same number of neutrons.

isotones for even numbers of neutrons throughout the periodic table is between three and four, seven naturally occurring nuclides have eighty-two neutrons, and six have fifty neutrons.

3. The number of isotopes. Tin, with fifty protons, has more stable isotopes than any other element. The heaviest and lightest differ by twelve mass units. The only other case of such a spread in the masses of the isotopes of an element is found in xenon, whose great span may be attributed to xenon-136, which has eighty-two neutrons.

4. Neutron capture cross section. Capture cross sections for nuclides with fifty, eighty-two, or one hundred and twenty-six seem abnormally low. At the same time, some of those with forty-nine or eighty-one neutrons seem unusually high.

This and other evidence leaves no doubt as to the particular stability of certain shells of both protons and neutrons. The interpretation of this stability and the assignment of spectroscopic terms to the levels is a very difficult task.

The early history of atomic structure is to be found in the annals of spectroscopy. It was from the energies of the spectral lines that the energies of the electron levels were calculated. Thus, it might be expected that a similar case might be that of nuclear levels and nuclear spectroscopy. This, however, is not the case as much as would be expected.

One of the reasons for this is the lack of a sufficient number of simple nuclei which give out radiations. The most common artificially and naturally radioactive nuclides are very complex in their structure. It may be noted here in comparison that the optical spectra of some of the more

complex elements are not yet completely resolved. Another great difficulty which must be met is that the interactions between and relations between the various nuclear particles are very strong and not at all well understood.

Three methods have been used, individually, or inconjunction with one another, for the assignment of terms to the various levels within the nucleus:

1. Wave mechanical calculations, beginning with Elsasser's work, have been set up for variously shaped potential wells, in which the nuclear particles are supposed to move. Nordheim utilizes the postulate of an approximately spherical well. Mayer and Feenberg use a square well, with possibly rounded corners. The 'wine bottle' potential well, as first suggested by Elsasser, is also used by Feenberg in the calculation of the energies of proton oscillators.

2. The analysis of the spins of nuclei, as determined from hyperfine structure in optical spectroscopy, and from angular correlation work in gamma-ray spectroscopy, gives considerable information as to the angular momentum of the nucleus. The simplifying assumption is made that an even number of identical nucleons are all paired to give zero angular momentum to the nucleus. Thus, the momentum of an odd nucleus may all be attributed to the odd proton or neutron or to the odd 'hole'. This assumption is characteristic of the 'individual particle model'. The spin and angular momentum contribution of the nucleons are calculated in a manner similar to that used in optical spectroscopy. The neutron and proton each have angular momenta -l- equal to h/2, while their spins -2- are each equal to $\frac{1}{2}$ h/2.

Thus, the spectroscopic terms of individual nucleons are analogous to those of individual electrons.

3. Nordheim uses magnetic moments of nuclei to determine whether the spin of a nucleon is parallel to or antiparallel to the orbital angular momentum. In all this work a strong spin-orbit coupling is assumed, so that the relative orientation of the two forms of momentum is of highest importance. Nordheim plots the magnetic moments of nuclei against the spectroscopically determined spin, I. The points are found to lie on or near two rather poorly defined lines. These lines are close to the theoretically determined lines for $I_j = l + \frac{1}{2}$ and $I_j = l - \frac{1}{2}$, as calculated by Schmidt. The fact that these points do not form a well defined pair of lines is attributed to the insufficiency of the one particle picture. There is thus some interaction of odd particles with those already paired.

Perhaps the most convincing paper to date is that of Mayer[47] in which the theory of the 'spin-orbit coupling model' is put forward. The assumptions made in that theory are:

1. The succession of energies of single particle orbits is that of a square well with strong spin-orbit coupling giving rise to inverted doublets.

1a. For a given value of I, the level $I = l + \frac{1}{2}$ has invariably lower energy than, and will be filled before that level where $I = l - \frac{1}{2}$.

1b. Pairs of spin levels within one shell, which arise from adjacent orbitals in the square well in such a way that the spin-orbit coupling tends to bring their energy closer, can and often will cross.

2. An even number of identical nucleons in any orbit with total angular momentum quantum number j will always couple to give a spin of zero,

and no contribution to the magnetic moment.

3. An odd number of identical nucleons in a state j will couple to give a total spin of j, and a magnetic moment equal to that for a single particle in that state.

4. For a given nucleus, the pairing energy of the nucleons in the same orbit is greater for orbits with larger j.

A good example of crossing of levels because of pairing energy of the level with higher angular momentum is afforded by the spins of odd-proton nuclei with Z lying between 28 and 38. From Table XIV, it is seen that the proton configuration for Z = 28 is $1s^2$, $2p^6$, $3d^{10}$, $2s^2$, $4f_{7/2}{}^8$. The next proton should be, then, according to the calculated energy of the levels, an $f_{5/2}$ proton. However, spins for the next five odd-proton nuclei indicate filling in the $P_{3/2}$ level. The only exceptions to this are $_{37}Rb^{85}_{48}$ and $_{30}Zn^{67}_{37}$, which have spins of 5/2. This shows that the proton or neutron goes into the $3p_{3/2}$ level, but that the next proton causes a re-arrangement, and both protons go into the lower-lying $4f_{5/2}$ level. The closeness of such levels during a cross-over is shown by the frequency of cases of nuclear isomerism in those regions.

Neutron structure of nuclei with fewer than fifty neutrons is identical to that for the same number of protons. After fifty, however, the coulomb repulsion has a considerable effect on the density of protons close to the center, whereas the neutrons are unaffected. Thus, proton levels with high angular momentum are favoured more than in neutron levels.

The present state of nuclear energy assignments may be summarized for the three important schools of investigation as in Table XIV.

TABLE XIV

Energy Levels and Spin Terms of Nuclei

(After Mayer)(44) 51

ν	2 6	12	30	32
$\Sigma\nu$	8	20	50	82
Feenberg and Hammack	$1s^2\ 2p^6$	$2s^2\ 3d^{10}$ - $3d^{10}$	$4f^{14}$ $5g^{18}$	$6h^{22}$ $4d^{10}$
Nordheim	$1s^2\ 2p^6$	$2s^2\ 3d^{10}$	$4f^{14}\ 3p^6\ 4d^{10}$	$5g^{18}$ $5f^{14}$
Mayer	$1s^2\ 2p^6$	$2s^2\ 3d^{10}$	$4f^{14}\ 3p^6\ 5g^{10}_{9/2}$	$5g^{8}_{7/2}\ 4d^{10}\ 3s^2\ 6h^{12}_{11/2}$
Square Well	1s 2p	3d 2s	4f 3p 5g 4d	3s 6h 5f 4p 7i

TABLE VIII

Yields of Cesium Isotope From Thermal Neutron Fission of Uranium-235

Mass	Absolute Yield
133	6.29[a]
135	6.18[b]
137	5.87[c]

[a] Value for Xe^{133} based on 2.8% yield of I^{131}

[b] Corrected for neutron absorption

[c] Corrected for decay using 33-year half-life.

LIST OF ILLUSTRATIONS

REFERENCES

1. H. G. Thode, and R. L. Graham, Can. J. Res., A27, 1, (1947).

2. J. Macnamara, C. B. Collins, and H. G. Thode, Phys. Rev. 78, 129, (1950).

3. M. G. Inghram, D. C. Hess, and J. H. Reynolds, Phys. Rev. 76, 1717, (1949).

4. L. E. Glendenin, Ph.D. Thesis, Massachusetts Institute of Technology, June, 1949.

5. E. W. Titterton, and F. K. Goward, Phys. Rev. 76, 142, (1949).

6. O. Hahn and F. Strassmann, Naturwiss. 27, 11, (1939).

7. E. P. Steinberg, J. A. Seiler, A. Goldstein, and A. Dudley, United States Atomic Energy Commission, MDDC 1632.

8. J. C. Jacobsen, and N. O. Lassen, Kgl. Danske Videnskab. Selskab. Matt-fys. Medd. 19, No. 6 (1941).

9. D. H. T. Gant, and R. S. Krishmann, Proc. Roy. Soc., (London) A178, 478, (1941).

10. E. Fermi and E. Segre, Phys. Rev. 59, 680 (1941).

11. R. O. Haxby, W. E. Shoupp, W. E. Stephens, and W. H. Wells, Phys. Rev. 59, 57 (1941).

12. M. Petrzhak, and G. N. Flerov, J. Exptl. Theoret. Phys. (U.S.S.R.) 10, 1013, (1940).

13. H. G. Thode and J. Macnamara, N.R.C. Progress Report, August 22, (1950).

14. W. Jentschke, F. Prankl, and F. Hernegger, Naturwiss. 28, 315 (1940).

15. I. Joliot-Curie, and F. Joliot, Ann. Phys. 19, 107, (1944).

16. G. Dessauer, and E. M. Haffner, Phys. Rev. 59, 840 (1941).

17. J. C. Jacobsen, and N. O. Lassen, Phys. Rev. 58, 867, (1940).

18. Amos S. Newton, Phys. Rev. 75, 17 (1949).

19. I. S. Panasyuk and G. N. Flerov, Compt. Rend. Acad. Sci. URSS 30, 704 (1941).

20. I. Perlman, R. H. Goeckermann, D. H. Templeton and J. J. Howland, Phys. Rev. 72, 352 (1947).

References (continued)

21. R. H. Goeckermann and I. Perlman, Phys. Rev. 73, 1127, (1948).

22. R. H. Goeckermann and I. Perlman, Phys. Rev. 76, 628 (1949).

23. Maria G. Mayer, Phys. Rev. 74, 235 (1948).

24. Lise Meitner, Nature 165, 561 (1950).

25. N. Bohr and J. A. Wheeler, Phys. Rev. 56, 426 (1939).

26. L. E. Glendenin, C. D. Coryell and R. R. Edwards, Plutonium Project Record, Vol. 9, Paper 52, (1946).

27. R. D. Present, Phys. Rev. 72, 7 (1947).

28. Plutonium Project, J. Am. Chem. Soc. 68, 2411, (1946).

29. W. E. Grummitt and G. Wilkinson, Nature 158, 163 (1946)

30. K. H. Kingdon, Phys. Rev. 76, 136 (1949).

31. C. W. Stanley, and S. Katcoff, J. Chem. Phys. 17, 653 (1949).

32. J. D. Kurbatov, M. L. Pool and H. B. Law, Phys. Rev. 59, 919 (1939).

33. Iw. Kurbatow, J. Phys. Chem. 36, 1241, (1932).

34. A. J. Dempster, Proc. Amer. Phil. Soc. 75, 755 (1935).

35. A. J. Dempster, Phys. Rev. 51, 67 (1937).

36. R. J. Horsley, M.Sc. Thesis, McMaster University, May 1950.

37. A. E. Shaw, United States Atomic Energy Commission, MDDC 30A (1946).

38. R. L. Graham, A. L. Harkness and H. G. Thode, J. Sci. Instr. 24, 119, (1947).

39. B. W. Smith, Private Communication.

40. L. E. Glendenin and R. P. Metcalfe, Plutonium Project Report CC-2219, (Feb. 1945).

41. H. G. Thode, and R. B. Shields, Reports on Progess in Physics, XII, 1, (1949).

42. R. I. Powers and A. F. Voigt, Phys. Rev. 79, 175 (1950).

43. J. R. Bartlett, Nature 130, 165 (1932).

References (continued)

44. W. Elsasser, J. de Phys. et Rad. 5, 625 (1934).

45. Maria G. Mayer, Phys. Rev. 75, 1969 (1949).

46. Maria G. Mayer, Phys. Rev. 78, 16 (1950).

47. Maria G. Mayer, Phys. Rev. 78, 22 (1950).

48. L. W. Nordheim, Phys. Rev. 75, 1894 (1949).

49. E. Feenberg, Phys. Rev. 75, 320 (1949).

50. E. Feenberg and K. C. Hammack, Phys. Rev. 75, 1877 (1949).

51. E. Feenberg, K. C. Hammack and L. W. Nordheim, Phys. Rev. 75, 1968 (1949).