particle model, while Goldhaber and Teller's theory does not consider this question. We believe that many of the features of the nuclear photo-effect can be understood merely from the fact that there are dipole transitions, without a special model.

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The Fission Yield of Xe^{133} and Fine Structure in the Mass Yield Curve

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A mass spectrometer investigation of the xenon isotopes formed from U^{235} fission has been made and the fission yield and half-life of Xe^{133} determined. The fission gases were extracted, purified, and the mass spectrometer abundance data obtained within two weeks of the end of the irradiation period. The fission yield of Xe^{133} was found to be 6.29 ± 0.01 percent, which is 20 percent higher than that expected from the mass yield curve. This is further evidence of fine structure in the mass fission yield curve. The half-life value of Xe^{133} was found to be 5.270 ± 0.002 days.

THODE and Graham[1] first determined mass spectrometrically the relative abundances of stable isotopes of Xe and Kr resulting from the decay of fission product chains in U^{235} fission. In this way the relative fission yields of eight mass chains—83, 84, 85, 86, 131, 132, 134 and 136, were determined with considerable accuracy. When these yields are normalized to the mass-yield curve at a value of 2.8 percent for mass 131, the values fit the curve nicely with the exception of Xe^{134} which is about 35 percent above the normal fission yield curve. Glendenin[2] has pointed out that the value obtained by Thode and Graham[1] of Kr^{84} is also above the experimental mass yield curve by about 35 percent. These results indicated for the first time "fine structures" in the mass yield curve. Recently, Inghram, Hess, and Reynolds[3] reported isotope abundance data for fission product cesium, which also indicate anomalies in the mass yield curve.

If it is assumed that the yields should fall on a smooth curve, then the abnormal yields of Xe^{134} and Kr^{84} might be explained by delayed or prompt neutron emission. If this fission chain branching does occur, then the fission yield of adjacent chains will be affected. It seemed important, therefore, to determine accurately the fission yields of the 133 and 135 mass chains. In the original mass spectrometer investigations of the fission gases, Xe^{133} and Xe^{135} did not occur because of their short half-lives, and because it was not possible to get samples immediately after irradiation. However, with the Chalk River facilities of the National Research Council available, it has now been possible to extract Xe gas from irradiated uranium disks without a long "cooling" period and thus permit the investigation of 5.3-day Xe^{133}. In order to calculate the fission yield of Xe^{133} from abundance data, (after a definite irradiation and cooling time), it is necessary to know its half-life with considerable accuracy. Since the previous values obtained by radio-chemical methods,[4–12] were only good to 1 or 2 percent, a new and more accurate value was determined mass spectrometrically. This determination of the half-life of Xe^{133} and the determination of the fission yield for the 133 mass chain are reported in this paper.

THEORY

Half-Life Determination

By comparing the abundance of a radio-active isotope with that of a stable one with a mass spectrometer over a period of time, it is possible to follow its decay rate and thereby determine its half-life. By this method, very accurate half-life determinations are possible for isotopes with half-lives ranging from about one day to ten years. The fundamental decay equation is

$$n/n_0 = e^{-\lambda t} = \exp(-0.6932t/t_{\frac{1}{2}}),$$

where n is the concentration at time t and n_0 is the concentration at zero time.

By substituting for example the 133/131+132 ratio obtained with the mass spectrometer at different time

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[2] G. L. Glendenin, Ph.D. thesis, Department of Chemistry, M.I.T. (August 1949).
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intervals for n/n_0 the half-life, $t_{\frac{1}{2}}$, of Xe^{133} can be determined.

Fission Yield Determination

The fission yield of the 133-mass chain for U^{235} fission is calculated from the mass spectrometer abundance ratio of Xe^{133} to Xe^{134} obtained after an irradiation time t, a "cooling" time to t_1 and at a time t_2 after extraction of the fission gases.

The two mass chains being compared are as follows:

$$Sb^{133} \xrightarrow{10\text{ min.}} Te^{133} \xrightarrow{60\text{ min.}} I^{133} \xrightarrow{22\text{ hr.}}$$
$$Xe^{133} \xrightarrow{5.270\text{ day}} Cs^{13}$$

$$Sb^{134} \xrightarrow{10\text{ min.}} Te^{134} \xrightarrow{43\text{ min.}} I^{134} \xrightarrow{54\text{ min.}} Xe^{134}\text{ stable}$$

For purposes of calculating the yield of the 133-mass chain, the following assumptions can be made when cooling times of 5 to 10 days are considered:

1. That the primary fission yield of Xe^{133} is negligible (from point of view of calculations) and that Xe^{133} produced comes from the decay of I^{133}.
2. That the fission yield of I^{133} can be taken as the yield of the whole chain in view of the short half-lives of its precursors.
3. That the 134 chain will have completely decayed to Xe^{134} at the time of extraction of the fission gases.
4. That the fission yield of the 134-mass chain is 7.4 percent as taken from previous mass spectrometer work.

With these assumptions a formula can be derived for the yield of the 133-mass chain in terms of the mass spectrometer ratio of Xe^{133} to Xe^{134}.

The number of atoms of I^{133} of fission yield Y_{133} and disintegration constant λ_1 produced at constant neutron flux f for a period of irradiation time t is:

$$n=\frac{fY_{I^{133}}}{\lambda_1}[1-\exp(-\lambda_1 t)]. \quad (1)$$

Now since we have assumed that all the Xe^{133} produced comes from the decay of I^{133}, the number of atoms of Xe^{133} of disintegration constant λ_2 produced during the irradiation period t will be:

$$n_1=\frac{fY_{I^{133}}}{\lambda_2}\left[1-\frac{\lambda_1\exp(\lambda_2 t)-\lambda_2\exp(-\lambda_1 t)}{\lambda_1-\lambda_2}\right]. \quad (2)$$

During the "cooling" period t_1, the time elapsed from the end of the irradiation until the xenon is extracted from the uranium metal, the number of Xe^{133} atoms produced will be:

$$n_2=\frac{n\lambda_1}{\lambda_2-\lambda_1}[\exp(-\lambda_1 t_1)-\exp(-\lambda_2 t_1)]. \quad (3)$$

The total number of atoms of Xe^{133} available at the time of dissolution is given by the sum of Eq. (2) multiplied by the decay factor $\exp(-\lambda_2 t_1)$ plus Eq. (3) with n taken from Eq. (1).

Therefore, the total number of atoms of Xe^{133} at the time of analysis is given by

$$N=[n_1\exp(-\lambda_2 t_1)+n_2]\exp(-\lambda_2 t_2), \quad (4)$$

where t_2 is the time elapsed from the time of dissolution of the uranium until the time of analysis with the mass spectrometer.

On substituting the expressions for n_1 and n_2 in Eq. (4) the total number of atoms of Xe^{133} at the time of analysis is then given by:

$$N_{Xe^{133}}=\exp(-\lambda_2 t_2)Y_{I^{133}}\left\{\frac{1-\exp(-\lambda_1 t)}{\lambda_2-\lambda_1}\times[\exp(-\lambda_1 t_1)-\exp(-\lambda_2 t_1)]+\frac{\exp(-\lambda_2 t_1)}{\lambda_2}\times\left[1-\frac{\lambda_2\exp(-\lambda_1 t)-\lambda_1\exp(-\lambda_2 t)}{\lambda_2-\lambda_1}\right]\right\}. \quad (5)$$

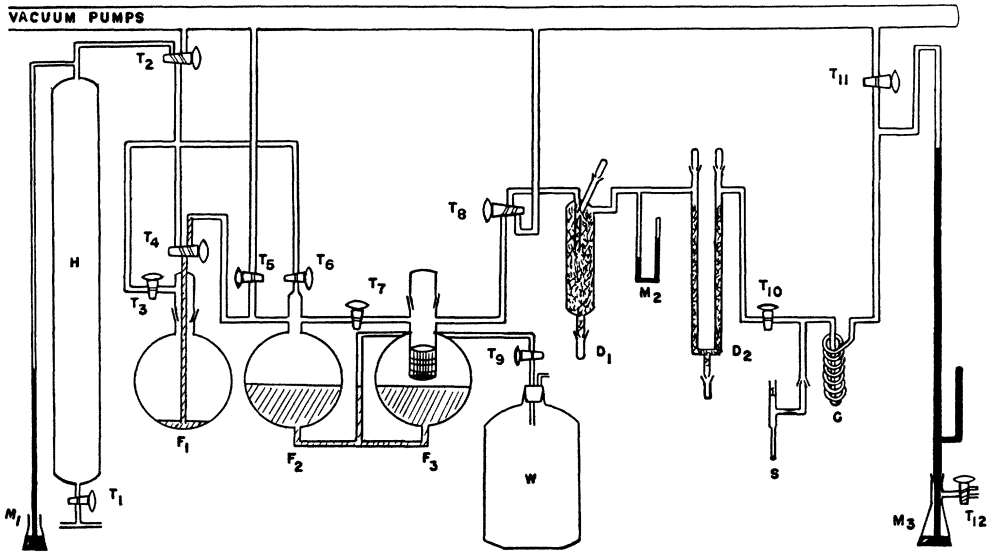

FIG. 1. Apparatus for extraction of fission product xenon and krypton from irradiated uranium.

TABLE I. Xe^{133} half-life from mass spectrometer abundance data.

Ratio of Xe mass ion currents ×100	Time after first analysis (hr.)			Xe^{133} half-life calculated (days)
	0	45.83	92.91	
133(131+132)	34.60±0.005	26.91±0.006	20.79±0.015	5.265
				5.275
				5.270
133/(131+132+134+136)		7.022±0.01	5.430±0.02	5.275
			Average	5.270±0.002

As the period of "cooling" is long compared to the half-lives of the Xe^{134} precursors, the total 134-mass chain will decay to Xe^{134}. Therefore, the total number of Xe^{134} atoms at the time of analysis is given by:

$$N_{Xe^{134}}=fY_{134}t, \tag{6}$$

where Y_{134} is the yield of the 134-mass chain. The ratio of the total number of Xe^{133} atoms to total number of Xe^{134} atoms at the time of analysis is therefore given by the equation:

$$\frac{N_{Xe^{133}}\text{ at }t_2}{N_{Xe^{134}}}=\frac{\exp(-\lambda_2 t_2)}{YXe^{134}t}Y_{133}\left\{\frac{1-\exp(-\lambda_1 t)}{\lambda_2-\lambda_1}\right.$$

$$\times[\exp(-\lambda_1 t_1)-\exp(-\lambda_2 t_1)]+\frac{\exp(-\lambda_2 t_1)}{\lambda_2}$$

$$\left.\times\left[1-\frac{\lambda_2\exp(-\lambda_1 t)-\lambda_1\exp(-\lambda_2 t)}{\lambda_2-\lambda_1}\right]\right\}. \tag{7}$$

The left-hand side of this equation is equal to the ratio of Xe^{133} to Xe^{134} at the time of analysis and is therefore given by the mass spectrometer ratio. Equation (7) was used to calculate the yield of the 133-mass chain.

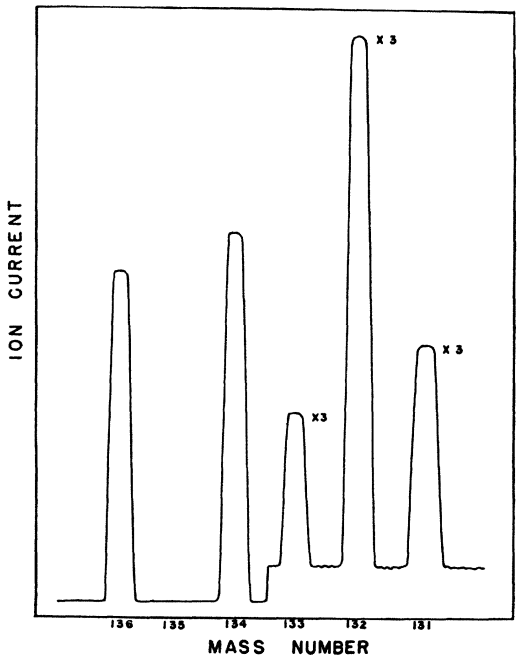

FIG. 2. Stable isotopes of fission product xenon, also Xe^{133}.

TABLE II. Fission yield of Xe^{133}. Yield of Xe^{134} taken as 7.4 percent. Thode and Graham results normalized to mass yield curve at 2.8 percent for Xe^{131}.

Sample No.	Time of irradiation hr. (t)	Time of cooling up to dissolution hr. (t_1)	Time after extraction before analysis hr. (t_2)	$\frac{Xe^{133}}{Xe^{134}}$	Yield of Xe^{133}
1	32.42	244.0	73.42	0.1644	6.285
2	25.88	211.90	51.42	0.2258	6.290
				Average	6.290±0.01
3*	68.45	193.67	50.83	0.2335	6.55

* Preliminary experiment, this result is probably high because of interruptions in the irradiation period.

EXPERIMENTAL

Preparation of Samples

Small uranium disks of about 20 g each, were irradiated with neutrons at as nearly a constant flux as possible in the pile at Chalk River. The uranium was irradiated for a sufficient period so as to produce approximately 10^{-3} cc at N.T.P. of fission product xenon and krypton. After "cooling" for ten days the uranium disks were dissolved in a saturated solution of cupric potassium chloride and the rare gases extracted by a method similar to that used by Arrol, Chackett, and Epstein.[13]

The extraction apparatus described by Arrol, Chackett, and Epstein required certain modifications to dissolve the more active disks (see Fig. 1). The apparatus was shielded with four inches of lead and stopcocks T_5, T_6 and T_7 were manipulated by remote control methods. About 800 cc of cupric potassium chloride solution in F_1 was flushed with dry hydrogen from the reservoir H to eliminate the air dissolved in the solution. The hydrogen was dried and purified by passing it through activated charcoal in a trap at liquid nitrogen temperatures. The uranium disk was placed in a platinum basket in F_3, and the reaction vessels F_2 and F_3 evacuated through T_5. The dissolving solution was then siphoned from F_1 into the reaction vessels through T_4. The rate of dissolution of the uranium disk was controlled by raising and lowering the solution in F_2 and F_3. The solution was raised and lowered by varying the pressure in F_2. Dry hydrogen was used when it was necessary to increase the pressure above the solution in F_2. The rate of dissolution was observed by the pressure readings on the manometers M_2 and M_3. M_3 was maintained at approximately 3 cm during the dissolution. The drying traps, D_1 and D_2 to eliminate HCl (gas) and water vapor, contained potassium hydroxide pellets and magnesium perchlorate, respectively. These traps were fitted with ground glass joints in order to change the drying agents during successive

[13] Arrol, Chackett, and Epstein, Can. J. Research, 27, 757 (1949).

TABLE III. Fission yields from mass spectrometer determinations and yield-curve data compared mass chains (131–138).

Mass chain	Mass yield curve	Mass spectrometer fission yield
131	2.8	2.8
132	4.2	4.2
133	5.0	6.3
134	5.5	7.4
135	5.9	—
136	6.1	6.1
137	6.2	—
138	6.2	—

dissolutions. This prevented further glass blowing once the line was contaminated. The fission gases and impurities were swept through the drying traps by the hydrogen gas produced during the dissolution of the uranium metal and absorbed on the charcoal in trap C at liquid nitrogen temperatures. Following the dissolution of the uranium, the remaining traces of Xe and Kr were flushed out with dry hydrogen from H and the system evacuated through T_{11}. The gas sample was then purified in a calcium furnace,[13] and the abundance of the xenon isotopes measured with the mass spectrometer.

The fission product solution was transferred automatically by evacuating the glass jar W and siphoning the solution from F_2 and F_3 into W.

Mass Spectrometer

The mass spectrometer used was a 180 degree direction focusing instrument.[14] The ion current measurements were made with a linear inverse feedback amplifier coupled to a Leeds and Northrup Speedomax recorder.[15] The relative abundance data reported in this paper are good to 0.1 percent or better.

RESULTS

Figure 2 shows a typical mass spectrogram for fission product xenon. Previous spectrograms[1] show no Xe^{133} because the uranium disks were allowed to cool for a number of months after irradiation to permit the various fission product chains to decay to the stable Xe^{131}, Xe^{132}, Xe^{134} and Xe^{136} isotopes. However, by analyzing the xenon produced in fission, within two weeks of the end of the irradiation period, it was possible to measure both the yield and the half-life of Xe^{133}.

Half-Life of Xe^{133}

The average value of the half-life of Xe^{133} from four determinations is 5.270 ± 0.002 days. This mass spectrometer method of determining half-lives, completely eliminates the difficulties from contamination with other radioactive isotopes which interfere with counting methods. It provides a most accurate means for determining half-lives in the range of several days to several years. Whereas the half-live values obtained by radiochemical methods are good to only 1 or 2 percent, the values obtained from mass spectrometer abundance data are good to 0.1 percent. Table I gives the results of the mass spectrometer analyses.

Fission Yield of Xe^{133}

The mass spectrometer abundance data for Xe^{133} and Xe^{134} was taken from spectrograms similar to Fig. 2. The results for three separate experiments are given in Table II. In columns 2, 3, and 4 are given the irradiation time of the uranium sample t, the cooling time t_1, and the analysis time after extraction of xenon gas t_2. Column 5 gives the abundance ratio Xe^{133}/Xe^{134} obtained with a precision of 0.1 percent and a probable accuracy of 0.5 percent and finally, column 6 gives the fission yield of the 133-mass chain, calculated by means of Eq. (7) where the half-lives of I^{133} and Xe^{133} were taken as 22 hours and 5.270 days respectively. It can be seen that the results for the two experiments check well within the limits of our precision. The results indicate that the fission yield for the 133 mass chain is also abnormal (see Fig. 3).

DISCUSSION

Table III summarizes the fission yield data which has been determined with considerable accuracy by mass spectrometer methods. These yields which include the yield of Xe^{133} reported in this paper are compared with

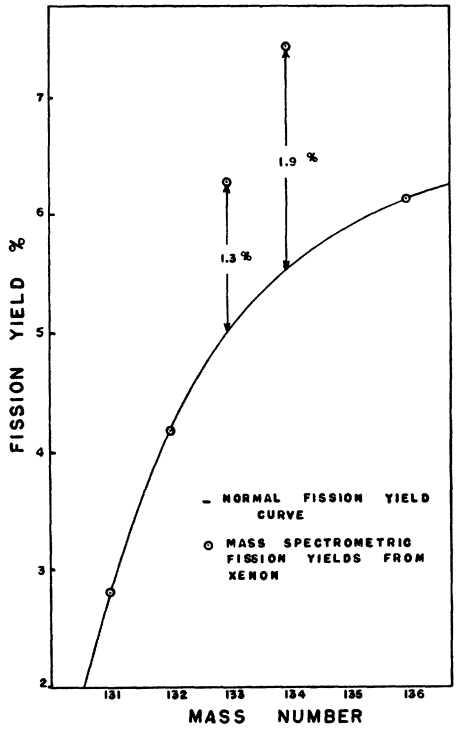

FIG. 3. Mass fission yield curve showing fine structure.

[14] Thode, Graham, and Zeigler, Can. J. Research **B23**, 40 (1945).
[15] Lossing, Shields, and Thode, Can. J. Research **B25**, 397 (1947).

corresponding values taken from the smooth mass yield curve.

It is seen that the mass spectrometric values fit nicely on the mass yield curve with the exception of Xe^{133}, Xe^{134}, and possibly Kr^{84} which are 20, 35, and 35 percent too high respectively (see Fig. 3).

Recently, C. W. Stanley and S. Katcoff[16] published a most interesting paper on the properties of 86-second I^{136}. According to their results the accumulative yield of I^{136} from U^{235} fission is 3.1 percent or about one-half the value predicted by the mass yield curve for the 136 chain. Furthermore, the yield of Xe^{136} determined mass spectrometrically is 6.1 percent. Stanley and Katcoff suggest several possible explanations for the low I^{136} yield. First, an independent isomer of I^{136} with a half-life less than 30 seconds would not have been observed in their experiments. Second, the daughter produced namely Xe^{136} might have a high primary fission yield, thereby accounting for the missing fraction of the total chain yield. This latter explanation seems very unlikely in view of the low yield found for Cs^{136} and in view of the low primary yields predicted for Xe^{136} by the theories of charge distribution. The known delayed neutron emissions from I^{137} through Xe^{137} must, of course, be taken into account.

It recently occurred to G. L. Glendenin[17] that the anomalous yields might be explained at least qualitatively on the basis of the extra stability for the nuclear closed shells of 50 and 82 neutrons which occur in fission products in the regions of mass numbers 85 and 135 (krypton and xenon ranges). There is very convincing evidence that special numbers of neutrons or protons in the nucleus form a particular stable configuration. M. G. Mayer[18] has recently summarized the experimental facts which indicate a particular stability of shells of 50, 82, and 126 neutrons.

Glendenin postulates that a primary fission product (which has already emitted the usual number of prompt neutrons) containing one neutron in excess of the closed shells (51 and 83) will often emit this more loosely bound neutron immediately rather than emit β-particles or γ-rays as in the ordinary case. This process could then account for abnormal fission yields in the neighborhood of nucleids with 50 and 82 neutrons.

Table IV illustrates the chain branching in the region of 82 neutrons as proposed by Glendenin. This scheme would explain qualitatively the abnormal yields of Xe^{134}, I^{136} and the abnormal yield of Xe^{133} reported in this paper. For example, the yield of the 134 chain is increased by the reaction

$$Te^{135}(83 \text{ neutrons}) \longrightarrow Te^{134}(82 \text{ neutrons}) + n \quad (3)$$

and is decreased by the reaction

$$Sb^{134}(83 \text{ neutrons}) \longrightarrow Sb^{133}(82 \text{ neutrons}) + n. \quad (2)$$

If Te^{135} has a higher primary yield than Sb^{134} then the 134 chain will gain more than it loses by this mechanism and its yield will be high.

TABLE IV. Branching.

Sn^{131}	Sb^{131}	Te^{131}	I^{131}	Xe^{131} Stable		
Sn^{132}	Sb^{132}	Te^{132}	I^{132}	Xe^{132} Stable		
$1\uparrow n$ Sn^{133}	Sb^{133}	Te^{133}	I^{133}	Xe^{133}	Cs^{133} Stable	
	$2\uparrow n$ Sb^{134}	Te^{134}	I^{134}	Xe^{134} Stable		
		$3\uparrow n$ Te^{135}	I^{135}	Xe^{135}	Cs^{135}	Ba^{135} Stable
	Sb^{136}	Te^{136}	$4\uparrow n$ I^{136}	Xe^{136} Stable		
			$5\uparrow n$ Te^{137}	I^{137}	Xe^{137}	Cs^{137}	Ba^{137} Stable
			Te^{138}	I^{138}	Xe^{138}	$6\uparrow n$ Cs^{138}	Ba^{138} Stable

Table IV shows the side reactions of this type numbered from 1 to 6. If it is assumed that the fine structure in the mass yield curve is due to these side reactions, then the extent of some of these reactions can be established from the experimental results so far obtained. For example, the mass spectrometric yield data for Xe^{131} and Xe^{132} are in accord with the mass yield curve, the primary yield of Sn^{133} must therefore be small and the extent of reaction (1) approximately zero. Reaction (2) on the other hand must amount to 1.3 percent of total fission since the Xe^{133} yield is found to be high by this amount. In the same way the extent of reaction (3) can be established. Since Xe^{134} yield is high by 1.9 percent (see Table V), the reaction (3) must amount to 1.9+1.3 or 3.2 percent of total fission to account for it.

Coryell, Glendenin, and Edwards[19] have proposed a theory for the distribution of charge based on equal charge displacement. The predicted primary fission yields of Sn^{133}, Sb^{134} and Te^{135} taken from their charge distribution curve are 0.6, 1.7, and 2.8 percent respectively. If we assume that the probability of emission of the extra neutron in excess of a closed shell is practically 100 percent, the extent of reactions (1), (2) and (3) are 0.6, 1.7 and 2.8 percent of total fission respectively. Similarly taking Present's theory[20] of charge distribution and again assuming 100 percent probability of emission of the extra neutron, these values are 0, 0.4 and 1.6, respectively. These results are compared in Table V.

The proposed mechanism of chain branching in the neighborhood of closed neutron shells does serve to

[16] E. W. Stanley and S. Katcoff, J. Chem. Phys. **17**, 653 (1949).
[17] G. L. Glendenin, Phys. Rev. **75**, 337 (1949).
[18] M. G. Mayer, Phys. Rev. **74**, 235 (1948).
[19] Coryell, Glendenin, and Edwards, Memo CL-LEG-I (July 25, 1946); Phys. Rev. **75**, 337 (1949).
[20] R. D. Present, Phys. Rev. **72**, 7 (1947).

TABLE V. Extent of reactions. (1–4 of Table IV).

	Reaction	Determined from mass spectrometer yield data	From Coryell Glendenin theory*	From Present's theory*
(1)	($Sn^{133} \rightarrow Sn^{132} + n$)	0	0.6	0
(2)	($Sb^{134} \rightarrow Sb^{133} + n$)	1.3	1.7	0.4
(3)	($Te^{135} \rightarrow Te^{134} + n$)	3.2	2.8	1.6
(4)	($I^{136} \rightarrow I^{135} + n$)	<3.0**	2.7	2.5

* Assuming probability of emission of extra neutron is 100 percent when nucleid formed as primary product.
** Yield determined by radio-chemical methods (Katcoff and Stanley).

explain at least qualitatively the anomalous fission yields reported above. However, the relative isotope abundance data for fission product Cs reported recently by Inghram, Hess, and Reynolds,[3] is not consistent with the above yield data if we accept the Glendenin mechanism. If their relative abundance data for Cs^{133}, Cs^{135}, and Cs^{137} are normalized to our value of 6.3 percent for the yield of the 133 chain, then the yields of these chains become 6.3, 7.8, and 6.1 percent, respectively. An abnormally high yield is, therefore, indicated for Cs^{135}. The extent of reactions (3) and (4) indicated in Table V, however, would predict a low yield for Cs^{135}.

Further yield data is necessary to verify the Glendenin explanation of fine structure in the mass yield curve. Work is in progress to determine directly by mass spectrometer methods the yields of Xe^{135}, Ba^{138}, and to re-determine the yields of Cs^{133}, Cs^{135}, Cs^{137} and Xe^{133}.

A study of the yields of the Xe and Kr isotopes in the fission of U^{233} will also be of considerable interest. With the mass yield curve shifted by 2 mass units the fine structure, if related to closed neutron shells, will appear at the same mass numbers but at different yield values. For example, the yields of the Kr isotopes will be considerably greater and any fine structure in the neighborhood of the 50-neutron shell will be more readily established. This investigation is now in progress.

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